HW-75007

PROCEEDINGS

PLUTONIUM

AS A

POWER REACTOR FUEL

DECEMBER, 1962

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HANFORD LABORATORIES

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TOPICAL MEETING

Richland, Washington -- September 13 and 14, 1962

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Co-Sponsor
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PREFACE

A National Topical Meeting of the American Nuclear Society was held in Richland, Washington, on September 13-14, 1962, in cooperation with the U. S. Atomic Energy Commission. The subject of the meeting was "Plutonium as a Power Reactor Fuel", and the program was arranged with the intent of informing the audience regarding what is known on the subject, what is not known, and what efforts are under way to get answers to the unknowns.

The program consisted of invited papers, most of which reviewed the activities at individual laboratories where substantial effort is being directed toward the development of plutonium utilization. The program was augmented by inclusion of "papers in miniature" as prepared discussion following the formal papers. The manuscripts used both for the papers and for the prepared discussion are published herein.

The proceedings have been edited by Messrs. F. G. Dawson and W. E. Roake. It is hoped that no serious errors have been made in transcribing the original papers. Grateful acknowledgment is given to Mr. J. E. Brown, Mrs. Dorothy P. Atkins, and Mrs. Ivadelle B. Daer of Hanford Laboratories for performing the task of typing and assembling the report, and to Miss Anne C. Jirus and Mrs. Mildred C. Strode, also of the Laboratories, for proofreading the typed report.

W. K. Woods
Program Chairman

PLUTONIUM AS A POWER REACTOR FUEL

U. M. Staebler
U. S. Atomic Energy Commission
Washington, D. C.

INTRODUCTION

There is no topic within the broad field of nuclear power development that has more current interest, in my opinion, than that of plutonium as a power reactor fuel. It is very appropriate and timely, therefore, that the American Nuclear Society has arranged this national topical meeting on the subject.

It is also appropriate that there is to be international participation in this meeting. Certainly the interest in plutonium as a nuclear fuel is as widespread as is the interest in nuclear power. Since the discovery of plutonium by Drs. Glenn T. Seaborg and A. C. Wahl, on February 23, 1941, there have been two other major international conferences devoted to the characteristics of plutonium as an unusual artificial element and nuclear fuel. As some of you may recall the first meeting was sponsored by the American Society for Metals and the Atomic Energy Commission in Chicago, Illinois in November, 1957. The second such meeting was held in Grenoble, France in April, 1960 under the sponsorship of the French Society of Metallurgy. The changing emphasis in subject matter of these meetings on plutonium is indicative of the increasing interest in plutonium fuels technology as time passes.

ROLE OF PLUTONIUM IN NUCLEAR POWER

There are two principal reasons for a widespread interest in the use of plutonium as a power reactor fuel.

First, one must recognize that the energy equivalent of the naturally occurring fissile isotope, U-235, in currently estimated economically available uranium resources, is not particularly large when compared with our fossil fuel reserves. A nuclear power complex sustained by U-235 alone would not represent a long term answer to our energy problem. Fortunately, the U-235 can be used to get the fission chain reaction system started and in turn can lead to the creation of fissile isotopes from the much more abundant isotope U-238. For all practical purposes, it is fission of artificially produced plutonium that makes uranium potentially capable of sustaining our future energy needs for many centuries. Of course U-233 plays a similar role in the thorium cycle. By making use of the plutonium produced in reactors, the nuclear fuel resources are many orders of magnitude greater than fossil fuel resources.

A second reason for widespread interest in use of plutonium is the economic effect of plutonium pricing on the nuclear plants operating now and expected to be operated in the immediate future. Without credit for plutonium, nuclear energy costs naturally are somewhat greater than they are with a

financial credit for plutonium. This effect is somewhat less important for natural uranium fueled systems and the seed and blanket core arrangement for water moderated and cooled reactors than for other systems. In these cases the plutonium is contained in depleted uranium of relatively little or no value and the cost of recovery may exceed its fuel value in the short term at least. For fuel systems using slightly enriched uranium fuel the economic incentive to recover slightly enriched uranium contained in the fuel material discharged from the reactor carries with it an immediate economic interest in an ability to reuse the simultaneously recovered plutonium. According to today's AEC charges for uranium and chemical processing, there is a break-even point at about 0.95% U-235 enrichment in total uranium. The economic incentive to reprocess irradiated fuel below that assay is dependent upon receipt of credit for the plutonium content of the fuel. In assigning a credit to the plutonium produced, one infers that it has a value. If nuclear power is to be commercially justified within its own rights that value must be based on the use of plutonium as a power reactor fuel. Hence, there is a real practical and immediate interest in developing the technology to permit economic fuel use of the plutonium which is produced in most power reactors.

There are many ways in which plutonium can be used as a nuclear fuel. It is inevitable that any reactor core containing U-238 will make plutonium and that some of that plutonium will be fissioned to produce energy without ever leaving the reactor core. In view of costs associated with handling and processing fuel outside of the reactor, it is naturally desirable to "burn" a large fraction of the plutonium "in situ". Some reactor designs are more favorable in this regard than others. Fuel management in any particular design can also improve this aspect of core performance. Fuel management may simply involve shuffling fuel elements from one region of a core to a region which gives the fissile plutonium atoms greater reactivity effectiveness. Depending upon the specific design and fuel management scheme, plutonium burned without recycle may produce 25 to 50 percent of the total energy produced by the reactor. The ability to burn plutonium "in situ" in thermal convertor reactors generally is limited by reactivity. In fast reactors, with their improved conversion or breeding ratio, it is likely that irradiation damage of fuel bearing materials will be limiting. There is a possibility that interchange of plutonium bearing fuel between fast and thermal reactors may offer advantages.

Whatever the reason for terminating fuel exposure, it is likely that there will be considerable amounts of plutonium remaining in the discharged fuel. Ultimately, at least, the recycling of this plutonium produced in one core for use in subsequent cores appears to be a necessity. The time at which this is necessary may, of course, be the subject of disagreement.

Recycle of plutonium in thermal-convertor reactor systems permit the production of several times the amount of energy that would otherwise be available from these systems for a given amount of uranium. Use of recycle plutonium as an alternate method of enrichment can sustain operation of some systems which require fuel enrichment by requiring only natural uranium feed. Of course, the plutonium available for recycle in such systems may not be sufficient to take full advantage of economic savings possible through enrichment.

Recycled plutonium may be used in subsequent cores of the same reactor or of a different reactor. The choice will depend upon the isotopic distribution of the plutonium produced and its relative value in reactor systems with different nuclear characteristics. There are important differences in the relative importance of various plutonium isotopes in thermal and fast reactor systems. For example, in thermal reactor systems the value of the plutonium appears to be related primarily to the content of the readily fissionable isotopes Pu-239 and Pu-241 whereas in reactors operating on a fast neutron spectrum the isotopic composition of the plutonium does not appear to be nearly as important. In fact, the "low quality" plutonium which has a significant fraction of the isotopes above Pu-239 actually appears to be a better fuel for these fast systems. This effect is due largely to the nuclear properties of Pu-241.

Also, because of the much better nuclear properties of plutonium, in general, in fast reactor systems, it appears that the value of plutonium in relationship to the price of U-235 will be of the order of twice as great in fast systems compared with thermal systems. Of course, plutonium would seek a single market value in free competition with thermal and fast reactor systems such that the plutonium value and power costs would be comparable in both systems.

We have barely scratched the surface in our consideration of plutonium recycle fuel schemes. It is quite likely that modifications in core designs of thermal reactors will improve their effectiveness of plutonium use. Certainly there are some interesting possibilities involving designs to maximize use of Pu-240 as a fertile material to provide long term reactivity stability.

There are clearly many variables which must be considered in evaluating the use of plutonium as a fuel. During the next 3 to 5 years it is likely that research and development activities including large scale critical experiments will require rapidly increasing amounts of plutonium. Totals of several hundred kilograms currently being used in such work in the United States undoubtedly will expand to several thousand kilograms during this period.

INTEREST OF THE ATOMIC ENERGY COMMISSION

The interest of the Atomic Energy Commission in plutonium as a power reactor fuel may be considered to be synonymous with that of the nuclear power industry. However, we do have certain responsibilities which give us some interest which may be considered unique. In addition to our responsibilities for fostering the development of nuclear power technology, the Atomic Energy Act currently provides that the Commission shall own all special nuclear material and shall compensate the producer in accordance with its fair value. The commission is considering legislative changes which would specify the fair value for Pu and U-233 based upon its estimated near term fuel value and permit private ownership of special nuclear material at some appropriate time in the future. It is recognized by the Commission that this transition to private ownership must be carried out over a sufficiently long period of time

that it will not introduce an abrupt economic penalty to nuclear power. On the other hand, American traditions argue that we should proceed toward private ownership of all of the materials and services required for the nuclear power industry to the extent and at rates which appear practical, considering the many factors involved.

Early establishment of a sound technical and economic basis for using plutonium as power reactor fuel is of particular importance to the Commission for a number of reasons. For example:

1. It will reduce or remove one more important uncertainty in estimating the lifetime economics of nuclear power plants which can be built today and which appear to be economically competitive in important areas of the country if certain assumptions are valid--one of these assumptions involves the value of plutonium as a fuel in recycle systems. Under these conditions utility management will have a better basis on which to decide whether or not to "go nuclear".
2. Having a sound technical and economic basis for use of plutonium as a fuel will improve the prospects that the Atomic Energy Commission can get out of the business of being a plutonium broker or custodian of a stockpile of plutonium fuel by modifying legislation to permit private ownership with appropriate provisions for a transition period. The establishment of a true market value for plutonium would bring with it the functioning of proper incentives for optimization of designs and of fuel management schemes.
3. The most important benefit that is in the offing from our nuclear power program is that it will provide a long-term solution to the problem of meeting our ever increasing needs for energy. Implicit in fulfilling this goal is the successful application of recycle of the bred fuels. Bear in mind that improving the utilization of nuclear fuels not only allows us to extract more useful energy from each unit of fuel mined, but equally important is the fact that improved utilization of nuclear fuels also allows economic use of lower grade uranium and thorium deposits which in turn vastly increases the resource base of the nuclear fuels. Thus, nuclear power systems which provide for good utilization of nuclear fuels can assure us of adequate energy availability for many centuries. Optimum use of plutonium as a nuclear power fuel will vary with time and technology. The Commission recognizes this and it will attempt to align its policies and development support to provide for both the short and long term aspects of the situation. While efforts are proceeding toward establishment of a better technical and economic basis for use of recycled plutonium as a power reactor fuel, it is recognized that the Commission must attempt to bridge the market gap. It is difficult to predict how long this will be necessary. Our development programs should provide a good technical base for recycle of plutonium in at least some large scale thermal power reactors within the next five to ten years.

STATUS AND MAJOR PROBLEM AREAS

The development of technology necessary for use of plutonium as a power reactor fuel requires extensive specialized facilities and equipment. This is due to the unusual physical and nuclear properties of the material and the special precautions required for safe handling of the materials. Thus, it has been necessary to provide a number of new facilities in order to be able to conduct a reasonable development program embracing the many facets of plutonium fuel technology. This job of providing the needed facilities has been occupying a major fraction of our effort during the past four or five years. Major new facilities devoted to plutonium are now in existence, primarily here at Hanford and at the Argonne National Laboratory. This does not mean to imply that progress has not been made in the form of research and development results. Important progress has been made but it has been mainly in the form of laboratory results on materials properties, reactor physics, and processes necessary to the fuel cycle, irradiation tests of a wide variety of relatively small samples of potential fuel materials, and analytical results on many fuel cycle variations. Great strides have been made in the development and demonstration of fuel fabrication techniques for handling plutonium bearing fuels. We are now entering the stage in the overall program where significant numbers of practical plutonium bearing fuel elements are being tested in environments typical of power reactors.

Perhaps the most significant deficiencies which exist in our knowledge of plutonium fuel cycles stems from the uncertainties in physics and extended burn-up behavior in different systems. Since the inception of the Plutonium Recycle Program at Hanford, it has been anticipated that once reliable fuel fabrication techniques had been demonstrated, the irradiation program would be expanded to include reactors other than the Materials Testing Reactor, Engineering Testing Reactor and Plutonium Recycle Test Reactor. This point has now been reached and the Experimental Boiling Water Reactor will be utilized for this purpose in a cooperative effort involving the Hanford and Argonne Laboratories. In time other experimental reactors also are expected to be used in a similar manner. The data thus generated will provide information which will permit better optimization of various thermal reactor core designs for plutonium utilization.

Work on plutonium fuels materials and fuel cycles for fast reactors is centered primarily at the Argonne National Laboratory, with very important contributions on oxide ceramic fuel mixtures being made at the Vallecitos Laboratories of the General Electric Company. Another very interesting development in advanced fuel concepts for fast reactors is the demonstration of a molten plutonium alloy as a fast reactor fuel in the Los Alamos Molten Plutonium Reactor Experiment being conducted for the AEC by the Los Alamos Scientific Laboratory.

Although the major portion of the plutonium utilization efforts in the United States are currently under AEC sponsorship, it is clear that the

ultimate application of the information being generated must be carried out by the nuclear power industry. I am encouraged by the current interest which exists throughout the industry in this respect as illustrated by the attendance at this very meeting. A wealth of knowledge is being generated concerning methods of fabricating and handling plutonium fuels, the nuclear and physical behavior of these materials during irradiation, and methods of reprocessing. Computational tools have been developed for the analysis of fuel cycles involving plutonium enrichment. I encourage the industry to make use of this information to the fullest extent possible in assessing the role of plutonium in your programs and reactor designs.

There are expanding research and development efforts on plutonium as a power reactor fuel not only in the United States but also in many other countries. I am very pleased that we will have an opportunity to hear more about these efforts as well as those in the United States during this symposium. I believe that the complimentary nature of work in different countries will become clear as the meeting progresses.

I consider it to be clearly established by now that nuclear power is to become a vitally important source of world energy in the years ahead. It is inevitable that this development will make plutonium a fuel of great importance. We are now in the very early stages of attempting to establish a technical and industrial base for using plutonium and of determining the best ways to make use of plutonium as a nuclear fuel. It is clear that our progress toward economically practical use of plutonium as a fuel will be accelerated by cooperation among the many groups actively participating in this challenging work.

THE RADIOLOGICAL PHYSICS OF PLUTONIUM

C. M. Unruh
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Richland, Washington

INTRODUCTION

Nearly 20 years of successful plutonium processing experience exist at Hanford and other AEC plutonium processing sites. This experience is available¹ to provide a sound basis for design and development engineers in the advancement of processing and handling techniques for the use of plutonium as a fuel in power reactors. By the end of this decade it is reasonable to expect the world's power reactors to generate plutonium in quantities measured in tons and possibly tens of tons per year. The use of this plutonium as a power reactor fuel is the next logical step in the short history of this element.

High exposure plutonium from power reactor fuels will generate plutonium radionuclides from mass number 238 through 241 in quantities sufficient to require a careful radiological analysis of the fuel processing operations. The radiological properties of each of these radionuclides and their daughters need to be considered. As the irradiation time of the plutonium fuel increases not only will the nuclear properties of the fuel change but also the radiological properties associated with the processing of the fuel will change.

MODES OF RADIONUCLIDE GENERATION

Table I shows the mode of formation of plutonium radionuclides and their daughters.

TABLE I

FORMATION OF PLUTONIUM RADIONUCLIDES AND DAUGHTERS

The plutonium radionuclides that result from high exposure plutonium (or high exposure uranium) fuel elements give rise to several products that affect the radiological properties of the purified fuel. For a typical plutonium fuel, neutron capture generates additional Pu^{240} , Pu^{241} , Pu^{242} , and even higher plutonium radionuclides at the expense of Pu^{239} . Pu^{238} will also be found in this irradiated fuel. In plutonium fuels Pu^{238} is formed by several processes: Pu^{241} with 13 year half-life decays by both alpha and beta disintegrations. Although only a few of the disintegrations are by alpha emission, these result in the formation of U^{237} . U^{237} decays with a 6.75 day half-life by beta emission to form Np^{237} . Neutron capture by Np^{237} yields Np^{238} which decays with a 2.1 day half-life by beta emission to form Pu^{238} . The second method of formation of Pu^{238} stems from the beta decay of Pu^{241} to form Am^{241} . Neutron addition to Am^{241} forms Am^{242} which decays with a 16 hour half-life by beta emission to form Cm^{242} . Cm^{242} decays by alpha emission with a 163 day half-life to form Pu^{238} . If U^{238} is present, a third process leading to the formation of Pu^{238} is available. The n, 2n reaction with U^{238} forms U^{237} which then, by the steps outlined for the alpha decay of Pu^{241} , leads to the formation of Pu^{248} . Each mode of Pu^{238} formation requires a neutron addition step, consequently, no Pu^{238} is formed after the fuel is removed from the reactor. The daughters of Pu^{241} , U^{237} , and Am^{241} , both continue to build up after the plutonium purification and contribute significantly to the gamma dose rate of purified plutonium.

INTERNAL DEPOSITION

In processing even the smallest quantity of plutonium, extreme care is necessary to control contamination and to prevent the deposition of plutonium in the body. The long biological life time, the high energy of the emitted alpha particles and the body's selective localization of plutonium in the bone combine to result in the low maximum permissible body burdens for plutonium radionuclides. The maximum permissible body burdens for the radionuclides of interest here are given in Table II.²

TABLE II
MAXIMUM PERMISSIBLE BODY BURDENS

<u>Radionuclide</u>	<u>μc</u>	<u>μg</u>
Pu^{238}	0.04	2.4×10^{-3}
Pu^{239}	0.04	0.65
Pu^{240}	0.04	0.18
Pu^{241}	0.9	8.2×10^{-3}
Pu^{242}	0.05	12.8
Am^{241}	0.05	1.6×10^{-2}
Np^{237}	0.06	89

The very small mass of the maximum permissible body burdens requires diligent attention to positive contamination control throughout all processing operations involving these radionuclides. Continuous monitoring of breathing air with equipment capable of sounding appropriate alarms before significant internal deposition can occur may be desirable in many facilities.

The radionuclides listed in Table II are alpha emitters with the exception of Pu^{241} which is a beta emitter. Detection of Pu^{241} contamination is difficult due to the relatively low 20 kev energy of the emitted beta particles. Control of air contamination to within the Pu^{239} maximum permissible concentration in air is not sufficient to assure that Pu^{241} is maintained below its maximum permissible concentrations in air unless it is known that the weight percent of Pu^{241} present in the material is less than about 4 percent.³ Similar considerations are necessary for the Pu^{241} maximum permissible body burden and maximum permissible concentrations in water when other plutonium radionuclides are present.

Containment for contamination control is now a universal practice in the Nuclear Industry. The use of glove boxes and hoods for the fabrication of plutonium is a common practice. Radiological engineering design of these hoods should consider such factors as air flow through the hoods, maintenance of appropriate pressure drops between the hoods and surrounding work areas, absolute filtration of discharged air, fire prevention and fire fighting within the hoods, and hood penetrations for performing manipulation within the hoods. Adequate hood design and sound operational practices have resulted in practical plutonium fabrication methods under conditions of absolute confinement.

A bioassay program is commonly established to assure internal deposition and is maintained below the applicable maximum permissible body burden. Experience in handling plutonium metal through the use of hood gloves has shown that small puncture wounds to the hands may occasionally occur and that relatively large fractions of a maximum permissible body burden may be deposited in the flesh at the site of the wound. The use of thin crystal "wound counters" adjusted to count the 17 kev X-rays from Pu^{239} have proven most helpful in estimating the quantity of Pu^{239} present in puncture wounds. Wound site depositions exceeding a few percent of the maximum permissible body burden can usually be removed without difficulty by excising the wound area.

EXTERNAL DOSE RATES

The handling of large quantities of plutonium will require appropriate shields to minimize personnel exposure during the processing of operations in addition to absolute containment for contamination control purposes. The surface dose rates from massive plutonium metal have been calculated by Roesch⁴ and others^{5,6} and are summarized in Table III.

TABLE III
GAMMA SURFACE DOSE RATES

Radionuclide	Half-Life, Years	Surface Dose Rate	
		rads/hr <40 kev	rads/hr >40 kev
Pu ²³⁸	86.4	980	2
Pu ²³⁹	2.4 x 10 ⁴	0.61	0.056
Pu ²⁴⁰	6.6 x 10 ³	14	0.62

The surface dose rate contributions from the daughters of Pu²⁴¹ are a function of the Pu²⁴¹ concentration in the mixture and the time since purification of the plutonium.

The surface dose rate contribution from Am²⁴¹ in a plutonium radionuclide mixture for times much less than 14 years is given by:

$$\text{rads/hr} = 0.2 \text{ p}_{241} t \quad (1)$$

Where:

t is time in days since purification of the plutonium.

p₂₄₁ is the fraction by weight of Pu²⁴¹ in the mixture.

The units of the constant, 0.2, are (rads/hr)/day.

The surface dose rate contribution from U²³⁷ in a plutonium radionuclide mixture for times much less than 14 years is given by:

$$\text{rads/hr} = 23 \text{ p}_{241} (1 - e^{-0.102t}) \quad (2)$$

Where:

t is the time in days since purification of the plutonium.

p₂₄₁ is the fraction by weight of Pu²⁴¹ in the mixture.

The units of the constant, 23, are rads/hr; the units of the constant, 0.102, are per day.

The contribution from each of these daughters to the surface dose rate of a plutonium radionuclide mixture is shown in Figure 1.

FIGURE 1

Surface Dose Rate Contributions from Pu^{241} Daughters

Initially the gamma dose rate from U^{237} will be the major dose rate contributor; however, the U^{237} buildup reaches equilibrium in a few weeks. The dose rate due to Am^{241} continues to increase and just equals the U^{237} dose rate at about four months (115 days) after purification. The dose rate due to Am^{241} continues to increase for many years until the parent Pu^{241} can no longer maintain the buildup of Am^{241} .

An analysis of the energy emitted by pure plutonium metal indicates that a major fraction of the dose rate originates from gamma rays and X-rays with energies below about 20 to 40 kev. As Am^{241} and U^{237} buildup in the purified plutonium, higher energy gamma rays (greater than about 40 kev) are emitted and more extensive gamma shielding may be required to maintain acceptable whole body dose rates during plutonium processing. Less than 10 percent of the X-ray and gamma radiation dose rate arises from Pu^{238} and Pu^{242} for fuel exposure not greatly exceeding 10,000 MWD/T.

The total X-ray and gamma radiation surface dose rate from massive plutonium may be calculated from the following equation:

$$\begin{aligned} \text{rads/hr} = & 980 p_{238} + 0.67 p_{239} + 14 p_{240} + 0.2 p_{241} t + \\ & 23 p_{241} (1 - e^{-0.102 t}) \end{aligned} \quad (3)$$

Where:

p_{238} is the fraction by weight of Pu^{238} ,
 p_{239} is the fraction by weight of Pu^{239} ,
 p_{240} is the fraction by weight of Pu^{240} ,
 p_{241} is the fraction by weight of Pu^{241} ,
 t is time in days since purification of the plutonium.

The surface dose rate arising from X-rays or gamma rays with energies greater than 40 kev may be calculated from the following equation:

$$\begin{aligned} \text{rads/hr} = & 2 p_{238} + 0.056 p_{239} + 0.62 p_{240} + 0.1 p_{241} t + \\ & 23 p_{241} (1 - e^{-0.102 t}) \end{aligned} \quad (4)$$

The symbols in Equation (4) are the same as those defined in Equation (3).

Appreciable neutron dose rates are also associated with massive plutonium. Spontaneous fission of the plutonium radionuclide 240 is the major source of neutrons from currently available plutonium metal. Less than 10 percent of the neutron dose arises from spontaneous fission of Pu^{238} and Pu^{242} for fuels exposure not greatly in excess of 10,000 MWD/T. The fast neutron emission from plutonium radionuclides is given in Table IV.

TABLE IV
FAST NEUTRON EMISSION RATES

<u>Material</u>	<u>Spontaneous Fission n/sec-g Plutonium</u>
Pu^{238}	3.4×10^3
Pu^{239}	2.0×10^{-2}
Pu^{240}	1.0×10^3
Pu^{241}	----
Pu^{242}	1.7×10^3

Neutron emission rates from plutonium in contact with light elements should not be neglected. The exact neutron emission rates from plutonium-light element mixtures or compounds are not well established. The emission rates calculated by M. J. Steindler⁷ for plutonium fluoride and plutonium oxide are given in Table V. The formation of these compounds during plutonium processing may lead to neutron emission rate increases of 1,000 times that observed for the pure metal.

TABLE V
FAST NEUTRON EMISSION RATES FROM PLUTONIUM
FLUORIDE AND OXIDE

<u>Compound</u>	<u>Emission Rate Due to (α, n) Reaction n/sec-g Plutonium</u>
Pu ²³⁸ F ₄	2.1 x 10 ⁶
Pu ²³⁹ F ₄	4.3 x 10 ³
Pu ²⁴⁰ F ₄	1.6 x 10 ⁴
Pu ²⁴² F ₄	1.7 x 10 ²
Pu ²³⁸ O ₂	1.4 x 10 ⁴
Pu ²³⁹ O ₂	4.5 x 10 ¹
Pu ²⁴⁰ O ₂	1.7 x 10 ²
Pu ²⁴² O ₂	2.7

There may be an advantage of lower neutron dose rates by avoiding certain light element compounds during plutonium processing. For example, the neutron emission rate from plutonium chloride is about 100 fold less than that from the fluoride. For a spherical mass of plutonium the dose rate due to fast neutrons can be estimated from the following equation:

$$\text{rems/hr} = \frac{(0.03 p_{238} + 0.009 p_{240} + 0.02 p_{242}) M}{r^2} \quad (5)$$

Where:

p₂₃₈ is the fraction by weight of Pu²³⁸,

p₂₄₀ is the fraction by weight of Pu²⁴⁰,

p₂₄₂ is the fraction by weight of Pu²⁴²,

M is the mass of plutonium in grams,

r is the distance in cm, and

the units of the constants are $\frac{(\text{rems/hr})\text{cm}^2}{\text{gram}}$.

Any increase in dose rate due to neutron multiplication has been neglected in Equation (5).

The high density of the plutonium and the relatively low energy of the emitted gamma rays result in a gamma dose rate which is largely a function of the surface area of the plutonium material being processed. This is particularly true for freshly purified material. Good housekeeping practices to prevent the distribution of even a light layer of plutonium material over the interior of a hood will greatly reduce the dose rate from the low energy, <40 kev, electromagnetic radiation. While such practices may not seem warranted when shields are provided for the processing of aged, high exposure plutonium, such practice may help to minimize hand exposures. The use of hood gloves for hood operations and maintenance can lead to hand exposure rates greatly exceeding the whole body exposure. The use of hood shielding when hood gloves are used for manipulation with the hood will not necessarily reduce or control hand exposures. Clean hoods, when practical, can alleviate this problem. Some experience is available with the use of lead loaded neoprene hood gloves and their application can reduce hand exposures.⁸ The use of remote processing equipment, whenever practical, is a better method to minimize the direct hand exposure during the fuel fabrication.

As the plutonium material ages, the gamma contribution from Am^{241} and U^{237} increases. The gamma dose rate from these daughters is a function of the mass of material present and the time since purification. The neutron dose rate is also primarily a function of the mass of material present. Neutron dose rates will not change with time and rapid processing of the purified material cannot be used to minimize neutron dose rates.

The gamma and neutron surface dose rates at a given time after plutonium purification will both increase as the fuel exposure increases. The rate of formation of each plutonium radionuclide is a function of the parameters of the reactor of interest and the composition of the initial fuel loading. In general, one may anticipate the surface dose rate arising from gamma rays with energies greater than 40 kev to increase by perhaps a factor of 2 for every few thousand megawatt days per ton of fuel exposure. Surface gamma dose rates (>40 kev) may reach a few r/hr at a week after purification following irradiation to ten thousand megawatt days per ton.

CRITICALITY

Throughout all plutonium handling operations criticality control must be considered. The critical mass of plutonium is a function of its nuclear composition, environment, and geometry. Criticality controls may be accomplished through the design and use of always critically safe equipment or by the strict application of administrative controls to processing operations. Although criticality control by equipment design is generally preferable, a judicious practice of both administrative and design criticality control is a common practice. A double contingency rule is commonly applied for criticality control. Under such conditions, the design would require more than two administrative or equipment failures to result in conditions that would lead to a criticality occurrence.

In the design of a plant, consideration should be given to personnel exposure following any possible criticality accident. Processing of plutonium materials in shielded hoods with remote operating equipment can provide protection for the operators in the event of a criticality occurrence while minimizing routine operating radiation exposures and maintaining containment of plutonium materials.

SUMMARY

The use of plutonium as a fuel for power reactors can be accomplished within a radiologically safe technology. Equipment design and processing operations should provide: (1) containment for contamination control; (2) shields for radiation exposure control; and (3) limitations on plutonium mass, concentration and solution volume for criticality control. The use of remote processing and handling techniques for plutonium materials can provide the necessary containment, shields, and criticality control is generally desirable, and is encouraged.

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THE PLUTONIUM-BASED FUEL PRODUCTION PROGRAMMES IN FRANCE

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INTRODUCTION

The construction of reactors using plutonium as fuel, and particularly RAPSODIE which will be our first fast neutron reactor, has raised the need outside the research laboratory for the building of a plant specialized in the questions of plutonium fuels. No such plant of plutonium technology existed in France until construction began at the Cadarache Centre in the Spring of 1960, and it was put into active operation in January 1962. It was designed to study the manufacture of plutonium fuel element prototypes, and also to produce small series of plutonium fuel elements for the loading of a research reactor. This is the case with our present programme which involves the manufacture of fuel for RAPSODIE and also the production of uranium-plutonium alloy slugs for the critical experiments AQUILON (study of heavy-water moderated lattices) and MARIUS (study of graphite moderated lattices).

I shall describe briefly the building, its equipment and its possibilities before going on to the manufacturing programme properly speaking.

The building is divided roughly into three sections (Figure 1). One contains 12 active plants of about 120 M² each powerfully ventilated, in which are placed most of the glove boxes for manipulations on pure plutonium, alloys or refractory compounds containing plutonium. The second part contains the general utilities of the building: mechanical plant, reception centre, stores, radiation protection centre, etc... In the third part is a hall and a number of less strongly ventilated rooms which are used for canning operations (welding, machining, polishing, cleaning of the finished elements) and for metallurgical transformation operations on alloys already canned (rolling, drawing, hammering, drawing-out, etc...).

This equipment was foreseen to allow varied types of fuel elements to be produced in limited numbers; when mass-production is necessary, the general equipment must be supplemented by installing or increasing certain work units in order to bring the output to the required level. We shall see how this principle is applied in the course of the detailed description of the processes involved in our present programme, which is divided into two broad sections:

Use of plutonium in thermal reactors,

Use of plutonium in fast reactors.

I - USE OF PLUTONIUM IN THERMAL REACTORS

Manufacture of fuel elements containing an alloy of uranium with a small proportion of plutonium.

Within the programme of the USA-EURATOM partnership to study the recycling of plutonium in thermal reactors, the participation proposed by the Commissariat à l'Energie Atomique consists of the study by substitution of uranium-plutonium fuels at low plutonium concentrations, in natural uranium heavy water lattices (AQUILON) and natural uranium graphite lattices (MARIUS), and also by oscillation in a natural uranium heavy water lattice placed in a core cavity of the swimming-pool pile MINERVE.

The problem which arises in the manufacture is to obtain rods of a homogeneous uranium-plutonium alloy containing 0.05% of plutonium in natural uranium. These rods must be canned with aluminium, forming air-tight slugs which can be assembled into 240 cm. bars (AQUILON and MARIUS experiments), then into 90 cm. bars (MINERVE experiment). The ends of the slugs must be thin enough to allow a space of less than 1 mm between two consecutive rods of alloy in a bar, including the thickness of a flux detector (a 0.2 mm thick disc of plutonium-aluminium or uranium-aluminium alloy placed in certain cases between two slugs). This distance between two rods must remain low in order that the perturbation of measurements caused by a rise in the thermal flux will be negligible.

The method established is as follows:

- the rod, 292.5 mm long and 29.2 mm in diameter, is obtained by casting the alloy.
- the canning is of aluminium A5, 0.8 mm thick.
- the plugs are of aluminium containing 0.5% magnesium, the ends being 0.3 mm thick.
- the plugs are glued with Araldite to the aluminium canning, since in fact it would be difficult to find a method of welding with such thin plugs. The technique of gluing is much less expensive and perfectly suitable, since these elements are not subjected to irradiation in a high flux nor to heat exchange. Preliminary tests have shown that slugs prepared in this way are impermeable to the helium spectrometer sweating test. The slugs thus precanned are assembled in aluminium tubes of 240 cm or 90 cm; after welding, the tube is filled with helium for a final airtightness test.

The final phases of manufacture are as follows:

1. Preparation of the Rods

The alloy is prepared at the final concentration desired by successive dilutions of a single mass of plutonium from which a mother alloy has been made, this having been divided into as many pieces as there are castings at the stage when the ingot is moulded. With the equipment we possess (Figure 2) this has involved three successive dilutions and 32 castings of 6 rods each.

FIGURE 1

Drawing of the Cadarache Plutonium Technology Plant

FIGURE 2

Vacuum Induction Furnace for the Preparation of Pu-U Alloys

A first dilution to 5.6% plutonium was made.

The ingot obtained was divided into 3 pieces and a second dilution brought each ingot to around the theoretical value of 3.6%. These 3 ingots were separated into 7 batches, and these in turn diluted in the form of slabs to a value of 2.2%. These slabs (Figure 3) are easily separated by breaking the piece which connects them to one another, and each one represents the quantity of mother alloy which must be added to about 25 kg of uranium to obtain 6 rods at a final dilution. This last casting is done in a copper ingot mould.

2. Analytical Check

It is essential that the plutonium concentration should be the same in all the rods, and therefore at each stage of the dilution the composition of the alloy was checked by two physico-chemical methods, after dissolution of samples taken at two different points of the ingot. The two methods used are coulometry and spectrophotometry. A series of glove boxes has been installed for these operations (Figure 4). The results showed in practice that the concentrations obtained did not differ, within the limits set by the precision of the analysis, from the theoretical concentrations calculated from the masses of alloy used.

3. Canning

The rough cast rods are machined on the lathe to obtain the precise dimensions necessary and the surface conditions suitable for canning (Figure 5); this operation is carried out dry in a glove box in a purified argon atmosphere.

After machining, the rod is freed of dust particles by ultrasonic washing in a tank of water; the extent of contamination is checked by rubbing the surface with a pad of cotton.

The rod is then inserted into the can (Figure 6), the ends of which have been machined to a conical shape, and Araldite is applied to a few points between the rod and the can to reinforce the slug. The ends are then capped by conical plugs previously coated with Araldite. The whole is placed in a press which holds the plugs in position on the ends of the rod while the resin hardens. After gluing, the excess metal is removed on the lathe and a perfectly cylindrical slug is obtained.

These slugs are then placed in tubes 240 cm in length (Aquilon, Marius). When the critical experiments are finished, these elements will be dismantled and the slugs recuperated will be fitted into 90 cm tubes for the Minerve experiment.

This production, already under way, is expected to be finished next November.

When this load is ready, two other sets will be prepared in order to repeat the critical experiments with 0.2% of plutonium in depleted uranium; the two sets will differ in the isotope 240 content of the plutonium used.

FIGURE 3

Pan-cakes of U - 2.2% Pu Mother Alloy

FIGURE 4
Analytical Control Chain

FIGURE 5
Pu-U Rod After Machining

FIGURE 6

Schedule for AQUILON - MARIUS - MINERVE Sheathing Operations

II - USE OF PLUTONIUM IN FAST REACTORS - STUDY AND PREPARATION OF HIGH PLUTONIUM CONTENT FUEL MANUFACTURE FOR RAPSODIE

The research began several years ago at the metallurgical laboratory of Fontenay-aux-Roses has been carried on in two directions:

- The study of a metal fuel, a uranium-plutonium-molybdenum ternary alloy and the study of a ceramic fuel, a mixed uranium and plutonium oxide.

a. Description of the Fuel Unit of RAPSODIE

Whichever of these two methods is used, the fuel appears after canning in the form of slugs, identical in outward appearance and known as needles because of their dimensions. The final mounting is thus the same in the two cases - Figure 7 and 8 - : the central needles containing the plutonium are grouped in a bundle of 37 in a tube of hexagonal cross-section, between two blanketing clusters of 7 needles containing depleted uranium. The hexagonal tube (Figure 9) is terminated at the lower end by a hollow foot which serves to place the unit in one of the holes in the base plate of the reactor. The base plate distributes the liquid sodium coolant, which is led through the hexagonal tube, passes through the clusters of needles and comes out through a piece known as a "head", which is also used for manipulations.

FIGURE 7

Setting Up of the Core Needle Bundle

FIGURE 8

Setting Into Place of the Outer Hexagonal Tube

FIGURE 9

Outer View of the RAPSODIE Subassembly

These different operations (Figure 10) are carried out on a series of machines for hammering the rivets, grinding down, mounting the needles, fitting the heads and feet before welding, and finally checking the dimensions and weights.

b. Metal Fuel

The investigations have led to the choice of a ternary alloy containing 75% uranium, 15% plutonium and 10% molybdenum. The fuel is protected by double canning (Figure 11): an outer sheath of stainless steel provides airtightness and good resistance to corrosion by liquid sodium, while an inner sheath of niobium prevents direct contact between the ternary alloy and the steel and avoids the risk of Pu - Fe eutectic. The space between the two sheaths is filled with sodium to ensure good transmission of heat to the outer can. This latter is wrapped around with a wire spiral, soldered here and there which maintains a constant space between the needles when they are assembled in groups; this enables the sodium to circulate at constant speed in the elementary channels bounded by the neighbouring needles.

1. Manufacture On a Pilot Scale

Several tens of needles are being manufactured at present, a certain number of which will be used for fuel irradiation tests.

The rod canned in niobium is obtained directly by "barometric casting" (Figure 12) in a furnace which can contain only one needle at a time (Figure 13). For this, the ends of the thin-walled (0.2 mm) niobium canning tube are pinched with a rotating hammer in such a way as to bring the external diameter to the size of the internal diameter of the original tube (Figure 22).

One of the ends is fitted with a temporary plug and the tube is placed in the furnace with the open end down. The U - Pu - Mo alloy is melted under vacuum in a crucible, and when the injection temperature is reached a mechanism raises the crucible so that the lower end of the mould dips in the liquid; a pressure of argon is then let into the furnace, the metal rises in the tube and solidifies. The rods obtained (Figure 14) are cut to the required lengths and the two ends capped by plugs which are resistance welded. One of these plugs leaves a certain free space to allow the alloy to swell on irradiation without deforming the can.

The outer canning of stainless steel is prepared apart, the lower plugs being welded by electron bombardment in batches of 12 (Figures 15 and 16). The rods and cans are brought together in a glove box in an atmosphere of very pure argon, a piece of sodium is passed into the canning, then the rod is introduced and the upper plug placed in position. The needles ready for welding are then carried in an airtight container to the welding machine and placed in the recipient (Figure 17).

FIGURE 10

Schedule of Setting Up of RAPSODIE Fuel Subassembly

3.13

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FIGURE 11
Fabrication Operations for the RAPSDIE Metallic Needle

FIGURE 12
Standard Schedule for "Barometric Casting"

FIGURE 13
Model Furnace for "Barometric Casting"

FIGURE 14

RAPSODIE Metal Rods: on the left, 5 rods as-cast with temporary plug
on the right, are two length-cut rods

FIGURE 15

A Twelve-Seat Barrel for the Welding of RAPSODIE Needle-Plugs

FIGURE 16

Detailed View of the Welding of the Lower Plug

FIGURE 17

Adjusting of Needles for the Welding of the Upper Plug

After welding, they are tested for airtightness by the helium spectrometer sweating technique. The next operation is the fixing of the helical wire (Figure 18). The needles are then centrifuged at high temperature in order to eliminate the defects in the sodium joint, and the quality of this joint is checked in an instrument using Foucault currents produced at high frequency (20 K HZ).

2. Manufacture of a Complete Load for RAPSODIE

If the metal fuel is adopted for the first core of RAPSODIE, all parts of the equipment described above except the barometric casting furnace are capable of an output sufficient to produce a core in about 6 months. They have in fact been tried out at the production rhythm for the manufacture of a core for the technological mock up of RAPSODIE, in which the fuel rods are replaced by tungsten rods of the same weight. The barometric casting furnace can prepare at best two needles per day, while there are about 2500 in a core. A furnace which could cast 20 needles at a time has been produced and will be used if metal fuel is to be manufactured.

c. Ceramic Fuel

The needle consists of a stainless steel tube containing a column of sintered oxide pellets (Figure 19), the oxide being a mixed uranium and plutonium oxide containing 25% by weight of PuO_2 . The lower plug is welded by electron bombardment and the upper one by arc welding under helium, in order to leave a helium atmosphere in the small space between the pellets and the canning. Contact between the different pellets in the column is maintained by a spring, which leaves a cavity in the upper part of the needle to collect the fission gases given off during irradiation.

1. Manufacture on a Pilot Scale

The pellets are produced by the Fontenay Laboratory, and are canned in a glove box which allows for the loading of one needle at a time. Several tens of needles will be prepared in this way before the end of 1962 and a certain number will be used for irradiation tests.

2. Manufacture of a Core for RAPSODIE

An assembly line capable of producing 800 to 1000 pellets per day has been equipped (a core represents about 80,000 pellets), occupying 3 cells in the plutonium technology plant. We have adopted the conventional process of pressing into pellets and sintering because this method is reliable and the results are well tried by several years' manufacturing experience with uranium oxide. The results obtained, at Hanford notably, by the process of cold compression by vibration were attained on tubes of rather large diameter and it is uncertain whether they would apply to thin-walled tubes of the diameter of those for RAPSODIE (5.7 mm). In the same way, the method of hot drawing under canning is difficult to perfect and requires heavy equipment which would not be easy to house in a glove box.

FIGURE 18
Winding the Helical Wire Over a RAPSODIE Needle

3. 20

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FIGURE 19

Schedule of the Setting Up of Ceramic RAPSODIE Needle

Working with the equipment installed at Cadarache, the raw materials used are ammonium diuranate for the preparation of UO_2 and plutonium oxalate for the preparation of PuO_2 .

All the furnaces were designed on the same principle (Figures 20 and 21), allowing for dismantling and repair in the glove box. They can fulfill different functions as the need arises: roasting, reduction or sintering.

The operation plan of the process is as follows:

The oxides, calcined then reduced, are fed into a ball-mill where they are mixed and crushed. The fine powder obtained, after addition of alcohol, camphor and stearine, is mixed to a thick paste which is granulated by sieving. This granulate is dried and fed at constant speed to automatic pellet-forming presses (Figure 22). The pellets obtained are freed from camphor by baking under vacuum, then sintered at 1700° in a current of hydrogen.

This programme, which was tested recently with UO_2 , will soon be in operation with plutonium.

Working Programme for 1963

Taking into account the date of divergence of RAPSODIE, the manufacture will only involve a few units of the type chosen (some hundreds of needles).

Working Programme for 1964

The manufacture of a complete core load is expected with a view to a divergence at the beginning of 1965.

FIGURE 20

Side-View of Hydrogen Atmosphere Sintering Furnace

FIGURE 21

Hydrogen Atmosphere Sintering Furnace, Loading Operation

FIGURE 22
Automatic Pelleting Press

RESEARCH ON THE FABRICATION, PROPERTIES AND
IRRADIATION BEHAVIOUR OF PLUTONIUM FUELS
FOR THE U.K. REACTOR PROGRAMME

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I. INTRODUCTION

The present U.K. nuclear power programme based on gas-cooled reactors is designed to provide an output of nearly 5,000 MW by 1968 and this implies that several tons of plutonium will be available by 1970; there is thus considerable interest in the efficient usage of this plutonium. Research at A.E.R.E. Harwell has been directed towards examining a wide variety of potential plutonium containing fuel materials for use in both fast and thermal reactors. In nearly every case, the plutonium has been heavily diluted with uranium which acts as a fertile diluent and the systems are therefore uranium-rich; thus their behaviour can be adjudged to some extent from that of the corresponding uranium system, and in such cases the fabrication behaviour can be more easily studied in the first place on the uranium system to avoid the α -active handling problem. The developed technique is then extended to the plutonium bearing material. This approach, however, is not always applicable, especially in the case of (UPu)-C as compared to U-C.

As with other laboratories, the materials studied have progressed from metals and alloys to ceramics and cermets as the need for higher temperature and longer burnup has arisen. Preliminary irradiation sorting tests on plutonium alloys had shown that within the limitations of metal fuels a range of compositions of Pu-Th, Pu-Zr and U-Pu-Mo alloys were all promising. Since the ternary system U-Pu-Mo gave the greatest freedom in choice of composition and allowed the presence of large amounts of uranium, it was considered worthy of more intensive examination than the other systems and specimens have been, or are being, irradiated over a wide range of burnups.

The ceramic materials studied have been (U, Pu)-O₂ and (U, Pu)-C. Initially the work was concentrated on the mixed oxides, but in view of the higher thermal conductivity, the higher fissile atom density and the possibility of bonding to canning materials, more attention has recently been paid to mixed carbides for fast reactor applications.

The cermet work has been concerned principally with UO₂-stainless steel and (U, Pu)O₂-stainless steel. The aim has been to produce cermets with as high a ceramic fraction as possible consistent with fabrication by conventional techniques such as rolling and to produce idealized structures from an irradiation standpoint, i. e., comparatively large spheroids of the ceramic phase evenly dispersed in a stainless steel matrix.

II. METAL FUELS - U-Mo-Pu ALLOYS

Interest in the ternary system U-Mo-Pu arose from the known good irradiation behaviour of U-Mo binary alloys coupled with the fact that work at Harwell had shown that an extensive single phase γ -U field existed in the ternary system at temperatures above 550 C. Results on a preliminary set of samples were reported at the second Geneva Conference¹ and were disappointing, excessive swelling and cracking having occurred at burnups of less than 5000 MWD/Te. However, it was thought that this was due to a poor choice of specimen compositions plus a failure to heat treat them to a homogeneous structure, taking account of constitution studies carried out since then,² a second batch of samples has therefore been irradiated. The compositions chosen were slightly lower in molybdenum than the original ones, being in the range 15-25 at. % whilst the plutonium contents ranged from 7-1/2 to 15 at. %. Only one sample of each composition has been irradiated to any particular burn-up level, but a range of burnup between 3,000 and 16,300 MWD/Te (0.4 to 1.6 % burn-up total atoms) has been covered.

The samples were in all cases machined cylinders 0.125" diameter x 0.250" high prepared from arc melted stock. An homogenizing heat treatment of 2 days at 900 C followed by oil quenching was given to the samples, a typical microstructure prior to irradiation is shown in Figure 1. A grain boundary network was often present (Figure 2) and x-ray examination suggested that this contained both oxide and carbide phases. Attempts to produce a fine grained structure by transforming below the eutectoid temperature of 550 C and then reheating for a short time in the γ -U field were unsuccessful, the prior γ -U grains being rapidly reformed.

FIGURE 1

U - 15 at. % Mo - 7-1/2 at. % Pu. Unirradiated

FIGURE 2

U - 15 at. % Mo - 7-1/2 at. % Pu. Grain Boundary Network

To date, only the more highly irradiated samples have been examined, the details of the results are given in Table I, the surface temperature being 600 C in all cases.

The S values quoted are only approximate since the swelling is not always uniform over the specimen (Figures 3 and 4). Bubbles containing fission product gases are nucleated throughout the alloys, but those at grain boundaries break away before those in the grains (Figure 5). At still higher swelling levels, bubbles within the grains also swell rapidly to produce the type of structure shown in Figure 6.

FIGURE 3

U - 15 at. % Mo - 15 at. % Pu
As-Irradiated to 8,170 MWD/Te
X12

TABLE I
IRRADIATION OF U-Mo-Pu ALLOYS

Composition, Atomic Percent				Density, G/CM ³		Percent Increase in Volume, ΔV	Burn-up MWD/Te	$S = \frac{\Delta V}{\text{Percent Burn-up}}$	Rating, Watts/G
U238	Mo	Pu	U235	Before Irradiation	After Irradiation				
66.9	15.0	7.6	10.5	17.72	11.52	53.8	7,630	51	73
"	"	"	"	17.69	9.77	81.1	14,760	55	103
69.6	15.0	14.9	0.5	17.78	10.02	77.4	8,170	70	78
"	"	"	"	17.78	9.28	91.6	14,880	59	110
57.2	25.0	7.5	10.4	17.06	13.49	26.5	7,880	25	75
"	"	"	"	17.05	11.73	45.4	14,440	29	103
59.7	24.9	15.0	0.4	16.99	15.22	11.6	8,720	10	85
"	"	"	"	17.03	10.42	63.4	16,300	41	119

X12

FIGURE 4

U - 25 at. % Mo - 15 at. % Pu As-Irradiated to 16,300 MWD/Te

X150

FIGURE 5

U - 25 at. % Mo - 15 at. % Pu As-Irradiated to 8,720 MWD/Te
Porosity at Grain Boundaries

X150

FIGURE 6

U - 15 at. % Mo - 7-1/2 at. % Pu As-Irradiated to 7, 630 MWD/Te
Widespread Porosity

It had been expected by analogy with U-Mo binary alloys that the samples with the higher molybdenum content would have the better irradiation behaviour and this seems to be true as judged on density measurements but microscopically it is less apparent. Increasing the plutonium content from 7-1/2 to 15 at. % produces no definite change in the case of the 25 at. % molybdenum alloy, but slightly worsens the behaviour of the 15 percent molybdenum sample. With the exception of the 15% plutonium 25% molybdenum alloy, the volume increase is approximately linear with increase in burnup. No figures for gas release were obtained since the samples were not individually encapsulated.

Comparisons may be made between the present results and others obtained on U-Mo-Pu, on U-Mo binary alloys and on uranium itself. Previous U.K. work on U-Mo-Pu alloys¹ showed volume increases of 99 to 113% at burnups of only 0.42 - 0.44% total atoms and temperatures of 500 C and 700 C respectively; the alloy composition was 28 at. % molybdenum - 18 at. % plutonium. A sample with a reduced plutonium content of 6.5 at. % only increased in volume by 3.3% at 700 C but the associated burnup was only 0.22% total atoms.

Recently Mustelier³ has published results on a series of U-Mo-Pu alloys containing 20 wt % plutonium. As can be seen from Figure 7 they are not very different in composition from the 15% plutonium samples of the present work. The surface temperatures employed ranged from 410 to 525 C and the burnups from 4,400 to 8,300 MWD/Te. The specimens were irradiated in the as-cast condition and volume increases of 30 - 105% were observed. It was not stated whether the neutron flux was sufficient to ensure that the specimens existed as metastable γ -uranium.⁴

FIGURE 7

Compositions of French and British Irradiation Samples U-Mo-Pu

In general, the present results appear to be better than Mustelier's and considerably better than the earlier UK ones. They are, however, very disappointing when compared to those for binary U-Mo alloys⁵ where volume increases of as little as 4% for burn-ups of 2 at. % at 600 C are observed. Recent results on uranium with controlled minor additions of iron and aluminium have given figures of only 9.7% increase in volume for a burn-up of 0.7% at centre temperatures of 550 C.⁶ Since it has been observed that the swelling of U-Mo alloys increases rapidly at temperatures above 600 C, it might be suggested that in the case of U-Mo-Pu alloys, the critical temperature for this change in behaviour is lower than for U-Mo alloys since the addition of plutonium causes a rapid lowering of the melting point.² However, if this were the explanation, Mustelier's experiments at 410 to 525 C would have been expected to give better results than the present ones carried out at 600 C. It is possible that the French alloys transformed

during the irradiation from the retained γ -uranium structure to a 2- or 3-phase structure containing ξ -uranium-plutonium and that the benefits of a lower irradiation temperature were thereby annulled.

Although the results are difficult to explain, the avoidance of severe swelling at temperatures as high as 600 C presents a difficult problem; the only promising results to date on the ternary alloys have been those of Kittell and Paine⁷ on alloys with 20 wt% plutonium - 5 wt% molybdenum, but the temperatures did not exceed 340 C.

III. CERAMIC FUELS

(a) Uranium-Plutonium Oxides

(i) Fabrication - The existence of continuous solid solubility between PuO_2 and UO_2 offers the possibility of combining the desirable irradiation behaviour of UO_2 with the use of plutonium enrichment in concentrations appropriate to both thermal and fast reactor systems. Work has thus centred around understanding the behaviour of compositions in the U-Pu-O system under conditions likely to be encountered during fabrication and irradiation and studying the conditions required to produce high density, stoichiometric $\frac{\text{O}}{\text{U+Pu}} = 2.00$ solid solution pellets.

Heating the solid solutions in argon at temperatures up to 1650 C produces materials which appear to be single phase as judged by X-ray diffraction examination. However, these oxides have $\frac{\text{O}}{\text{M}}$ ratios up to 2.10 and show separation of a U_4O_9 type phase in the uranium-rich compositions.⁸ Solid solutions which have been melted in argon remain single phase with PuO_2 concentrations up to ~30%. Since the lattice parameters of these phases do not vary with composition (from the UO_2 figure) it appears that under these conditions, these fluorite-type phases $(\text{UPu})\text{O}_{2-x}$ can tolerate considerable oxygen deficiency before splitting into $(\text{UPu})\text{O}_2$ and $\text{UO}_2\text{-Pu}_2\text{O}_3$ phases.

Pellets of $(\text{UPu})\text{O}_2$ heated in hydrogen at 1550 C show behaviour which is similar to that of pellets melted in argon but a long time is taken to reach equilibrium. A 40 mol % PuO_2 specimen heated for 3 hours at 1650 C in dry hydrogen had a lattice parameter identical with that of a stoichiometric pellet where $\text{O/M} = 2.00$. However, when the same pellet was heated in hydrogen for 12 hours, a fluorite type phase having a parameter of $5.49 \text{ \AA}$ ($\text{UO}_2\text{-Pu}_2\text{O}_3$) and amounting to about 40% of the total material present had appeared. The lattice parameters of the 10, 20, 30 and 40% PuO_2 pellets heated for 3 hours in hydrogen remained very close to those expected from a Vegards Law relationship indicating that the O/M ratios had not been reduced very far below 2.00.⁸

The phases present in U-Pu-O compacts after sintering or arc melting are largely independent of whether the starting material was the mixed separate oxides or a solid solution powder. However, the sintering behaviour of these materials is strongly affected by the nature of the feed powder. Comparing the effects of argon and hydrogen sintering atmospheres

at 1650 C, the physically mixed oxides containing a high proportion of uranium sintered to a higher density in argon whereas the plutonium-rich mixtures sintered to a higher density in hydrogen.⁹ The densities of the argon fired mixtures were close to a line connecting the densities of PuO_2 and UO_2 , whereas, the density-composition curve for hydrogen sintered pellets passed through a minimum at about 30-40% PuO_2 .

The sintering behaviour of $(\text{UPu})\text{O}_2$ solid solution prepared by co-precipitation of $\text{Pu}(\text{OH})_4$ and A. D. U. followed by calcination in hydrogen at 800 C was quite different.⁸ Here, the densities obtained in hydrogen were slightly higher than those in argon at all compositions except those very close to UO_2 . The UO_2 -rich compositions sintered to almost identical densities in hydrogen and argon, but if the argon-fired pellets were corrected for the effect of the excess oxygen, this would probably bring them below the densities obtained in hydrogen. As the plutonium concentration is increased, the sintered density obtained in hydrogen increases until a maximum density is obtained for an addition of about 10% PuO_2 . Thereafter, with further increase of PuO_2 concentration, the density could be obtained from a line joining the sintered densities of PuO_2 - UO_2 . Generally, the densities of argon sintered pellets follow the same trend, but there appears to be a minimum in the density/composition curve at $\sim 60\%$ PuO_2 .

More detailed studies of these phenomena have been made at selected compositions and Figures 8 and 9 show the sintering behaviour as a function of atmosphere for $(\text{U}_{70}\text{Pu}_{30})\text{-O}_2$ prepared from the mechanically mixed oxides and from co-precipitated $\text{Pu}(\text{OH})_4$ + A. D. U.

With the mechanically mixed oxides, the lowest densities are obtained in the most reducing sintering atmospheres and the highest densities are found when the sintering atmosphere is CO_2 containing 10% CO . Mixtures sintered in this atmosphere seemed to be less sensitive to the effects of variation of powder preparation conditions than mixtures sintered in more oxidizing or reducing atmospheres.

FIGURE 8

Density of Mechanically Mixed $(\text{U}_{70}\text{Pu}_{30})\text{-O}_2$ Sintered in Various Atmospheres at 1500 C for 4 Hours

FIGURE 9

Density of Co-Precipitated (U₇₀Pu₃₀)-O₂ Sintered
in Various Atmospheres at 1500 C for 4 Hours

The co-precipitated material sinters to the highest density for a given calcination treatment in dry hydrogen. However, more oxidizing atmospheres consisting of CO₂ containing 1-20% CO produced sintered densities which are about as high as those obtained in dry hydrogen.

Uranium-rich (UPu)O₂ solid solutions have a propensity for becoming oxygen deficient before the plutonium content is high enough to show second phase precipitation. Irradiation experiments have, therefore, been carried out to compare the behaviour of oxygen deficient and stoichiometric (U₉₀Pu₁₀)O₂.

The powders were mixed and milled in polythene mills using porcelain balls. Pellets were pressed at 30 tsi using a 5 wt% binder addition to form pellets with a green density of 6.5 g/cm³. Oxygen deficient pellets were prepared by sintering for 4 hours at 1550 C in CO₂ to promote the formation of a solid solution followed by cooling in hydrogen. The stoichiometric pellets were prepared by sintering for 4 hours at 1550 C in CO₂ and cooling in CO₂ containing 1 vol of CO per 5 vol of CO₂. This method of controlling the O/M ratio of sintered pellets by varying the CO/CO₂ ratio has been widely used in our (U, Pu)-O₂ work.¹⁰

The density of all pellets was in the range 10.5-10.6 g/cm³, but the stoichiometric pellets had an O/M ratio of 2.000 ± 0.005 while the O/M ratio for the oxygen deficient pellets was 1.990 ± 0.005 . (See the following)

(ii) Irradiation - The irradiation behaviour of mixed oxides has been studied at the 1% and 10% plutonium levels. The behaviour of $(U_{99}Pu_1)O_2$ compared with low enrichment UO_2 was described by Sayers and Worth at a recent I.A.E.A. Symposium.¹¹ In brief, five mixed oxide specimens and five enriched UO_2 specimens were irradiated under pressurized water conditions, the fissile contents being adjusted to give similar power ratings and centre temperatures in the two types of specimen.

The pellets were all 0.5-inch diameter by 0.5-inch long and each specimen comprised a 0.020-inch wall thickness stainless steel can 3.0-inch long containing five of these pellets and two MgO end spacers as 0.25-inch long annuli. These cans were contained in outer stainless steel cans and different fuel centre temperatures were achieved by varying the width of the helium filled gap between the two cans. The specimens were placed in the reactor loop symmetrically about the maximum flux position to give comparable burnups in pairs of UO_2 and $(U, Pu)O_2$ specimens. The specimen burnups were determined by calorimetry of the loop and by radiochemical analysis, fair agreement being obtained between the two methods.

The specimen data are given in Table II. The centre temperatures of these specimens were all estimated to be well within the temperature for grain growth and the extent of the grain growth area is given in this table. Generally speaking, the microstructures and gas release values show no marked differences between the UO_2 and the mixed oxide. Some interesting microstructural features were observed in these specimens, e.g., the possibility that radial cracks can act as sources of lenticular pores, the migration of which is believed to be the mechanism of columnar grain growth and central cavity formation (Figure 10).

FIGURE 10

$(U_{99}Pu_1)O_2$ Irradiated to 3000 MWD/Te, Lenticular Pores
Originating from a Radial Crack

TABLE II
IRRADIATION OF (U₉₉Pu₁)O₂ AS COMPARED TO UO₂
UO₂ 3.5% U235

Specimen No.	Max. Inner Can Temp. °C	Oxide Surface Temp. °C	Oxide Central Temp. °C	$\int_{T_s}^{T_o} k d\theta$	Heat Rating watts/gm	Burn-up MWD/Te Oxide	Burn-up Fissions/cc x 10 ¹⁹	Grain Growth Dia. Pellet Dia.	% Gas Release	
									Fiss. Kr	Fiss. Ke
6109	485	960	1890	25.8	26.6	2578	7.66	0.561	9.34	11.5
6110	695	890	1920	28.3	29.0	2850	8.47	0.586	Not sampled	
6111	480	770	1860	30.1	31.1	3040	9.02	0.513	5.06	6.31
6112	560	880	1960	30.5	32.4	3175	9.43	0.616	27.2	34.6
6113	490*	1250	2360	31	33.5	3240	9.62	0.818	39.5	49.1
1% PuO ₂ in UO ₂ 2% U235										
16114	585	1170	2170	28.1	30.3	3010	8.94	0.761	50	55.9
16115	585	1070	2090	28.1	29.4	2970	8.82	0.71	20.5	24.1
16116	545	800	1800	27.5	27.9	2820	8.37	0.473	8.3	10.3
16117	630	1000	1960	26.6	26.7	2690	7.99	0.641	10	12.2
16118	500	970	1830	24.0	24.5	2435	7.23	0.561	12.3	10.6

N.B. The ratio $\frac{\text{GRAIN GROWTH DIAMETER}}{\text{PELLET DIAMETER}}$ is calculated from the pellet diameter observed after irradiation and not on the original dimension.

Oxide temperatures have been calculated from these ratios using Fig. 3 in CRFD-835.

This temperature has been calculated on the measured diameter of the swollen can.

Electron microscope replica studies on these specimens also yielded interesting information. The 0.002-inch Bex film replicas of polished and etched regions of columnar grain growth in both the irradiated UO_2 and $(\text{U}, \text{Pu})\text{O}_2$ specimens were obtained and examined in an unshielded Philips model EM 100 electron microscope. Figure 11 shows a typical picture in which bubble formation in the grains is apparent. These bubbles are about $400 \text{ \AA}$ diameter and tend to be clustered into distinct groups in the mixed oxide, possibly due to nucleation along dislocation lines. These fine bubbles may be the forerunners of the large bubbles seen in oxide fuels at much higher burnups. These studies are continuing using replication of fracture surfaces so that the bubbles may be seen more clearly.

FIGURE 11

$(\text{U}_{90}\text{Pu}_{10})\text{O}_2$ Irradiated to 2820 MWD/Te. Fission Gas Bubbles
in Equiaxed Grain Growth Region X10,000

More recently, the irradiation behaviour of $(\text{U}_{90}\text{Pu}_{10})\text{O}_2$ was compared with that of 10% enriched UO_2 . The method of preparation of the mixed oxide was as described previously. The UO_2 was prepared by the standard method, viz: ball milling powder obtained from calcination of A.D.U., adding "Cranco" binder, cold pressing, removal of binder and sintering at 1650 C in a hydrogen atmosphere. The pellets measured 0.25-inch diameter by 0.25-inch long and were canned in 0.020-inch thick 20 Ni/20 Cr stainless steel tubes. Each specimen comprised a 2-inch long miniature fuel element containing eight pellets, the cans being sealed by arc welding under an atmosphere of helium. The estimated fuel/can gas gap was 0.001 inch. Six specimens were made-- three containing UO_2 and three mixed oxide, of which two had O/U + Pu ratios of 2.00 and one a ratio of 1/99. All pellets were carefully characterized before canning, external dimensions were measured to $\pm 2.5 \mu\text{m}$ on a Sigma comparator, real and apparent densities were measured to determine open and closed porosity, and finally, the end face of each pellet was polished for metallographic examination. After canning, the external dimensions of the cans were measured at a number of points to $\pm 2.5 \mu\text{m}$.

For irradiation, the samples were sealed in groups of three in sodium filled stainless steel capsules measuring 4.5-inch long by 2.5-inch diameter. Insulated thermocouples passed through seals in the capsule lid to each specimen and cobalt wire flux monitors were attached to each specimen. The object of the sodium filling was to assist in the removal of the fission heat. Surrounding each capsule was a helium filled gas gap of controlled size, which determined the specimen temperatures, and a water cooled heat sink. The maximum undisturbed thermal flux in the reactor was 4.5×10^{13} and the rig remained in the reactor for 232 operating days, resulting in the ratings and burnups shown in Table III. In all cases the can surface temperature was approximately 600 C.

Postirradiation examination included:

- (i) Measurement of can external dimensions.
- (ii) Can punctured and gases removed to an evacuated glass sample tube for mass spectrometric analysis.
- (iii) Can sectioned at the interface between the central two pellets and the exposed face prepared for microscopic examination.

The results of (i) and (ii) are listed in Table III, the fission gas release being recorded as percentage release of the total xenon and krypton formed in the fuel. In general, the results show negligible increases in can dimensions and fairly low fission gas releases. In particular, we may compare the UO_2 and $(\text{U}, \text{Pu})\text{O}_2$ specimens taken to 12,700 MWD/Te burnup with similar centre temperatures. The gas release values are 0.37% and 0.144% which are in good agreement, bearing in mind the experimental errors involved; the level of release is also reasonable for the estimated centre temperature of 1200 C. The gas release for the two highest burnups is higher than expected; this

TABLE III
IRRADIATION OF (U₉₀Pu₁₀)O₂ AND OF (U₉₀Pu₁₀)C

Specimen Composition	Rating		Estimated centre temperature °C	Estimated burn-up MWD/te	Estimated volume of Xe produced cm ³	Total gas collected cm ³	Vol. of Xe cm ³	% of gas release	Can diameters	
	watts/cm	watts/gm							Before irrad. (inches)	After irrad. (inches)
UO ₂	119	36	880	9,000	2.93	3.62x10 ⁻²	3.62x10 ⁻⁴	0.01*	0.2930	0.2931
UO ₂	169	51	1015	12,700	4.16	8.07x10 ⁻²	1.54x10 ⁻²	0.37	0.2917	0.2918
UO ₂	247	75	1275	18,700	5.53	0.409	0.31	5.53	0.2925	0.2926
(U ₉₀ Pu ₁₀)O ₂ ¹	168	51	985	12,700	4.72	2.7x10 ⁻²	6.8x10 ⁻³	0.144	0.2925	0.2927
(U ₉₀ Pu ₁₀)O ₂ ²	168	51	985	12,700	4.72	0.25	Not available due to gas contamination		Not available	
(U ₉₀ Pu ₁₀)O ₂ ²	197	60	1050	14,900	5.39	0.457	0.278	5.2	0.2923	0.2932
(U ₉₀ Pu ₁₀)C	49.5	76.5	~ 700	19,000	1.28	4.64x10 ⁻³	3.4x10 ⁻⁴	2.65x10 ⁻²	0.1437	0.1438
(U ₉₀ Pu ₁₀)C	59.5	91.5	~ 800	22,800	1.13	1.0x10 ⁻²	1.32x10 ⁻⁵	1.1x10 ⁻³	0.1437	0.1437

* Inaccurate due to argon leak from cell during gas sampling

$$^1 \frac{O}{U+Pu} = 1.990$$

$$^2 \frac{O}{U+Pu} = 2.000$$

could be due to underestimate of centre temperatures but no grain growth was evident in the microstructures. The microstructures in either case show no significant changes on irradiation which, again, is to be expected from the fact that the centre temperatures did not reach the level for grain growth (Figures 12A and 12B). Under these conditions, therefore, there is no noticeable difference in the irradiation behaviour of UO_2 and $(\text{U}_{90}\text{Pu}_{10})\text{O}_2$.

(b) Uranium-Plutonium Carbides

(i) Fabrication - Specimens of solid solutions of (U Pu)C have been fabricated both by powder routes and by the skull arc melting method. In both cases, particular attention has been paid to the problem of compositional control. Plutonium contents of up to 30% have been studied, but most work has been done on the 15% composition.

The arc melting technique was initially established using uranium carbide; the 3-inch diameter crucible held a maximum of 700 g of carbide and skulls were prepared by melting together uranium and graphite, initially filling only one-third of the crucible, remelting this up to 6 times, turning the charge over between each melt and then progressively making further additions to the skull. When full; about 25% of the contents could be poured.

The graphite electrodes were 1-1/4 inch diameter with a 1/4- to 1/2-inch diameter axial hole in the end--the function of the latter was to focus the arc and thus stabilize the melting conditions but in practice, if spattering occurred, the arc tended to emanate from the spatter rather than the hole. It was thought that this could be due to the lower work function of the spatter, and the inside of the hole was therefore coated with a material of low work function; caesium chloride being chosen. This technique proved very successful and the coating survived several melts. In addition, carbon transfer from the electrode to the melt was minimized.

Casting was carried out by the Durville method using massive copper moulds, graphite moulds having resulted in more piping in the product and a worse surface finish. It was found that the casting rarely had the desired carbon content whether using uranium plus graphite or premelted carbide as the charge. The results for 45 melts intended to be stoichiometric are shown in Figure 13 and the distribution is markedly different from the normal Gaussian error curve for a random distribution; the absence of the stoichiometric composition suggested some type of instability at this composition. The liquid appears to pick up either carbon or uranium from the skull and to have an equilibrium composition of either 4.6 wt% or 5.0 to 5.1 wt%. Reference to the published diagram¹² suggests that this would correspond to a change of only about 20 C in the liquidus temperature. A test experiment was performed in which a sample was poured, a small section of the casting analysed, and the rest replaced and remelted. This process was repeated 6 times and the carbon content of all the castings was between 4.98 and 5.08 wt% (Figure 14). The composition, therefore, changed during the course of the first melt to the equilibrium value and was thereafter essentially constant. Since the time at maximum power was approximately 1-1/2 minutes,

FIGURE 12A

$(U_{90}Pu_{10})O_2$ Unirradiated X275

FIGURE 12B

$(U_{90}Pu_{10})O_2$ Irradiated to 12,700 MWD/Te with Estimated
Centre Temperature 1000 C X300

FIGURE 13

Change of Composition on Skull Arc Melting
of Stoichiometric UC Charges

FIGURE 14

Variation in Carbon Content of Uranium Carbide
During a Series of Remelts

it was decided to reduce this to 5 seconds. The test was carried out on $(U_{85}Pu_{15})C$ and was highly successful in the case of prealloyed charges; nine consecutive specimens having compositions between 4.76 and 4.85 wt% carbon with standard deviation from the mean of 4.80 wt% being only 0.03 wt%. These preliminary results show clearly that the skull technique can be used for the melting and casting of mixed carbides.

Experiments on the sintering behaviour of uranium carbide and uranium carbide-plutonium carbide have been carried out on powders which have been prepared by ball-milling arc-melted buttons. Work in other laboratories^{13, 14} has indicated that very high temperatures (>1800 C) are required to produce satisfactory densification of nuclear carbides unless sintering aids are employed. However, it has recently been found that if the carbide particles are sufficiently small and are prevented from reacting with water vapour and oxygen, compacts prepared by cold-pressing such powders will sinter to $>95\%$ theoretical density at temperatures as low as 1550 C.

The behaviour of pellets during sintering is strongly affected by particle size which should be in the range 1 to 3 microns and is also determined by the care taken in the previous handling of the powders and by the purity of the sintering atmosphere. Variations in the carbon content and sintering time produce little effect on the final density as illustrated in Table IV. With reasonable care in powder handling and during pellet preparation, the carbon content of pellets will not alter by more than 0.05 wt% from that of the initial powder.

TABLE IV
THE EFFECT OF SOME SINTERING VARIABLES ON THE DENSITY
OF U-C AND $(U_{85}Pu_{15})$ -C PELLETS

Pellet	C content (wt %)	Sintering time (hrs)	Sintering temperature (°C)	Bulk density (g/cm ³)
(U Pu)-C	4.69	4	1585	13.11
(U Pu)-C	4.89	4	1585	13.11
(U Pu)-C	5.25	4	1550	13.12
U-C	4.70	5	1550	12.99
U-C	4.70	12	1550	12.94

Typical microstructures of sintered high density pellets are shown in Figures 15 and 16. Figure 15 is of a $(U_{85}Pu_{15})-C$ pellet containing 4.66 wt% carbon in the electropolished condition and shows the single-phase solid solution. Figure 16 is of a $(U_{85}Pu_{15})-C$ pellet containing 4.85 wt% carbon chemically etched in nitric acid/acetic acid/water; small white particles of sesquicarbide, reasonably uniformly dispersed in the monocarbide matrix, are visible.

FIGURE 15

Sintered $(U_{85}Pu_{15})-C$ Containing 4.66 wt% C.
Apparently Single Phase X500

FIGURE 16

Sintered $(U_{85}Pu_{15})-C$ Containing 4.85 wt% C. Small Particles of
Sesqui-Carbide Uniformly Dispersed in the Monocarbide Matrix. X750

Both of these photographs indicate a discrepancy between the actual microstructure and that expected from the determined carbon content bearing in mind that the stoichiometric monocarbide contains 4.8 wt% carbon. Present indications are that oxygen picked up during powder handling dissolves in the monocarbide lattice during sintering resulting in a shift of the microstructure-composition relationship to higher carbon contents.

(ii) Thermal Conductivity - Determinations of the thermal conductivity of mixed uranium-plutonium carbides have been carried out in a linear heat flow apparatus similar to that described by Schroder.¹⁵ The results are quoted in Table V and are compared with those obtained for uranium monocarbide in the same apparatus. For the arc-cast U-C specimens the variation in thermal conductivity for a carbon content range of 0.6 wt% is only 0.006 cal/sec/°C/cm/cm². The microstructure showed only UC and uranium in the carbon deficient samples, whereas the carbon-rich sample showed UC with UC₂ plates. The appearance of the hot-pressed sample was very different since this consisted of UC with a U₂C₃ network present at all grain boundaries.

TABLE V
THERMAL CONDUCTIVITIES OF UC AND (U₈₅Pu₁₅)-C CYLINDERS

Fabrication Method	Density (g/cm ³)	Carbon content (wt. %)	Thermal conductivity (uncorrected for porosity) (Cals/sec/°C/cm/cm ²)
<u>Uranium Carbide</u>			
Arc Cast	13.87	4.40	0.052
" "	13.64	3.77	0.052
" "	13.40	5.06	0.058
Hot Pressed	13.08	4.91	0.041
<u>Uranium Plutonium Carbide</u>			
Arc Cast	13.67	4.55	0.031
" "	13.55	4.71	0.036
" "	13.24	4.84	0.041
Hot Pressed	13.21	4.92	0.030
Arc Cast	-	5.03	0.040
Hot pressed	12.90	~ 5.2	0.030
" "	13.04	5.41	0.030

With the mixed carbides, the thermal conductivities of the arc-cast stoichiometric and carbon-rich specimens are in good agreement (0.041 and 0.040 cal/sec/°C/cm/cm²) but the decrease with increasing metal concentration is somewhat steeper than with U-C. All carbon rich specimens contained the monocarbide together with a sesquicarbide phase. The distribution of the phase varied greatly, and was present as a continuous grain boundary phase in all arc cast specimens, whereas, in the hot pressed specimens, the distribution depended on the pressing temperature. The pressing temperature used for the U-C specimen was sufficiently high that the sesquicarbide was distributed along the grain boundaries as in the arc cast (UPu)-C, whereas the microstructure of the hot-pressed mixed carbides was similar to that of the sintered pellets illustrated in Figure 16.

The thermal conductivity values of the carbon-rich carbides are more affected by the method of preparation than they are by carbon content. For both U-C and (U-Pu)-C, the thermal conductivities of hot pressed samples, (not corrected for density), are about 70% of those of the corresponding arc-cast material. For both methods of manufacture, the addition of 15% PuC to UC has reduced the thermal conductivity to about 70% of its original value.

(iii) Irradiation - Two samples of (U₉₀Pu₁₀)C were irradiated in the same rig which was used for the irradiation of the 10% enriched oxide specimens. The details of the irradiation were as described for the oxide with the exception that the fuel diameter was 0.1 inch, but the can wall thickness remained at 0.020 inch. The pre- and postirradiation examination procedures were the same as for the oxides and the results are recorded in Table III.

These two specimens were arc-cast by the drop casting technique into 1/8-inch diameter copper moulds and contained approximately 4.6 wt% carbon. This was evident in the microstructure (Figure 17A) which showed light etching areas of metallic phase. After irradiation, the microstructures showed no cracking except for some areas of intergranular cracking in one specimen which has been interpreted as inter-granular corrosion due to an excessively high moisture content in the hot cell atmosphere (Figure 17B). The central temperatures estimated for these specimens are quite low--about 700-800 C--and in consequence the fission gas release values are also low. In fact, they are somewhat lower than those calculated from a recoil release mechanism. In a more recent experiment, further mixed carbide specimens have been irradiated together with samples of sintered UC and liquid-phase sintered UC + UFe₂. Burn-up levels were about 15,000 MWD/Te and, since these specimens were also of 0.1-inch diameter, the centre temperatures were about 700-800 C.

FIGURE 17A

(U₉₀Pu₁₀)-C Containing 4.6 wt% C. Unirradiated.
Free Metal Present X1250

FIGURE 17B

(U₉₀Pu₁₀)-C Containing 4.6 wt% C. Irradiated to 20,500 MWD/Te.
Grain Boundary Cracking Probably Due to Atmospheric Corro-
sion During Postirradiation Examination. X100

IV. CERMET FUELS

(a) $(U, Pu)O_2$ - Stainless Steel Cermets

(i) Fabrication - The benefits to be derived from the use of dispersion type fuel elements have been described by Weber and Hirsch¹⁶ but it is only recently that techniques have been developed in the case of UO_2 -stainless steel cermets for producing structures approximating the idealized one, namely a homogeneous dispersion of large UO_2 spheroids in a stainless steel matrix. Recently, there has been considerable interest in the UK in the development of $(U, Pu)O_2$ -stainless steel cermets containing ceramic fractions as high as 50 v/o. With such high oxide contents, the nonhomogeneous dispersion of angular ceramic particles that had proved satisfactory in cermets containing less than 30 v/o ceramic is not acceptable and attention has therefore been focussed on the fabrication of UO_2 and $(U, Pu)O_2$ spheroids.

A technique for producing UO_2 spheres has been developed by Williams.¹⁷ This involves pressing milled UO_2 into green pellets, breaking the pellets down by pushing through sieves to give the desired size fraction and then vibrating the crude granules in a deep track impregnated with a suitable abrasive material. Subsequent sintering in cracked ammonia gave densities greater than 95% theoretical and by recycling the fines produced at the sieving stage, 90% of the original oxide could be converted into excellent spheroids.

Attempts to produce $(U-Pu)O_2$ spheroids by using the same technique on mixed oxides were not immediately successful. An investigation of the variables involved showed that the problem was due principally to the presence of moisture in the PuO_2 . After drying the PuO_2 for 16 hours at 70 C, some 70% of the crude granules spheroidized within 15 hours on the vibrator. The addition of 0.4 wt% of stearic acid as a binder increased the yield to 95%.

To promote the formation of a solid solution of PuO_2 in UO_2 , the green spheroids were sintered in a 90 v/o CO_2 10 v/o CO mixture for 4 hours at 1500 C. Since this is liable to give $(U, Pu)O_{2+x}$, the product was then heat treated for 4 hours at 850 C in a stream of damp hydrogen to obtain the stoichiometric $(U, Pu)O_2$ composition. The size range required was 350-500 microns diameter and it was found that the granules increased in size during spheroidization, but this was compensated for by shrinkage during sintering (Figure 18). Thus, a batch of granules in the range 350-500 microns would typically give a product with 80% in that range, 10% < 350 microns and 10% slightly greater than 500 microns. X-ray examination of the spheroids showed a single face-centered cubic pattern, the cell size corresponding closely to that for the appropriate solid solution.

FIGURE 18

Spheroids of $(U_{70}Pu_{30})O_2$ X17

The fabrication of cermets by tumbling the prepared spheroids with stainless steel powder and pressing into compacts gave a very non-uniform dispersion. A coating technique similar to that developed by Cunningham¹⁸ was therefore employed to form a thick layer of stainless steel powder around each of the mixed oxide spheroids. The yield of coated spheroids was normally about 50% with the remainder being as aggregates which were sieved out and the oxide spheroids recovered by washing with solvent.

The coated spheroids were then compacted as required and a typical compact for the fabrication of a plate by the roll bonding technique is shown in Figure 19. At present, UO_2 -stainless steel cermets have been successfully fabricated into plates by the picture frame method using reductions of 60-70% at 1100 C for the 50 v/o ceramic material¹⁹ (Figure 20). Lower reductions gave unsatisfactory bonding and lower density in the core material whilst higher reductions lead to fragmentation and stringering of the UO_2 particles. The work is currently being extended to $(U, Pu)O_2$ cermets.

FIGURE 19

50 v/o $(U_{70}Pu_{30})O_2$ - Stainless Steel Cermet. Sintered Compact. X5

FIGURE 20

50 v/o UO_2 - Stainless Steel Cermet. As Roll Bonded. X23.5

FIGURE 21

50 v/o ($U_{70}Pu_{30}$) O_2 - Stainless Steel Cermet. As Hot Pressed.
Carbide Network in the Steel Matrix. X145

(ii) Mechanical Properties - Tensile test pieces were machined from the 50 v/o UO_2 stainless steel plates prepared as described above. Both transverse and longitudinal sections were prepared and in all cases, the gauge length was pure cermet and the stainless steel from the cladding only existed in the region beyond the shoulders.

The samples were tested in a Hounsfield tensometer in the as-rolled condition and also after annealing at 1050 C for 15 minutes and air cooling. A series of runs were carried out at different temperatures and values obtained for the U. T. S. and the elongation (Table VI); the latter was determined by carefully fitting together the fractured pieces after testing.

TABLE VI

MECHANICAL PROPERTIES OF 50 v/o UO₂ - STAINLESS STEEL CERMET

	Temperature °C	Transverse		Longitudinal	
		Hot rolled	Annealed	Hot rolled	Annealed
Elongation %	700	7.8	7.9	6.5	6.3, 7.7
U.T.S. Tons/sq. in.		3.59	3.79	4.62	4.06 , 4.87
Elongation %	600	6.3	6.7	8.1	9.5
U.T.S. Tons/sq. in.		4.9	3.88	5.20	5.34
Elongation %	400	5.7	11.4	5.7	11.7, 8.4
U.T.S. Tons/sq. in.		5.02	4.51	5.93	6.02, 6.22
Elongation %	200	4.2	-	5.5	6.0, 7.6
U.T.S. Tons/sq. in.		5.64	-	6.56	5.74, 6.93
Elongation %	Ambient	5.8	6.3	5.5, 5.4	8.9
U.T.S. Tons/sq. in.		8.11	5.74	8.80, 8.13	7.72

As expected, there is scatter in the results, but in general the properties appear to be superior in the longitudinal as compared to the transverse direction and annealing raises the ductility in both cases. The superior properties in the longitudinal direction are probably associated with the fact that the UO₂ deforms during rolling into ellipsoids, thus altering the effective UO₂ content of the X-sections for the different directions. The high ductility does not appear to be over-estimated by the fitting together of the fractured pieces since the cases where the fitting is worst have not given the highest ductility figures. No definite signs of necking are visible in the microstructures (Figures 22 and 23).

FIGURE 22

50 v/o UO_2 - Stainless Steel Cermet. Fractured Test Pieces. X1.3

FIGURE 23

50 v/o UO_2 - Stainless Steel Cermet. Fracture Surfaces. X1.3

(iii) Irradiation - Preliminary to the irradiation of 30, 40, and 50 v/o (U, Pu)O₂ - stainless steel cermet, postirradiation annealing experiments have been carried out on cermet specimens previously irradiated at a surface temperature of about 300 C in a pressurized water loop. These cermets consisted of a dispersion of irregularly shaped UO₂ particles of 50-200 microns diameter dispersed in Swedish iron, the UO₂ content being 18.7 wt% (14.2 v/o). The specimens were of two forms; rolled plates clad in aluminium-killed mild steel, and rods clad in 18/8/Ti stainless steel cans. Burnups were in the range 10 to 14% of uranium atoms with centre temperatures of about 370 C in the plates and 700 C in the rods. As irradiated, the specimens showed negligible dimensional changes. The initial density (93% theoretical) remained unchanged after irradiation.

Small specimens were cut from one of the rods and annealed in vacuo for 48 hours at a series of temperatures in the range 900 C to 1200 C. After annealing the densities were measured, surfaces were prepared for microscopic examination and plastic replicas were taken for electron microscopy. Briefly, the results showed a steady decrease in density from 900 to 1200 C although the extent of this decrease was only 4% at the highest temperature. The microstructures showed bubble formation in the UO₂ before heating and this was more extensive after annealing. Bubbles were observed in a region about 5 microns thick surrounding each oxide particle after annealing at the highest temperature. These bubbles were formed by the gaseous fission products which had recoiled into this region of the matrix. The electron micrographs showed evidence of fine bubble formation even before annealing. After annealing, fine bubbles were visible at the outer fringes of the recoil zones but those nearer the fissile particles had become too large to replicate (Figures 24 and 25).

FIGURE 24

14 v/o UO₂ - Fe Cermet.
As-Irradiated to 14% b.u.
of Heavy Atoms. Fine
Bubble Formation in
Recoil Damaged Matrix.
X5,500

FIGURE 25

14 v/o UO_2 - Fe Cermet
Annealed 48 Hours
1200 C After Irradiation.
Bubble Growth Has
Occurred in the
Damaged Matrix.
X5000

Associated with these effects was a change in the micro-hardness of the damaged regions of the matrix, i. e. , a rapid decrease in hardness occurring between 900 and 1050 C (Figure 26). It is difficult to proportionate the amount of swelling between the ceramic phase and the matrix, but undoubtedly the combined swelling will be serious for reasonable burnups at irradiation temperatures above 900 C although no catastrophic failures have been currently observed. In a cermet power reactor fuel, the matrix would probably be stainless steel rather than iron which would be expected to improve the resistance of the matrix to swelling, but also, the time at temperature will be very long compared to the 48 hours of the present experiments.

FIGURE 26

Variation of Micro-Hardness with Annealing Temperature
for Irradiated 14 v/o UO_2 - Fe Cermet

Experiments are now in progress to examine the irradiation behaviour of the types of cermet material whose fabrication behaviour has been discussed above. The specimens comprise 30, 40 and 50 v/o oxide in stainless steel in the form of plates or pellets. The plate specimens contain enriched spherical UO_2 particles in the size range 250 to 700 microns, the UO_2 being dispersed homogeneously throughout the matrix. The plate dimensions are 2.0-inch long, 0.4-inch wide, 0.50-inch thick (fuel) + 0.020-inch cladding. The pellets were prepared by the hot pressing route and contain $(\text{U}_{70}\text{Pu}_{30})\text{O}_2$ particles in the size range 250-350 microns and are 0.20-inch diameter with a 0.020-inch thick can.

These cermet specimens are being irradiated to burnups in the range 3 to 13% heavy atoms at ratings of 70 to 220 watts/g to cover a wide range of possible fast reactor operating conditions.

(b) (U, Pu)C-Fe Cermets

(i) Constitution Studies - Briggs et al.²⁰ established that a pseudo binary exists between UC and iron or chromium. Interest was thereby raised in the possibility of producing UC - Fe or UC - Cr cermets by melting and casting. The iron cermets were considered first in view of the volatility of chromium.

The possibility of enriching with plutonium for fast reactor application led to a cursory examination of the Pu-Fe-C system. The samples were prepared by arc melting and examined by metallographic, thermal analysis and X-ray diffraction techniques. The results showed that because of the instability of PuC as compared to UC, iron was capable of removing plutonium with the formation of PuFe_2 so that the tie lines Pu_2C_3 - PuFe_2 and PuC - PuFe_2 exist. A study by Marples²¹ of the uranium-rich portion of the UC-PuC-Fe system, however, has shown that up to 50% of the uranium can be replaced by plutonium without altering the as-cast microstructure; on raising the plutonium content to 70%, $(\text{U, Pu})\text{Fe}_2$ appeared in the microstructure. The variation in solidus temperature has also been studied in portions of both the U-Fe-C and U-Pu-Fe-C systems. A value of 1116 C was obtained for the UC-Fe eutectic as against the 1160 C reported by Briggs, whilst eutectics were also observed between UFeC_2 -Fe and UFe_2 -UC at 1160 and 1201 C respectively. A ternary eutectic is thought to occur between UC-Fe- UFe_2 at approximately 24 a/o U - 70 a/o Fe - 5 a/o C and 1037 C. Replacement of 30% of the uranium by plutonium on the UC-Fe tie line only reduced the eutectic temperature from 1116 to 1100 C.

(ii) Fabrication - The skull melting technique developed for UC has been extended to UC-Fe and by using the longer melting time at full power, i. e., 1-1/2 minutes, it was found that the carbon content tended to increase. The final values lay in a very narrow band about 4.5 wt% for a 10 wt% iron cermet, whilst for cermets containing 2-1/2 and 5 wt% iron, the behaviour was less regular. Since stoichiometric UC - 10 wt% Fe contains 4.32 wt% carbon, comparison with the histogram for UC melts (Figure 13) shows that the increase corresponds to the most probable value for the UC case. As expected from the data of Briggs,²⁰ the product being hyper-stoichiometric contains the ternary compound UFeC_2 in addition to primary UC and the UC-Fe eutectic (Figure 27).

FIGURE 27

Skull Arc-Melted Carbon - Rich UC - 10 wt% Fe X400

(iii) Bonding Studies - Operating ceramic fuels with high centre temperatures could lead to appreciable fission product gas release: this, in turn, may result in failure of the can. The centre temperature may be lowered either by reducing the fuel diameter or by improving the heat transfer between fuel and can. The former is usually not desirable whilst the latter method may be effected in a variety of ways, e.g., using a liquid metal bond or alternatively bonding fuel to can with a solid metal. The solid metal bond is intrinsically the most stable of the possibilities and since carbides are in general more readily wetted by metals than oxides, it is especially attractive for (UPu)C fuels. On the grounds of well established technology, high temperature strength, availability, and cost, stainless steels are a natural first choice for the canning material. The formation of eutectics between UC-Fe and UC-Cr suggests that a eutectic bond might be produced between a (U, Pu)C fuel and a stainless steel can. In view of the comparatively large differences in thermal expansion of the carbide and the steel, cracking of the bond may be a problem.

Preliminary experiments at Harwell have shown that such bonds can be formed by a variety of techniques but it is too early to say whether the thickness of the bond can be satisfactorily controlled. The thermal cycling behaviour of such bonds will also require investigation.

V. DISCUSSION

Since an integrated national nuclear power system requires the development of various types of reactors and the maintaining of a suitable balance between them, general development on plutonium bearing fuels has been carried out on a broad front with a view to determining the limits of behaviour. For a variety of reasons, the fast reactor appears to be the most suitable type for plutonium utilization and it is for this reason that the present investigations have been concerned with metallic, ceramic and cermet fuels with appreciable plutonium contents. For the metal fuels, a burnup of 2 a/o at a temperature of 600 C or above would be necessary to make the fuel attractive. The present results show that the ternary U-Mo-Pu alloys are unlikely to satisfy these conditions.

Extensive phase equilibria and sintering studies have been carried out in the $\text{PuO}_2\text{-UO}_2$ system and the conditions necessary for obtaining high density, solid solutions have been established.

The use of oxide fuels to achieve high burnup at high temperatures has been widely demonstrated for UO_2 and the present results indicate that the mixed oxide $(\text{U, Pu})\text{O}_2$ behaves similarly both at the 1% and 10% PuO_2 levels.

The use of a dispersion of $(\text{U, Pu})\text{O}_2$ in stainless steel to overcome the problem of the low thermal conductivity of $(\text{U, Pu})\text{O}_2$ has received considerable attention. The work of Lambert reported here has indicated that for cermets with low ceramic contents, substantial burnups can be achieved without dimensional instability at temperatures up to 800-850 C and that the nucleation and growth of bubbles in the matrix is an important factor in determining the stability above these temperatures. However, there is an economic incentive to raise the ceramic content and it is for this reason that the range of composition 30 - 50 v/o ceramic is being studied. At the 50 v/o level, it is essential that there shall be sufficient ductility to minimize the formation of cracks by thermal stresses and the results suggest that an idealized dispersion will be reasonably ductile prior to irradiation.

For carbide fuels, the higher thermal conductivity of UC as compared to UO_2 has been held up as a major advantage. Present work indicates that though the conductivity is lowered by dissolving PuC in UC, the value is still high compared to that of UO_2 . By paying attention to particle size and atmosphere conditions, direct sintering of the mixed carbides to high density at reasonable temperatures has been shown to be feasible. The irradiation behaviour is essentially unchanged at the 2 a/o burnup level by the incorporation of 10% PuC in UC and as already pointed out, the fissile atom density is higher for $(\text{U, Pu})\text{C}$ than for $(\text{U, Pu})\text{O}_2$. These features make $(\text{U, Pu})\text{C}$ an economically attractive fast reactor fuel. Furthermore, since the carbide is wetted by many metals it is possible to consider bonding the fuel to can with consequent reductions in the centre temperature and avoidance of grain growth and associated fission product gas release. The "bond" could either take the form of a liquid metal between the fuel and the can or a direct solid metal bond.

Addition of certain metals such as iron to (U, Pu)C using a melting and casting route leads to a structure consisting of discrete carbide particles in a fine eutectic matrix of the metal and carbide. By analogy with the cemented carbides, this would be expected to confer improved mechanical properties compared with the pure carbide. In view of the relatively high thermal conductivity of UC-rich (U, Pu)C, little improvement is to be anticipated in this property, but it is possible that bonding of the fuel to the can will be made easier. An objection to these possible fuels is the decrease in fissile atom density with increasing metal content--thus UC-10 w/o Fe has a uranium density of 10.9 g/cm^3 as compared to 13.0 g/cm^3 for UC, though this still compares favourably with 9.7 g/cm^3 for UO_2 .

The use of plutonium as low enrichment for thermal reactors has been considered, particularly for oxide fuels. The results of both fabrication and irradiation experience at the 1% PuO_2 level show that no new limiting phenomena are to be expected. The possibility of using plutonium in a high temperature reactor has also received some consideration. An all ceramic fuel element would be required, possibly of the "Dragon" type using a mixture of carbides dispersed in a graphite matrix or alternatively as an oxide ceramic. In the latter case, PuO_2 could be dispersed together with UO_2 in an oxide matrix of BeO or Al_2O_3 , or a combination of oxides to form a glass. Phase equilibria and sintering studies are being carried out with a view to proceeding to an irradiation investigation once the basic features have been established. A further possibility would be to use a perovskite compound²² of plutonium in which the plutonium is more firmly bound as the dispersed phase. The physical and mechanical properties of these compounds are being investigated and appear favourable for this type of application.

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HANFORD PROGRAMS FOR ADVANCED PLUTONIUM FUEL DESIGNS AND FABRICATION METHODS

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I. INTRODUCTION

A goal of the power reactor fuels program of Hanford Laboratories is to develop simple, inexpensively fabricated, plutonium-bearing metallic and ceramic fuel elements for safe operation at high specific power levels for long periods of time. Achievement of this goal requires integrated fuel design, fabrication, testing operations, and supporting fundamental studies. The fuel development effort also must be extremely sensitive to interactions with other parts of the overall fuel cycle, such as reactor design and operation, chemical processing, physics, and fuel cycle analysis. For example, fuel element designs are being modified as data accumulate which indicate that the core melting behavior of a high performance fuel element can be controlled by changing reactor start-up procedures.¹

The following discussion is not intended to be a comprehensive survey of all possible plutonium fuel element designs and fabrication methods. It simply points up typical promising developments in each area of fuel technology which, when adequately simplified and integrated, will reduce plutonium-bearing fuel costs to a surprisingly low level. It also points up fertile ground for inventions and development which will hasten the arrival of that day.

II. FABRICATION STUDIES

Throughout the nuclear industry one finds high density fuel materials used to obtain a relatively high thermal conductivity, fission gas retention, nuclear reactivity, mechanical stability, fail-safe behavior, and freedom from gross relocation during irradiation. Typical densification methods are compared in Figure 1 and Table I. If particles are used instead of conventional sintered shapes, high densities are achievable by several methods.

FIGURE 1

Ceramic Fuels Fabrication Techniques

TABLE I**DENSIFICATION OF CERAMIC MATERIALS**

Process	Density (UO ₂ , % Theoretical)	Status of Technology	Versatility			
			Alternate Materials	Pu Fuels	Recycle Fuel	Enrichment Or Batch Size
Fusion	> 99	Commercially Available	Medium	Low	Low	Low
Electrodeposition	> 99	Development Scale	Medium	Medium	High	Low
High Energy Impact	> 99	Development Scale	High	High	High	High
Compaction and Sintering	< 98	Commercially Available	Medium	Low	Low	High

In one commercial process, approximately 1600 pounds of UO_2 can be fused in each lot. The cost of fused UO_2 has decreased markedly and the quality is much improved, but further process improvements are essential to minimize voids, contamination by nitrogen, and chemical disproportionation. Although the fusion process is increasingly promising for naturally enriched UO_2 , applications to plutonium enriched fuels (and to alternate fuel materials such as uranium nitride and uranium sulfide) will require techniques less apt to introduce toxicity, criticality, and other problems. An alternate route for uranium ceramic fuel elements enriched with plutonium is to fuse only the UO_2 and to add finely divided, nonsintered PuO_2 during final compaction in the fuel assembly.

High energy impaction is the most versatile process available for either preparing or densifying fuel particles.² With the equipment illustrated in Figure 2, oxides, nitrides, carbides, and cermets are compacted to essentially theoretical density at pressures to one-half million psi in high impact strength dies. Pressures to 880,000 psi or more have been achieved by modified "Bridgman Anvil" techniques. The equipment and process are being modified to accommodate plutonium-bearing, and partially decontaminated, recycled fuels. Notable advantages of this process also include high throughput and no fuel hold-up as in the fusion process.

Economics of the process appear attractive.

Electrodeposited UO_2 crystals of high density and purity have been prepared on a pilot plant scale.³ The greatest potential of this process may be in the preparation of partially decontaminated, high density particles from irradiated ceramic fuels. Studies of the properties, fabrication behavior, and irradiation performance of this and other partially decontaminated fuels will continue.

Development of the desired cladding materials and geometries, at acceptable costs, is a continuing, major problem. Crucial tests of new fuel element concepts can be delayed or jeopardized by lack of cladding of adequate quality, particularly when large diameter, novel shapes, or thin walls are required.

A remarkably versatile method for fabricating fuel cladding is the recently developed, high voltage electron beam welding process. The cladding shown in Figure 3 was fabricated in small quantities at a reasonable cost; fabrication by extrusion would have been prohibitively expensive. The inner coolant tubes of this assembly were welded together by passing the electron beam tangentially between them. They were then attached to the inner surface of the outer tube by welding from the outside (the electron beam actually penetrated the outer tube to complete the internal weld). The electron beam welding process also provides the best method for bonding straight or spiral spacing members. PRTR cluster and tubular fuel elements with such ribs are being evaluated. One-piece 19-rod clusters for PRTR tests are also being successfully fabricated by this process.

FIGURE 2

FIGURE 3

An extremely versatile process for assembling the fuel core and cladding materials is high energy vibrational compaction (Figure 4). This process plays an increasingly important role in Hanford fuel development programs because of its amenability to (1) many fuel and cladding materials, (2) decreased cladding tolerances, (3) thin-wall cladding, (4) novel fuel geometries and fuel enrichment schemes, (5) varied production rates, (6) automation, and (7) remote handling operations.⁴

FIGURE 4

It is now difficult to standardize reactor fuel element designs sufficiently to permit low cost, mass production. Versatility of the vibrational compaction process means that many fuel designs can be evaluated at acceptable costs without the advantages of mass production. Future Hanford fuel element fabrication studies will be concerned less with further development of the process than with using it to evaluate quickly and inexpensively the most promising of a growing number of possible fuel concepts or combinations of fuel geometries, materials, designs, fabrication methods and irradiation operations.

For example, the number of fabrication steps involving plutonium-bearing materials can be minimized by using heterogeneously enriched cores (Figure 5). These are easily fabricated by vibrational compaction. Typically a very small amount of plutonium is distributed throughout a relatively large amount of matrix material during vibrational compaction. To avoid segregation of the enriched phase, incremental loading equipment and techniques are used.⁵ These are examples of fabrication steps for which automation studies are urgently needed!

FIGURE 5

The most versatile closure technique under development for rod-cluster fuel elements is Magnetic Force Resistance Butt Welding⁶ (Figure 6). Zircaloy, beryllium, and many other candidate cladding materials have been welded in air, inert gas, or other atmosphere by this process. It will be particularly valuable for plutonium-bearing fuels when combined with a "dry cleaning" technique for cleaning components just prior to completing the weld (Figure 6). The equipment and closure techniques will be remotized and automated.

FIGURE 6

Another approach to the plutonium-bearing fuel element closure problem involves welding fuel element end plugs into the filled cladding tubes before the fuel material is compacted by swaging, or other similar processes. This eliminates the usual post-swaging cutting of the fuel rod to length, counter-boring, decontaminating, etching, cleaning, and welding.

Remote fabrication techniques are being developed to permit (1) fabrication of fuel elements containing gamma-active material such as partially decontaminated, irradiated UO_2 , and (2) disassembly of irradiated fuel elements, followed by rearrangement, replacement, or re-enrichment of components to extend the fuel element lifetime or to permit re-use of the elements in other reactors. Remote fabrication operations of the first type are illustrated in Figure 7.

Recycle Fuel Fabrication

FIGURE 7

In developing these processes high priority is assigned: (1) pre-assembly and nondestructive testing of as many fuel element components as possible; (2) keeping the gamma-active material separate from other core and cladding materials until the last possible moment; (3) automating the fabrication equipment and minimizing maintenance costs by utilizing modular concepts; and (4) developing equipment which requires location of only a few essential parts in the hot cell. For example, the amplifiers and transducers employed in vibrational compaction are being located outside the cell, with energy transmitted to the fuel element by a resonant structural beam or other connecting member.

The first remotely fabricated PRTR fuel element is expected to be of a tubular design. Prior to insertion into the hot cell, all the fuel element components will be fabricated, tested, cleaned, assembled, and inspected. Manipulations inside the hot cell will be limited, essentially, to (1) incremental loading of UO_2 and PuO_2 fuel, (2) welding of the top end closures and fixtures, and (3) finally inspecting the fuel element assemblies.

To modify elements for reuse in the same or different reactors, methods must be developed not only for disassembly and reassembly of components, but also for opening and closing the fuel compartments--possibly underwater. Underwater disassembly and some fabrication work with PRTR fuel elements have been performed successfully, although adequate closure methods and other fabrication techniques, as well as nondestructive tests, still require development.

III. FUEL DESIGNS

Two geometries of UO_2 and $\text{UO}_2\text{-PuO}_2$ PRTR fuel elements are illustrated in Figure 8. The fuels were compacted by cold swaging, hot swaging, and vibrational compaction.

These fuel element designs are relatively simple, using powder fuel, nonsegmented components, and, for the 19-rod clusters, nonbonded spacing members. Their performance has been generally satisfactory. One of the UO_2 elements with a pinhole prior to irradiation was operated for more than three months without evidence of waterlogging or progressive deterioration.

The possibility of fretting corrosion has strongly affected all fuel element designs involving Zircaloy cladding. Although catastrophically rapid corrosion of Zircaloy components has been demonstrated under certain combinations of coolant conditions, vibration, and contact between components, it has been avoided to a remarkable degree in the PRTR. Encouraging evidence that fuel designs can be developed which will eliminate or adequately reduce the fretting corrosion which has been observed at a number of sites was obtained by increasing the width of ribs on fuel element end fittings. The nested tubular fuel element design also is promising, in that no fretting corrosion has been detected after irradiation of a full-size element for six months. Design modifications undoubtedly will be required as these fuel elements are pushed to higher performance levels, but it is quite possible that these relatively simple elements can be operated at four times the heat generation rate of elements in most existing water-cooled power reactors. As increased

performance demands are made of such fuel elements, eventual failures must be anticipated--in many cases, because of "man-made limits" such as defects or impurities in the cladding and core materials, rather than "fundamental limits" such as brittleness, low thermal conductivity, core melting, or fission gas release.

PRTR FUEL ELEMENTS

FABRICATION METHODS

1. VIBRATIONAL COMPACTION
2. COLD SWAGING
3. HOT SWAGING

1. VIBRATIONAL COMPACTION
2. SINTERED COMPACTS

FIGURE 8

The design of more advanced fuel elements is increasingly dependent upon realistic concepts of the properties and behavior of ceramic fuel materials at elevated temperatures. For example, at operating temperatures for which most ceramic fuel elements are now being designed uranium dioxide becomes quite ductile (Figure 9), and the thermal conductivity becomes much higher than previously assumed (Figure 10).

FIGURE 9

THERMAL CONDUCTIVITY OF URANIUM DIOXIDE

FIGURE 10

The most fundamental design problem for future plutonium fuel designs remains: "How do you usefully remove the heat?" The general nature of this problem is indicated by a graphic illustration of the approximate color temperatures existing in several present and future fuel elements (Figure 11). The outer edge of the lightest region in each of the elements is at a temperature of approximately 1200 C. Until the fuel melts, at least, the temperature steadily increases as the center of the fuel rod is approached. The temperatures and heat removal problems encountered in designing and developing these and other elements can be illustrated by considering heat transfer in several consecutive steps: (1) through the core material, (2) across the fuel-cladding interface, (3) through the cladding, (4) across the cladding-coolant interface, and, finally, (5) useful recovery of the heat from the bulk coolant. The most recent quantum jump in understanding these processes has occurred in the first step, heat transfer through the fuel.

FIGURE 11

At moderately elevated temperatures, little is known of the nature of ceramic fuel cores at any given time during irradiation, because of the bonding, irradiation damage, grain growth, cracking and healing of cracks, disproportionation, redistribution of fission products, and other changes which continuously occur (Figures 12 and 13). Behavior at temperatures near and beyond the melting point is even less understood.

Zircaloy-2 Cladding, 0.505 inch ID $\times$ 0.025 inch Wall
 Diametral Clearance, 0.004 $^{+0.000}_{-0.001}$ inch at Assembly
 Polished Linde A and Linde B Abrasive, Trace Chromic Acid
 Etched, 60 ml 95 percent H_2SO_4 + 40 ml 30 percent H_2O_2
 Large Black Areas Indicate UO_2 Fragments Lost During Grinding and Polishing

Exposure 900 MWD/ T_U (2.4×10^{19} fissions/ cm^3)
 Maximum Heat Rating, 31 kw/ft
 Maximum Surface Heat Flux, 230 watts/ cm^2 (730,000 Btu/(hr)(ft 2)
 Calculated UO_2 Surface Temperature, 450 C
 Fission Gas Release, 41 percent of Xenon and Krypton
 Pellet Density, 10.3 g/ cm^3 at Assembly; Sintered in 1700 C Hydrogen
 Ground to 32 μm . Surface Finish

Irradiated, Sintered UO_2 Pellet Cross Section, Test GEH-14-189

FIGURE 12

Fission Product Distribution in Ceramic Fuels

FIGURE 13

It is often assumed that the design of fuel element cores would be a relatively simple matter if accurate thermal conductivity data were available. Actually, "thermal conductivity" has a rather limited meaning because, at the temperatures usually encountered in advanced design fuel elements, much of the heat transfer is by convection and radiation.⁷ This is apparent in the thermal conductivity data now available for single crystal UO_2 .⁸ For liquid or vapor phase ceramic fuels, the heat transfer situation is more complex, because of the chemical disproportionation or other reactions. Use of the term "ceramic" becomes questionable! One way of exploring ceramic fuels at such temperatures is to simulate in-reactor thermal conditions by internal, self-resistance heating of sections of externally-cooled fuel rods. These experiments are being conducted. At the same time, data are being obtained directly from the reactor, as indicated by the fuel cross-section shown in Figure 14. This originally sintered UO_2 fuel element is assumed to have operated with approximately one-half of the core diameter molten, because the tungsten marker wires apparently fell to the bottom of the fuel container when the reactor came to full power. The fuel design picture is complex, even without introducing extremely high temperatures and drastic irradiation alterations. This is particularly true of powder-type elements because of continually changing particle and bulk densities, sintering, and low temperature bonding encountered during irradiation. Near the center of a high performance fuel element, however, characteristics of the core become very similar for both powder or sintered-type materials (Figure 15). Bonding between ceramic fuel particles occurs at relatively low temperatures during hot swaging or impact forming, and, most interestingly, during irradiation. When a 19-rod PRTR fuel assembly containing six thermocoupled rods was irradiated, improved heat transfer at relatively low (300 - 400 C) fuel centerline temperatures was revealed. Hopefully, fuel elements can be designed and fabricated in which this effect is increased by choosing, for example, a more "in-reactor sinterable" material. Fundamental studies of the nature of this low temperature bonding, and methods of accelerating it, are in progress.

The oriented, single crystals which develop during irradiation have important fuel design implications. For example, as indicated above, they reveal relatively high thermal conductivities, particularly at elevated temperatures. From this, Horn and deHalas¹ have postulated that a profound decrease in the maximum temperature of a fuel element occurs early during irradiation, with initially molten cores solidifying as the irradiation continues. The significance of this is simply that heat can be removed from a fuel element much more rapidly than has always been assumed--provided you demand enough of the element to begin with!

Another important effect of the sintering and extensive growth of oriented single crystals during high temperature fuel irradiation is that the design of fuel elements containing plutonium or thorium is simplified because the effects of nonhomogeneous enrichment can be minimized (Figure 16).

FIGURE 14

FIGURE 15

Heterogeneously Enriched Fuels

FIGURE 16

Designs must also take into account effects of cracking, voids, impurities, and central cavities. Cracking, like the presence of initial interfaces between fuel particles, reduces heat transfer rates. Photomosaics of irradiated fuel element cross-sections reveal fewer cracks than anticipated--even after two hundred or more thermal cycles of the fuel elements. This and other evidence indicate that cracks undoubtedly heal during irradiation, thereby improving the heat transfer.

Effects of the many types of voids developed during high temperature irradiation have been investigated intensively only recently. Future fuel element design studies must take them into account.

Effects of impurities on the thermal conductivity of partially decontaminated fuels have not been determined. Preliminary irradiation test data indicate no effects on the extent of sintering or cracking.

The central cavity formed in a high performance fuel element during irradiation has the practical effect of relieving the hottest portion of the core by simply displacing the offending material. This may have disadvantages, however, because of the decreased internal support of "collapsible clad" elements. Cavities formed in fuel geometries such as tubes might radically decrease the mechanical stability of the fuel element. "Inverted-cluster" type fuel elements, in which the ceramic fuel is cooled by relatively small diameter, internally located tubes may be easier to design, because the cavities formed during irradiation cause the nonuniform fuel cross-section to adjust itself to receive relatively uniform cooling. This has been demonstrated by irradiation tests.

Additives to UO_2 have often been investigated with the hope of obtaining improved thermal conductivity, resistance to thermal shock, and other desirable properties. In general, the migration of carefully "positioned" additives at temperatures where improvement is needed precludes any pronounced, lasting effect of the type desired. This is illustrated by the relocation of fission products during irradiation, as well as by the gross fuel relocation observed in most high performance ceramic fuels.⁹ Strangely, but happily, plutonium oxide seems to be the one additive that is an exception. More rapid sintering during irradiation, lower fission gas release rates, and improved resistance to thermal shock appear to result from small additions of PuO_2 to UO_2 . Otherwise, additives seem to provide little hope for obtaining radically improved fuel element designs and performance in thermal reactors.

Alternate fuel materials to the oxides of uranium, thorium, and plutonium are being investigated, particularly at Argonne National Laboratory¹⁰ and Battelle Memorial Institute.¹¹ Hanford test elements containing a number of these alternate materials, or mixtures of them, await irradiation. With the exception of those for uranium carbide, few data are now available from which fuel elements using these materials can be designed and the irradiation performance predicted. Fabrication behavior of these alternate materials in processes such as high energy impact also are being investigated. Supporting fundamental studies, such as melting point determinations on irradiated materials, also are in progress.

Penetration of fuel cladding by the coolant makes the already complicated fuel element core behavior even more complex. The chain of events affecting fuel performance includes (1) decreased thermal conductivity between core particles, (2) oxidation of at least part of the core materials, (3) some dissolution or peptization of $(U, Pu)O_2$ or ThO_2 , (4) possibly wash-out or waterlogging, and (5) hydriding of cladding materials such as Zircaloy, with release of oxygen to further attack the core material. Data concerning continued operation of defected elements are perhaps even more urgently needed than the data from elements under normal operating conditions. It is unrealistic to assume economic fuel cycles that demand fabrication of fuel elements which are invariably perfect. Even if this were technically and economically feasible, the reactor designer and operator sooner or later would reasonably demand freedom to "push the element to the limit"--preferably with continued, safe operation for at least some time after the limit was passed.

We believe the most important discontinuity in the heat flow pattern is that at the core-cladding interface. The most recently published study of interfacial heat transfer coefficients is by Ross and Stoute.¹² It summarizes in detail the effects of composition and pressure of filler gas, interface pressure and pressure cycling, movement of contact points, and surface roughness on the interfacial heat transfer coefficient between cylindrical UO_2 pellets and Zircaloy-2. But there are two paths--through direct contact and through gas films--and the relative importance of each is different in vibrationally compacted or in swaged fuel rods (Figure 17). The relative importances are not known. Certainly, in the case of vibrationally compacted fuel rods, heat transfer through the interface gas might be expected to be relatively more important than in the case of pellet rods in which thermal expansion applies interfacial pressure between two machined surfaces. Swaging, however, creates a relatively large interface area and much mechanical keying of fuel to the cladding; therefore, contact conductance would be assumed to be particularly important.

Improved heat transfer between the fuel cladding and the reactor coolant could be very important because of the direct effect upon the fuel-cladding interface temperature. That temperature is limiting with respect to solid state chemical reactions between the fuel element core and cladding materials. This may be particularly important if the core is heterogeneously enriched, since core-cladding compatibility might be exceeded at relatively low average heat transfer rates. The degree to which fuel homogenization during irradiation alleviates such problems is being investigated.

Bonding materials such as lead increase the rate at which heat can be removed through the interface, but these bonding layers have many undesirable features. The bond must be complete and subject to reliable nondestructive testing, or it is literally worse than nothing. The bonding layer also involves disadvantages with respect to reactivity, expansion, and incompatibility of materials.

FIGURE 17

Oak Ridge out-of-reactor experiments¹³ revealed that heat transfer rates to 55 million Btu/hr-ft² are achievable. An imaginative approach is needed to adapt those results to nuclear fuel element designs. Heat generation rates of several million Btu/hr-ft² may be achievable in reactors like the PRTR by using vortex cooling, interrupted heat generation along lengths of fuel elements, surface roughening of the cladding, and coolant additives.

Major design problems become quickly evident when the temperature drop through the cladding material is considered as a function of surface heat flux (Figure 18). Again, the importance of material compatibility at the core-cladding interface cannot be over-rated! For example, it appears that the Zr-UO₂ reaction which occurs at approximately 1100 F would be encountered at a heat flux of approximately 1,500,000 Btu/hr-ft², in a fuel element having 0.030-inch thick Zircaloy-2 cladding and a coolant temperature of 500 F. Interface temperatures at a heat flux of 5,000,000 Btu/hr-ft² become wondrous to contemplate (Figure 19). New or composite cladding materials may be required.

FIGURE 18

FIGURE 19

Mechanical properties of cladding such as Zircaloy also will be deleteriously affected by the high temperatures. Although alternate metallic, ceramic, or cermet cladding materials may be more adequate, this conclusion indicates that collapsible, or non-self-supporting cladding must be studied more extensively. Thin-walled, collapsible cladding also would decrease the temperature drop, of course, as well as the reactivity loss associated with the cladding.

Another approach is to use spacing members, or other mechanical supports, to improve the structural properties of the cladding (Figure 20). Out-of-reactor autoclave tests have revealed that the ribs surrounding the 2.3-inch diameter fuel rod have a significant effect in increasing the external pressure which can be tolerated without collapse of the cladding.

Advanced Fuel Concepts

FIGURE 20

An exploratory test element of the large diameter fuel rod (Figure 20) was irradiated for two principal, typical reasons. First, simplification of fuel assemblies to a degree indicated would represent an economic breakthrough for fuel fabrication, particularly with recycled fuels. (The 2.3-inch OD rod contains as much UO_2 as nineteen 0.56-inch OD PRTR fuel rods of the same length.) Second, and more important at present, the irradiation testing of an element as untried or extreme as this inevitably points up the research and development studies most urgently needed to overcome performance limits. Conceivably, the above fuel element could be integral with the reactor coolant tube, as shown in Figure 20, thereby eliminating possible problems such as fretting corrosion. This concept would be particularly interesting if the core material

provided "Phoenix" fuel characteristics; i. e., uniform, long-life reactivity due to physics characteristics of higher plutonium isotopes during irradiation. The technology developed with this and similar fuel element-coolant tube combinations is applicable to "inverted-cluster" type fuel elements. These are of particular interest to superheat or supercritical reactors, in which plutonium fuel would have important advantages over uranium if steel alloys are used as cladding.^{1 4} The ultimate answer, for certain types of materials and reactors, may be to eliminate the cladding. Irradiation for an extended period of a fuel element having porous steel cladding is now being scheduled.

IV. CONCLUSIONS

In summary, relatively simple, versatile processes developed for plutonium fuel and cladding fabrication, and for fuel element assembly operations must be further simplified, adapted to remote operation, and integrated to obtain an overall process which can be conducted remotely in the smallest possible hot cell. The number of operations and the amount of material handled within the hot cell must be minimized by methods such as preassembly of components, heterogeneous enrichment, and automation of the fabrication equipment. Similar processes must be developed for reconstruction or re-enrichment of fuel elements for reuse in the same or different reactors.

Fuel design studies will continue to profit from (1) a rapidly increasing body of fundamental information relating out-of-reactor and in-reactor properties and behavior of UO_2 , PuO_2 , and other ceramic fuel materials, and (2) data concerning the in-reactor behavior, physics characteristics, and chemical processing of fuel assemblies.

Although presently designed fuel elements probably are capable of much higher performance than is now demanded, they will undoubtedly eventually encounter difficulties with compatibility of the core and cladding materials, hydriding of the cladding, and fretting corrosion.

In general, the greatest progress in studies of advanced plutonium fuel element design and fabrication processes will come from (1) improved understanding of the performances of carefully chosen and imaginatively tested materials and fuel assemblies; (2) rapid communication and correlation of fundamental and engineering data so that time and effort are not wasted on meaningless debate; and, (3) leapfrogging the supposed limits of materials at extreme conditions by encouraging the inventions and discoveries which provide technical and economic breakthroughs.

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PLUTONIUM FUEL EXPERIENCE AT HANFORD

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Research and development work on plutonium reactor fuels was begun at Hanford in mid-1956 on authorization of the Plutonium Recycle Program.¹ Additional developmental fuel fabrications in support of other Atomic Energy Commission programs have also been performed. Most notable of these are a plutonium loading for the operation of the Materials Testing Reactor² and plutonium target rods for the synthesis of trans-plutonium elements, mainly californium. An average of about 1000 individual fuel components, rods or plates, containing plutonium have been fabricated each year since 1958, Figure 1, and all but the Plutonium Recycle Critical Facility fuel have been irradiated. This has involved about 180 kilograms of plutonium to date.

FIGURE 1

Plutonium Fuel Element Irradiations

The relative effort expended on various fuel developments is illustrated in Figure 2. Initial developments were largely concerned with aluminum-plutonium alloys clad in either aluminum or Zircaloy.³ In all cases, aluminum claddings were metallurgically bonded to the aluminum-plutonium cores by either hot rolling or hot coextrusion⁴ techniques. Bonds have been formed with Zircaloy cladding by injection casting of molten aluminum-plutonium alloys directly into the cladding tubes.⁵ Although the bonding layer is brittle, the major portion of the bond remains intact when subjected to out-of-reactor or in-reactor thermal cycling. This is quite surprising since these materials have markedly different coefficients of thermal expansion.

FIGURE 2

Plutonium Fuel Development Work at Hanford

Unbonded, Zircaloy-clad, 19-rod spike fuel bundles have been evaluated in the Plutonium Recycle Test Reactor.⁶ These unsegmented, 8-foot long rods were fabricated⁷ by hot extrusion of the one-half inch diameter, corrosion resistant aluminum-plutonium alloy cores. The cores were slipped into the Zircaloy cladding tube with a 0.004- to 0.008-inch diametral gap which is partially closed by differential thermal expansion at operating temperature. Spacing of rods in the bundle is achieved by means of spiral wire wraps on individual fuel rods. Loosening of wire wraps has been observed after irradiation due to shortening of the fuel rods as a result of intermittent in-reactor bonding between the core and the cladding. Data indicate these wires retighten at operating temperatures. In general, irradiation performance of aluminum-plutonium alloys has been excellent⁸ up to powers as high as 42 kw/ft/(1000 watts/cc) and coolant temperatures to 310 C.

Other spike fuel concepts offering fabrication economies are also under development: ceramic spikes containing MgO-PuO₂ and ZrO₂-PuO₂ and Zircaloy-clad plutonium-zirconium alloy extended surface fuel fabricated by roll cladding.

Development work on plutonium ceramics was initiated in 1959 and fuels fabricated by pelleting of UO₂-PuO₂⁹ and cold swage compaction and vibrational compaction of UO₂-PuO₂ particle mixtures have been tested. Emphasis has been placed on the particle compaction technique and approximately 23 19-rod fuel bundles of this type are under irradiation in the PRTR. Replacement of the remaining portion of the PRTR spike loading with these uniformly enriched UO₂-PuO₂ elements is scheduled over the next few months.

Arc-fused natural UO₂ particles are mechanically enriched with fine PuO₂ particles during the fuel rod loading step. Distribution is obtained by simultaneously loading small increments of PuO₂, mixed with the fine UO₂, along with increments of the coarse UO₂ fraction¹⁰ (Figure 3). Compaction to fuel densities of about 88 percent of theoretical is then accomplished by cold swaging¹¹ or by means of high-energy vibration.¹² Initial fuel made by this technique showed homogeneity of enrichment on a large scale, but layering of enrichment increments was observed.

FIGURE 3

Incremental Loading of UO₂-PuO₂ Particle Fuels

A prototypical seven-rod cluster, shown in Figure 4, containing UO_2 -0.5 wt % PuO_2 was irradiated in an ETR high temperature loop.¹³ This 42-inch long cluster was fabricated by incrementally loading UO_2 and PuO_2 into three-quarter-inch diameter by 0.035-inch wall Zircaloy tubing, welding in swageable end caps and cold swaging the assembled rods to 0.565-inch diameter and final length. The assembled cluster was irradiated at a maximum power of 14 kw/ft in 250 C coolant at 2000 psi to a maximum exposure of 800 MWD/T. Postirradiation examination included autoradiography on glass of the uncut rods as shown in Figure 5. Plutonium segregation or layering is clearly shown and the periodicity correlates with the number of fine increments loaded into each tube. Figure 6 shows a longitudinal cross section and autoradiograph of a rod section which operated in the highest neutron flux. In-reactor sintering is fairly complete in the high plutonium area but is negligible in the depleted areas. Very little plutonium relocation has occurred under these conditions. A similar cluster was loaded with variable enrichment along the length for longitudinal power flattening and irradiated at the same maximum surface heat flux. The element generated about 30 percent more power and contained about twice as much plutonium enrichment as the equivalent uniformly enriched cluster. Figure 7 shows central void formation and columnar grain growth which occurred in a segregated increment of this fuel. Reduction of increment size and increment mixing by low energy vibration prior to compaction has improved homogeneity.

FIGURE 4

Prototypical Seven-Rod Cluster, Swage Compacted
 UO_2 - 0.5 wt % PuO_2 Test Element

SWAGE COMPACTED UO_2 -0.5 W/O PuO_2 SEVEN ROD CLUSTER

UNIFORM ENRICHMENT
 INCREMENTALLY LOADED (60 INCREMENTS/ROD)
 IRRADIATED IN THE ETR AT PRTR CONDITIONS

FIGURE 5

Autoradiographs of Irradiated Fuel Rods Fabricated by Swage Compaction
 of UO_2 and PuO_2 Particles

FIGURE 6
Longitudinal Section and Autoradiograph
of an Irradiated $\text{UO}_2\text{-PuO}_2$ Particle Fuel

FIGURE 7

Longitudinal Section and Autoradiograph of Cosine-Enriched
UO₂-PuO₂ Particle Fuel

High-exposure plutonium containing larger amounts of Pu²⁴⁰ and Pu²⁴¹ has been used for a portion of the aluminum-plutonium spike elements and for all the uniformly enriched elements. This material was obtained by the irradiation of coextrusion clad aluminum-plutonium alloy fuel rods in another reactor. Use of high exposure plutonium allows an approach to recycle equilibrium in the PRTR at an earlier date. Although radiation levels from high exposure aluminum-plutonium alloys (16 to 30 wt % Pu²⁴⁰) are from 5 to 10 fold higher than comparable low exposure alloys, moderate shielding such as lead-impregnated gloves, quarter-inch thick lead glass and lead impregnated aprons has allowed dose rates to operating personnel to be kept comparable to dose rates experienced with low exposure plutonium. The shielding

effect of UO_2 in handling UO_2 - PuO_2 fuels further reduces this problem. Largest increases have been observed in hand exposures but this is effectively reduced by leaded gloves, use of tongs, shielding of material not being worked on, and a conscientious effort to reduce handling times by operating personnel.

Many estimates have been made in the past of the increased fuel fabrication cost for plutonium enrichment in comparison with U^{235} enrichment. A commonly used value is double the cost of a comparable uranium base fuel. Table I shows some actual out-of-pocket fuel fabrication costs accumulated during the fabrication of fuel for the Plutonium Recycle Test Reactor. While these figures do not include uranium costs, inventory charges, amortization of capital investment, etc., they do reflect additional handling costs required by the toxicity of plutonium. The magnitude of the difference in labor cost for the swaged element is \$7.38/pound of uranium which is equivalent to about 0.20 mils/kwh_e (10,000 MWD/T exposure and 30 percent conversion efficiency).

TABLE I
SOME ACTUAL FUEL FABRICATION COSTS^(a)

	<u>\$/Assembly</u>	<u>\$/Pound-Uranium</u>		
		<u>Zircaloy and Material</u>	<u>Labor</u>	<u>Total</u>
1. Swaged 19-Rod Zircaloy-Clad UO_2 End 1960 Campaign	3500	29.75	3.25	33.00
2. Swaged 19-Rod Zircaloy-Clad, PuO_2 - UO_2 , Early Pilot Plant Stage	3443	21.89 ^(b)	10.63 ^(c)	32.47
3. Vi-Pac, 19-Rod, Zircaloy-Clad PuO_2 - UO_2 , Early Pilot Plant Stage	3595	20.88	13.88	33.96

(a) Hanford out-of-pocket costs only.

(b) Reflects lower Zircaloy prices.

(c) Labor costs reflect considerable hand manipulation which will be reduced by minor amounts of mechanization. Higher labor cost for Vi-Pac over-swaging process is because more development work has been done on swaging. This is expected to be reversed.

Experimentally determined processing rates and yields have been utilized for a very detailed and comprehensive fuel element fabrication cost estimate of rod type fuel bundles enriched with either U^{235} or plutonium.¹⁴ Data has been extrapolated to a manufacturing plant having a capacity of 20 tons of uranium per month. The reference fuel is a 19-rod cluster of the PRTR design made by vibrational compaction of UO_2 or UO_2 and PuO_2 dense particles in Zircaloy tubing. More than 60 cost factors were individually calculated, followed by estimates of normally expected requirements of plant and equipment, direct manpower, and materials. In addition, an FEFC (Fuel Element Fabrication Cost) code was developed¹⁵ and used to determine the effect of increased reject rates and increased labor and equipment costs on overall fuel costs. A result of these studies is shown in Figure 8. The detailed estimate gave a cost of \$28.34/pound uranium (0.787 mils/kwh_e at 10,000 MWD/T) for uranium enriched fuel and \$30.35/pound uranium (0.855 mils/kwh_e at 10,000 MWD/T) for plutonium enriched fuel of equal reactivity. This is about 9 percent premium for plutonium handling or 0.068 mils/kwh_e. If reject rates at each process step in the plutonium case are higher than those assumed, Figure 9 shows a regular increase with no sharp discontinuities. A similar effect results from increased labor and equipment costs.

FIGURE 8

Some Actual Plutonium Fuel Fabrication Costs

FIGURE 9

UO₂-PuO₂ Fuel Element Fabrication Costs - Effect of Increasing Plutonium, Labor, and Equipment Costs at Various Reject Rates (Equal Reactivities)

Reactivity changes with burnup are being measured for plutonium irradiation specimens containing 6 percent, 16 percent, and 27 percent Pu²⁴⁰ plutonium as a Zircaloy-clad aluminum-plutonium alloy.¹⁶ This is preliminary to optimization of the "Phoenix" extended reactivity fuel concept.¹⁷ Extended reactivity lifetime results from the ingrowth of fissionable Pu²⁴¹ by neutron capture to offset the burnout of Pu²³⁹ and the ingrowth of fission product poisons. Full exploitation of this technique required optimization of both neutron spectrum and plutonium isotopic ratios. Reactivity changes illustrated in Figure 10 exhibit the expected relative trends. The higher Pu²⁴⁰ samples lose reactivity at a slower rate than the lower Pu²⁴⁰ samples. The crossover of the 27 percent Pu²⁴⁰ reactivity curve is not understood, but may not occur after correction of the exposure data when flux wire measurements are complete. Additional burnout experiments in hardened spectrums are planned.

FIGURE 10

Reactivity Change with Exposure as a Function of Initial Plutonium Isotopic Content

A plutonium ceramics research program of about one-sixth the size of the development program is illustrated in Figure 11. This program is designed to provide synthesis, properties, chemical stability, and irradiation data on potential plutonium ceramic fuel materials. Initial studies involved UO_2 - PuO_2 solid solution formation and sinterability¹⁸ and currently are concerned with phase equilibria in the plutonium-oxygen system. Preliminary phase and property studies on the PuO_2 - ZrO_2 ,¹⁹ PuO_2 - MgO , and PuO_2 - ThO_2 systems have also been conducted. Plutonium carbides, silicides, and nitrides have been synthesized as pure single-phase compounds and are being studied. Investigation of plutonium sulfides has been initiated.

FIGURE 11

Plutonium Ceramics Research at Hanford

A recent highlight of this program is the proposed²⁰ Pu-O phase diagram, Figure 12. Congruent melting points have been found for $\text{PuO}_{1.5}$ (β - Pu_2O_3) and $\text{PuO}_{1.62}$ (α' - Pu_2O_3) of 2085 C and 2360 C with a eutectic in between. A melting point minima at 2280 C exists between PuO_2 and α' - Pu_2O_3 which always results when PuO_2 is melted in an atmosphere other than oxygen. BCC α - Pu_2O_3 transforms at a temperature less than 900 C to α' - Pu_2O_3 which is FCC and forms a complete series of solid solutions with PuO_2 . A eutectoid is also observed at $\text{PuO}_{1.70}$ and 300 C.

FIGURE 12

Proposed Pu-O Phase Diagram

Many improvements in the processes for fabricating reliable thermal reactor fuels with plutonium enrichment are possible which would result in reduced manual manipulations, increased fabrication rates, and corresponding monetary savings. While some fabrication cost penalty for plutonium enrichment will always exist, it may not be significant in the overall fuel cycle economics.

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PLUTONIUM FUEL PROGRAMS AT ARGONNE NATIONAL LABORATORY

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Most of the plutonium fuel programs at ANL are aimed at the utilization of plutonium in fast reactor power systems. Our principal incentive and instrument for the development of plutonium-bearing fuels is the second loading of EBR-II. EBR-II is a small fast reactor power plant operating on a closed fuel cycle and will be described in some detail by L. J. Kock in Paper 16 during Session III. The composition of the fuel depends to a large extent upon the nature of the reprocessing method chosen for recycling the irradiated fuel. In the interests of speed and simplicity, a melt refining process has been developed by the Chemical Engineering Division of ANL. Short cooled fuel is melted and held molten in a zirconium oxide crucible whereby fission gases and volatile fission products are vaporized and rare earth elements are oxidized by reaction with the crucible. Zirconium, Mo, Tc, Ru, Rh, and Pd are not at all or only partially removed and remain with the fuel alloy. Zirconium can be removed by reaction with carbon but this now appears to be an unnecessary and indeed an undesirable operation. The mixture of residual fission products is called fissium (Fs) if low in zirconium and fizzium (Fz) if higher in zirconium. To prevent the continual buildup of residual fission products, a portion of the melt refined fuel is subjected to a secondary operation which removes these residuals. After many recycles, the equilibrium fuel will contain residual fission products as alloying elements; the relative amounts (i. e., the composition of Fs or Fz) depends upon the fission products spectrum of the fuel and the efficiency of removal by the main line reprocessing method; the level of alloying (i. e., the total amount of Fs or Fz) depends upon the fraction of the main line stream diverted to the secondary, more complete decontamination process. In any case, the reprocessed fuel is still highly radioactive and must be refabricated into fuel elements by remote methods in heavily shielded enclosures.

REFERENCE FUEL CORE II OF EBR-II

1. The fuel alloy should be based on uranium-238 so that some measure of internal breeding is possible.

2. The plutonium concentration should be sufficient to operate the reactor at the desired power level. For EBR-II this would be in the neighborhood of 35 to 40% Pu. A look at the U-Pu phase diagram (Figure 1) indicates that an alloy containing this much plutonium is not likely to be a practical fuel material and that metallurgical considerations would set a limit on plutonium at about 20%. Actually this backoff in plutonium concentration is not a serious problem. Large and intermediate reactors would operate with plutonium concentrations in the range of 10-20%. The additional reactivity required to operate a small reactor such as EBR-II can be supplied by additions of U-235 without altering the metallurgical characteristics of the fuel. The reference fuel therefore contains 20% Pu and study material covers the range from 10 to 20% Pu.

URANIUM - PLUTONIUM
(Ellinger; Elliot and Cramer)

FIGURE 1

Uranium-Plutonium Phase Diagram

ANL Negative Number 106-6826

ALL GE RICHLAND WASH

3. Alloying elements of some kind and amounts are to be added to the basic U-Pu alloy to improve the metallurgical properties and resistance to irradiation damage. Fortunately, all of the fission elements are desirable alloying elements in uranium and indeed the U-Fs alloys (without Pu) are among the most irradiation stable of the uranium base alloys. We will therefore take this seminatural mixture as our alloying material. A minimum of 5 w/o Fs is required to produce much improvement in properties; 15 w/o Fs represents a quite considerable dilution of the fuel by inert elements; 10 w/o has been chosen as the reference level. It should be noted that the fraction of the main stream reprocessed material that must be diverted to the secondary reprocessing cycle is inversely proportional to the fission level.

4. The basic reference alloy for EBR-II is therefore a U²³⁸-20 w/o U²³⁵-20 w/o Pu-10 w/o Fz. Variants of this basic composition are of interest for use in larger reactors, in particular, the U²³⁵ could be reduced to residual levels and the plutonium reduced to 10-15 w/o. The Fs level can also be adjusted to achieve optimum stability with uranium drag-out reprocessing. Typical compositions are given in column 1, Table I.

TABLE I
TYPICAL REFERENCE FUEL ALLOYS

<u>Element</u>	<u>Recycle Fuel (7-12 w/o Fz)</u>	<u>Initial Loading Core II</u>
Zirconium	0.5-1.0	1.0
Molybdenum	1.7-2.9	2.92
Technetium	0.6-1.1	0.00
Ruthenium	3.5-4.3	4.20
Rhodium	0.4-0.5	0.65
Palladium	1.4-2.5	2.17
Uranium-238	Balance	Balance
Uranium-235	15.0-18.0	15.0-18.0
Plutonium	18.8-15.0	18.8-15.0

THE INITIAL LOADING

It is desirable that the feed material to the recycle plant be relatively fixed in composition, even on the first recycle of relatively low burnup fuel, before process equilibrium has been established. Further, the residual fission product elements are necessary to achieve phase stability during irradiation in the reactor. We will, therefore, choose an initial loading composition which approximates the equilibrium composition as closely as possible. It is not possible to duplicate the equilibrium composition even if we knew accurately what it is going to be. Two examples

will demonstrate the problem. Technetium exists only as a fission product and not enough is available to make the required amount of fuel alloy. We approximate the alloying effect of technetium by assuming that it behaves like Mo and Ru and we arbitrarily increase the amounts of Mo and Ru each by half of the expected amount of Tc. There is also uncertainty regarding the behavior of Zr during recycle. We will somewhat arbitrarily set the initial Zr concentration at 1 w/o. As a result of these adjustments, the initial loading will have the composition given in column 2 of Table I.

PROPERTIES OF U-Pu-Fs ALLOYS

Another reason for limiting compositions is that the property measurement problem became impossibly large if we were to consider all possible compositional variables. Small variations in composition normally encountered in melting and casting these complex alloys do not appear to have any appreciable effects upon the properties. The nominal compositions under study are given in Figure 2. A condensed summary of the pertinent properties is given in Table II. The Table also includes a detailed listing of the nominal fissium and fizzium compositions. Figure 3 (106-6906) illustrates one property, thermal conductivity, in somewhat greater detail. It is evident that the heavy alloying has adversely affected thermal conductivity but that variations in plutonium and fizzium within the chosen ranges have relatively little effect. Thermal cycling stability of the reference alloy is excellent to 500°C; small but tolerable shrinkage occurs upon cycling to 700°C. The reference alloy can be cast into long thin fuel pins (L/D ratio ~100) by the injection casting process. Figure 4 (106-5743) is an assembly drawing of such a machine designed for remote operation in the Fuel Cycle Facility of EBR-II. Partially decontaminated fuel from the melt refining operation is remelted in the crucible shown at the center of the Figure. The furnace is then evacuated, the crucible raised to submerge the open ends of the cluster of Vycor molds (120-160 molds) shown in the upper part of the Figure. The furnace is pressurized with argon to force the molten metal into the molds. A batch of U-Pu-Fs alloy pins cast in a similar but smaller furnace is shown in Figure 5 (106-3153).

The reference alloy suffers from one serious and perhaps lethal defect: it is not very resistant to irradiation damage. Figure 6 (106-5471 Rev.) illustrates the nature and magnitude of the problem. Serious swelling occurs at burnups as low as 1% at temperatures as low as 400°C. This tendency to swell badly at low burnup and at low temperature is entirely unacceptable and some method of dealing with this problem must be developed if the U-Pu-Fs alloys are to be used at all in fast power breeders. One approach is illustrated in Figure 6; the U-10% Pu-10% Fs alloy is more resistant to swelling than is the U-20% Pu-10% Fs alloy by a factor of 9. Indeed the swelling problem pretty well vanishes even at much higher burnups and temperatures if the plutonium content is reduced to zero but this solution is unacceptable from the reactor breeding standpoint. A second approach is through the use of a strong refractory alloy jacket to mechanically restrain the swelling. This

approach is illustrated in Figure 7 (106-6383 Rev.). These specimens have been carried to somewhat higher burnups at considerably higher temperatures without appreciable deformation of the jacket itself. The fuel has swelled, mostly radially, with a volume expansion of about 15%, which is all the initial geometry would allow without deformation of the jacket. Without the jacket restraint, the fuel would probably have expanded more than 100% under these conditions of burnup and temperature. The question as to whether jacket restraint will allow us to reach our goal of 5% burnup at central metal temperatures of 700°C is not yet answered. Recent experience has not been favorable in that some failures have been observed at burnups as low as 1.3% at temperatures of 700°C. It may be that both approaches will be necessary; i. e., the plutonium content could be cut back to 10 or 15% to improve the inherent stability of the fuel alloy, and jacket restraint used to further improve stability. In this connection, it can be mentioned that the Core I fuel alloy (U-5 w/o Fs, no plutonium) jacketed in refractory alloy tubing has been carried to 5% burnup without difficulty.

FIGURE 2

Uranium-Plutonium-Fissium Study Compositions

TABLE II
PROPERTIES OF URANIUM-PLUTONIUM-FISSIUM ALLOYS

Property	Alloy			Type ^(a)
	U-20 Pu-5 Fs	U-20 Pu-10 Fs	U-20 Pu-15 Fs	
Density, gms/cm ³ , 25°C	17.86	17.03	16.38	Fs
	17.72	16.84	16.14	Fz
Hardness, R _A , cast	75	65	76	Fs
	84	75	73	Fz
Solidus, (°C)	860	820	805	Fs
Liquidus, (°C)		1010		Fz
Thermal Expansion 10 ⁻⁶ /°C, average coefficient, 25°C to 500°C	18	16		Fs
Thermal Conductivity cals/sec-°C-cm at 500°C		0.052		Fs
	0.060	0.062	0.060	Fz
		0.064		Fs
	0.071	0.077	0.074	Fz

(a) Fs refers to low zirconium fissium
Fz refers to high zirconium fissium

Low Zirconium Fissium Composition for a 20 w/o Pu²³⁹-20 w/o U²³⁵ Fuel

Zr, w/o	0.04	0.08	0.12
Mo, w/o	1.88	3.76	5.64
Ru, w/o	2.05	4.10	6.15
Rh, w/o	0.31	0.62	0.93
Pd, w/o	<u>0.72</u>	<u>1.44</u>	<u>2.16</u>
Total Fs, w/o	5.00	10.00	15.00

High Zirconium Fizzium Composition for a 20 w/o Pu²³⁹-20 w/o U²³⁵ Fuel

Zr, w/o	1.40	2.80	4.20
Mo, w/o	1.87	2.75	4.62
Ru, w/o	1.98	2.95	4.93
Rh, w/o	0.25	0.50	0.75
Pd, w/o	<u>0.50</u>	<u>1.00</u>	<u>1.50</u>
Total Fz, w/o	5.00	10.00	15.00

FIGURE 3

Thermal Conductivity of U-Pu-Fz Alloys,
U-5 Fs and Uranium

FIGURE 4
Injection Casting Furnace

FIGURE 5
Cast U-Pu-Fs Fuel Pins

EFFECT OF Pu CONTENT ON THE BEHAVIOR OF U - 10 w/o Fs ALLOY
IRRADIATED TO 1.1 a/o BURNUP AT 400°C

FIGURE 6

Effect of Pu Content on the Behavior of U-10 w/o Fs Alloy
Irradiated to 1.1 a/o Burnup at 400°C

Vanadium-clad

304 S.S.-clad

Nb-1 Zr-clad

FIGURE 7

Clad U-20 Pu-10 F's Alloy After 1.6 a/o Burnup at $\sim 650^{\circ}\text{C}$

The use of restraining jackets has involved us in two other programs not directly concerned with the development of plutonium-containing fuel materials. The first concerns the selection of refractory alloys having suitable high temperature strength and creep resistance and their procurement or fabrication in the form of small diameter, thin wall tubing, (approximately 0.15-inch I.D. with 0.009-inch wall). There is a large national effort on the development of high hot strength refractory alloys and there is little we can add to this already massive effort. Very few of the potentially useable materials are available in small diameter, thin walled tubing, and we have started a small program on special drawing processes for fabricating such tubing. Since the fuel alloy will swell tightly against the jacket a second program involves the compatibility of fuel and jacket material. Figure 8 (106-5972) illustrates some of the preliminary results. It is evident that stainless steels are not satisfactory even at temperatures as low as 550°C, and that several of the refractory metal elements are satisfactory. Testing of a variety of refractory alloys is now in progress. The jacket material must also be resistant to attack by the sodium coolant. All of the refractory metals are resistant to very high purity sodium. It is not yet known how resistant suitable high strength refractory metal alloys are to reactor grade sodium containing the expected 20-30 ppm of oxygen and perhaps some hydrogen.

OTHER METALLIC ALLOY FUEL SYSTEMS

The Th-U-Pu ternary system contains all of the fertile and fissionable isotope combinations. The three binaries have been studied but so far as we know, no work has been done on the ternary system. We have started a study of the ternary system with particular attention to the Th and U corners and to the band of composition lying parallel to the U-Th binary and extending out to about 20% Pu. Pertinent properties of compositions which look promising on the basis of the phase study will be determined as part of the program. Our interest in the thorium corner of the diagram stems from the well-established fact that uranium is a pretty poor carrier for fissionable isotopes and requires considerable doctoring up by alloying before being even marginally useable in high burnup, high temperature fuel systems. Fairly extensive irradiation experiments have demonstrated that Th-U alloys containing up to 20% U have good stability at burnups up to 5% at temperatures up to 700°C. Figure 9 (106-6825), the Russian version of the Th-Pu diagram, indicates extensive solubility of plutonium in f.c.c. thorium. Preliminary irradiation tests on U-Pu alloys containing 5 and 10% Pu have also been encouraging. A more extensive set of specimens containing higher concentrations of plutonium is being prepared. These will be irradiated to higher burnups at central metal temperatures of approximately 700°C.

Inert metals such as zirconium can also be considered as carriers for plutonium. Figure 10 (106-6824) shows the Harwell version of the Zr-Pu diagram and indicates appreciable solubility of Pu in α -Zr. That the use of an inert carrier for plutonium does not automatically solve all irradiation stability problems is illustrated in Figure 11 (106-5478). A very remarkable amount

FIGURE 8
Diffusion Penetration of Potential Clad Materials
by U-20 w/o Pu-10 w/o F's

FIGURE 9
Thorium-Plutonium Phase Diagram

ZIRCONIUM-PLUTONIUM
(Marples, Poole, Waldron,
Williamson and Angwin)

FIGURE 10

Zirconium-Plutonium Phase Diagram

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Before
Irradiation

Zr - 5 w/o Pu
1.4 a/o Burnup

Zr - 7 w/o Pu
1.9 a/o Burnup

THE INITIAL IRRADIATION TEMPERATURE WAS NEAR 500° C.

FIGURE 11

Effect of Irradiation on Cold-Worked Zirconium-Plutonium Alloys

of anisotropic growth has occurred in this cold worked alloy. Presumably the anisotropic growth could be reduced or eliminated by proper fabrication and heat treatment procedures but no experiments to demonstrate this have been performed.

REFRACTORY COMPOUNDS AS FUELS

Finally there are a variety of high melting refractory compounds that can be considered as fuel materials in high temperature reactors. Our interests have been principally in the UC-PuC pseudobinary system and we have recently completed a study of the U-Pu-C ternary out to the 50 a/o C pseudobinary. Our interest in this system has been stimulated by the discovery by the reprocessing group in Chemical Engineering that U in Zr-Mg solutions can be precipitated as UC by reaction with finely divided carbon. We have undertaken a study of the sintering characteristics of these powders with rather encouraging results. Plutonium carbide has also been precipitated from metal solutions by the same process. Rather soon we expect to receive coprecipitated (U, Pu)C powders. If these powders can be sintered to a high density body with good irradiation stability, we would have available a pyrorefining process for the rapid recycling of carbide fuels. Uranium sulfide (US) and uranium phosphide (UP) are also interesting possibilities. Methods of preparing such materials on a reasonable scale have been developed and we are now in a position to prepare PuS and PuP by similar methods. Property studies of the (U, Pu)S and (U, Pu)P pseudobinary systems will follow. It is also very possible that these refractory compounds can be precipitated from liquid metal solutions.

SUPPORTING FUEL PROGRAMS

To this point, the discussion has been concerned with fuel materials containing modest concentrations (up to 20%) of plutonium and which are of interest as fuel for power producing reactors. There are other classes of reactors which are of importance in the development of power reactors; I refer to zero power reactors (critical experiments) such as ZPR-III or very small, low power reactors such as EBR-I which operate on nearly pure fissionable isotopes. For these applications we have been using plutonium base alloys containing 8-10 a/o Al. Some 350 kilos of a Pu-8.25 a/o Al have been procured from an outside fabricator for use in ZPR-III and we have recently completed the fabrication of the Mk-IV loading of EBR-I (about 35 kg of Pu-10 a/o Al).

The pertinent phase diagram is given in Figure 12 (106-5487). The feature of particular interest is the marked expansion of the temperature range of existence of the face centered cubic delta phase. In the neighborhood of 10 a/o Al the delta phase is stable from room temperature to about 650°C. These alloys are soft and ductile; they can be easily cast into ingot and rolled or swaged to plate, sheet, or rod. The alloys have a moderate freezing point and can be easily cast into long, thin pins by the injection casting technique described earlier.

7.18

FIGURE 12
Phase Diagram of Plutonium-Aluminum System

HW-75007

A summary of some of the more important properties is given in Table III. Of particular importance is the effect of aluminum upon the thermal expansion coefficient of delta plutonium. Pure δ -Pu has a small negative expansion coefficient, the 8-10 a/o Al alloy has a moderate, and positive coefficient, a feature of importance in reactor safety considerations. Figure 13 illustrates this effect.

TABLE III
SOME PROPERTIES OF PLUTONIUM-RICH
PLUTONIUM-ALUMINUM ALLOYS

	Composition	
	1 w/o (8.23 a/o) Al	1.25 w/o (10.02 a/o) Al
Density (gm/cm ³)	15.3	15.1
Mean Linear Thermal Expansion (20° to 450°C) (μ in/in/°C)	10.9	14.1
Thermal Conductivity (cal/cm-sec-°C)		
100°C	0.022	
500°C	0.045	
Hardness (R _F)	41	72

The Pu-1.25 w/o Al alloy is very soft (R_F 72) and weak and some concern has been expressed about its tendency to slump under its own weight and under small, externally applied loads. Figure 14 shows this effect in an as cast rod of Pu-8.25 a/o Al alloy. At each holding temperature, the initial rate of slump is high but decreases with holding time at temperature. Increasing the aluminum content to 10 a/o Al decreases the amount of slumping at any particular temperature and, as suggested by Figure 14, annealing to homogenize the cored as cast structure is also beneficial. The 10 a/o Al alloy appears to be useable up to temperatures of about 500°C.

FIGURE 13

Mean Linear Expansion of Pu-Al Alloys Between 100°-500°C

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AEL GE RICHLAND WASH

FIGURE 14
Change in Length of a Pu-1 w/o Al Specimen as a Function
of Time on Heating and Holding at Several Temperatures

PLUTONIUM FUEL PROGRAM IN CANADA

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INTRODUCTION

The Canadian plutonium fuel program had its origin more than a decade ago when a demand arose for several enriched fuel rods for the NRX Reactor. Processes for the preparation and fabrication of aluminum-plutonium alloys were developed and some 50 rods of different designs were irradiated. The results were encouraging. The alloys proved to be easily prepared and manipulated in simple air-filled glove boxes and they were dimensionally stable in the reactor.^{1, 2, 3, 4} After irradiation times extending up to three years with plutonium burn-ups of 65%, for example, no measurable distortion was produced in 10-foot long aluminum-clad fuel assemblies.

Once the interest in Canada shifted from water-cooled research to power reactors it was natural that an attempt should be made to utilize the experience already gained with aluminum-plutonium alloys. The zirconium-base alloy Zircaloy-2 which had been developed as a sheathing material in the USA was becoming available commercially and it appeared to have chemical and mechanical properties which would make it compatible with Al-Pu cores at coolant temperatures near 300 C. Although metallurgical bonds between Zircaloy-2 and Al-Pu were not expected to be stable during thermal cycling because of the four-fold difference in the thermal expansion coefficients of the two, it seemed feasible to design an element with a diametral gap at room temperature which would close up at the operating temperature. Thus, both out-pile and in-pile tests were initiated to determine the limitations of unbonded assemblies. The results have indicated that Zircaloy-clad Al-Pu fuel cylinders should be highly satisfactory as a form for the recycling of plutonium in water-cooled power reactors.⁵

The interest in Al-Pu alloys remains high at Chalk River but there has been a growing conviction that an economical method for plutonium utilization is one where the plutonium is fissioned in situ in the parent uranium fuel, hence obviating the necessity for chemical reprocessing, fission product disposal and refabrication steps.⁶ The uranium oxide fuel in the CANDU reactor serves as a good example of such a concept. About half of the energy from the expected output of 10,000 MWd/tonne U will come from the fissioning of plutonium. That plutonium remaining in the fuel will have grown to an appreciable amount, 0.4%, before the elements have been discharged from the reactor ready for disposal. Thus an obvious motivation exists for the study of the interactions between PuO₂ and UO₂.

The Canadian plutonium fuel program is, therefore, centered around two possible fuels, Al-Pu alloys and $\text{UO}_2\text{-PuO}_2$ ceramics. Since no requirement exists at present for the development of a specific plutonium-containing fuel, advantage is being taken of the interlude to learn more of the fundamental properties of the two systems.

As an aid to such investigations a new plutonium laboratory at Chalk River has been completed recently. A schematic layout of the laboratory which measures some 2500 ft² in area is shown in Figure 1. Equipment has been installed to permit alloy preparation and metal-working, ceramic sintering and grinding, corrosion testing, metallography and mechanical testing. In Figure 2, two sintering furnaces in the ceramic fabrication line may be seen. All of the glove boxes are constructed of stainless steel with plastic viewing panels. Their atmosphere is filtered air at a pressure of 1 inch H₂O below that in the laboratory.

FIGURE 1

Schematic Layout of the Chalk River Plutonium Laboratory
Illustrating the Air-Flow Pattern

FIGURE 2

View of a Glove-Box Line Containing Ceramic Fabrication
Equipment in the Chalk River Plutonium Laboratory

ALUMINUM-PLUTONIUM ALLOY STUDIES

a) Alloy Constitution

Many data on Al-Pu phase equilibria have been published during the past decade but there is still much to learn about the system. The solid solubility limits of plutonium in aluminum for instance have not been specified for a range of temperatures although a value for one temperature has been published. Hall reported from autoradiographic studies at Hanford on diffusion couples that 0.26 wt % plutonium dissolved in aluminum at 600 C.⁷ Recent studies at Chalk River by Roy have given much lower values.⁸ At the eutectic temperature of 650 C, for example, he found that the solid solubility was only 0.05 wt % plutonium.

The position of the solidus was determined by the metallographic detection of liquation in alloys heated above the eutectic temperature. When the solubility limit was exceeded the excess plutonium collected at

grain boundaries and boundary junctions. The curved configuration of the interfaces at the ternary junction, illustrated in Figure 3, is clear evidence that liquation has occurred. An electron probe microanalysis* of such a junction in a comparable Al-U alloy showed that it was of eutectic composition with a uranium content of 13 wt %.

FIGURE 3

Polished Section of an Aluminum - 0.095 wt % Uranium Alloy Heated for 4 Hours at 652.5 C and Furnace-Cooled Showing the Concentration of UAl_4 at the Grain Boundaries. The Liquation Effect on the Ternary Junction is also Visible.

Sub-eutectic heat treatments of Al-Pu alloys have been carried out by Roy also.⁸ Metallographic observations have indicated that the solid solubility of plutonium in aluminum decreases with decreasing temperature.

* Courtesy of the International Nickel Company Research Laboratories, Bayonne, N. J.

Other phase-studies on aluminum-rich alloys have revealed that the intermetallic compound PuAl_3 behaves in a surprisingly complex manner when heated and cooled. For nearly 10 years the stable atomic configuration of PuAl_3 was thought to be hexagonal.^{9, 10, 11} Recent work has shown, however, that hexagonal PuAl_3 is a high temperature modification only, forming from a phase of rhombohedral crystal structure in heating past 1027 C.¹² Normally hexagonal PuAl_3 crystals are plate-like in appearance (Figure 4). The new phase looked somewhat similar but occasionally trigonal crystals of markedly different morphology could be found as indicated by the arrow in Figure 5. The pseudo-hexagonal cell of rhombohedral PuAl_3 has lattice dimensions of $a_H = 6.155 \text{ \AA}$ and $c_H = 21.13 \text{ \AA}$ and contains 9 plutonium and 27 aluminum atoms.

FIGURE 4
Single Crystals of Hexagonal PuAl_3

FIGURE 5

Several Crystal Habits for Single Crystals of Rhombohedral PuAl₃
(Rhombohedral edges are visible in the trigonal crystal indicated by
arrow)

It has also been found that when the hexagonal phase is heated to 1210 C a transformation to cubic PuAl₃ occurs.¹² On cooling, the cubic to hexagonal transformation appears to be quite rapid since it was difficult to preserve the cubic phase except by a rapid quench to room temperature. Large cubic single crystals have been recovered from alloys made with only 95% pure plutonium where the major impurities were iron, chromium and nickel (Figure 6). Using plutonium of 99.8% purity polycrystalline cubic aggregates have been retained in alloys quenched from as high as 1395 C.

FIGURE 6

Single Crystals of Cubic PuAl₃

In the latest version of the Al-Pu phase-diagram published by Ellinger, et al.¹³ the transition from PuAl₂ to PuAl₃ was reported to occur at 1220 C. The Chalk River results indicate that the thermal arrest they reported was probably due instead to the polymorphic transformation from cubic to hexagonal PuAl₃.

Two unidentified X-ray powder diffraction patterns have been obtained lately from samples of alloys on the plutonium-rich side of PuAl₂.

Perhaps still further modifications of the phase-diagram may be required.

b) Chemical and Physical Properties

The phenomenon of eutectic spheroidization affects many of the properties of binary Al-Pu alloys. Jones has found, for instance, that their corrosion resistance is markedly reduced when as-cast structures are spheroidized by a 20 h heat treatment at 610 C. Whereas a cast Al - 5 wt % Pu cylinder of 0.25-inch diameter had a corrosion film only 0.004-inch deep after 20 h in static water at 280 C, the spheroidized sample had corroded completely through.¹⁴

It has been observed that the effective corrosion rate of Al-Pu alloys is lowered appreciably once enclosed in a defected Zircaloy sheath.⁵ Only 0.002-inch penetration of the cast Al - 5 wt % Pu alloy core illustrated in Figure 7 had occurred after 20 h in water at 340 C, the bare alloy would have been completely corroded.

FIGURE 7

Cross-Section of a Defected Zircaloy-2 Clad Al - 5 wt % Pu Cylinder Which had been Heated for 20 h in Water at 340 C.⁵
(Note that little corrosion of the core has occurred.) X 45

In spite of the high corrosion resistance of the Zircaloy-clad assemblies there may be a need to further improve the corrosion properties particularly for those highly-rated elements which might reach a core temperature where spheroidization could occur. Jones has made ternary additions of several elements such as Ni, Cu and Si to Al - 5 wt % Pu alloys and has determined their effect on the corrosion behaviour of the resulting alloy.¹⁴ Additions of 1% Ni have proved particularly effective. The corrosion film on Al - 5 Pu - 1 Ni was only 0.0004-inch deep after heating a cast cylinder for 20 h in H₂O at 380 C. In addition, spheroidization did not increase the corrosion rate of the ternary alloy.

Jones has also carried out thermal conductivity measurements on Al-U alloys at 60 C¹⁵ as a prelude to similar measurements now underway on Al-Pu alloys. No significant changes were produced by heat treatments at 620 C of cast alloys containing up to 12 wt % U. A 21 wt % alloy, however, showed a 23% increase in conductivity after spheroidization. Above 30 wt % U, heat treatments had a detrimental effect on conductivity. After polished sections had been studied the results could be accounted for qualitatively if one assumed that most of the heat was flowing through the α -aluminum solid solution around the uranium-containing intermetallic barriers.

Wauchope observed several years ago that inverse segregation occurred in castings of Al-Pu alloys.⁴ More recent work at Chalk River on 1.4-inch diameter cylinders containing 3.8 wt % Pu has demonstrated that there is an appreciable plutonium exudation to the surface.¹⁴ Increases of up to 15% have been measured in a 0.012-inch thick outer layer while depletions at the center of some castings have fallen to 40% below the mean concentration.

c) Irradiation Tests

Some additional metallographic data on the X-2-m loop charge have become available¹⁶ since the first results were reported at the Grenoble conference.⁵ From Figure 8 it is clear that the interaction between an Al - 15 wt % Pu core and a Zircaloy-2 sheath for an interfacial temperature of 350-400 C is only slight after four months' irradiation in the X-2 loop of the NRX reactor. The intermetallic layer at the interface is about 0.0004-inch thick.

FIGURE 8

Interaction Between an Al - 15 wt % Pu Alloy Core (Light Phase) and a Zircaloy-2 Sheath (Dark Phase) after 4 Months' Irradiation in the X-2 Pressurized Water-Cooled Loop at an Interfacial Temperature of 350 - 400 C.¹⁶ X 725

Metallographic examination has also indicated that the anodic layer which was formed on one of the Al - 10 wt % Pu alloys prior to irradiation disappeared during the loop test.¹⁶ The suspicion is that the oxygen from the anodized layer has moved into the 0.001-inch thick diffusion zone shown in Figure 9 forming a solid solution with the zirconium.

FIGURE 9

Postirradiation Polished Section of an Anodically-Treated Al - 10 wt % Pu Alloy (Dark Phase) Sheathed in Zircaloy-2 (Light Phase) After 4 Months' Irradiation in the X-2 Loop at an Interfacial Temperature of ~ 400 C.¹⁶ X 725

Seven alloys containing from 2 to 10 wt % Pu sheathed in 0.010-inch thick stainless steel cans were irradiated successfully in the X-7 organic-cooled loop of NRX for one month.¹⁷ Fuel temperatures of up to 570 C were recorded by thermocouples during the course of the irradiation. Sheath-core interfacial temperatures ranged from 380 to 440 C. No detectable sheath-core interaction occurred for temperatures below 420 C and only slight amounts above that. Eutectic spheroidization was observed in one of the elements irradiated at a core temperature near 500 C. (See Figure 10).

FIGURE 10

Microstructures of an Al - 7.2 wt % Pu alloy before (a) and After (b) an Irradiation of 1 Month at 500 C in the X-7 Organic-Cooled Loop.¹⁷ (Note that some spheroidization has occurred.) X 500

URANIUM DIOXIDE-PLUTONIUM DIOXIDE STUDIES

a) Sintering Behaviour

The development of a low temperature sintering process using an inert gas atmosphere for the preparation of UO_2 - PuO_2 pellets offers many advantages for glove box operation, not the least of which is the elimination of the use of hydrogen gas during the sintering period. At Chalk River the effort has been concentrated on an investigation of argon-sintering techniques.

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Michaud and Boucher¹⁸ reported that mechanical mixtures of 2% PuO_2 with UO_2 pressed at 60,000 psi could be sintered to densities in excess of 95% of the theoretical maximum after 2 h in Ar at 1450 C. In later experiments on UO_2 alone, Boucher found that even higher densities could be achieved at lower temperatures.¹⁹ After 2 h at 1050 C, for example, pellet densities reached 98% of the theoretical maximum.

The rapid sintering rates are attributed to the slight oxidation of the uranium oxide to the nonstoichiometric form by traces of water in the flowing argon stream. As is well-known the sinterability of UO_{2+x} is much higher than that of the stoichiometric oxide.²⁰ Since it is desirable, however, to reduce the O:U ratio to 2.0 to improve the performance under irradiation,²¹ a hydrogen reduction step was carried out at the end of the sintering period.¹⁸ A reduction temperature above 1250 C was required to prevent cracking of the pellets during the removal of the excess oxygen.¹⁹

Some of Boucher's sintering tests indicated that the sintered density of UO_2 was reduced by about 5% when 2% PuO_2 was added to UO_2 , but only when the PuO_2 dissolved in the UO_2 lattice.¹⁹ That is, if coarse, dense PuO_2 particles were used, prepared from say 1450 C-sintered PuO_2 , little interaction with the UO_2 occurred and there was no significant reduction in the pellet density. A polished section of such a pellet is shown at low magnification in Figure 11. PuO_2 particles measuring up to 0.5 mm diameter are readily visible. One of the particles is seen at higher magnification in Figure 12. It will be noted that a void exists around the particle and it is denser along its periphery. This and other metallographic evidence indicated that some uranium had diffused into the PuO_2 lattice. After two weeks at 1500 C in Ar the porosity in the PuO_2 had coalesced but there had been little Pu diffusion into the UO_2 (See Figure 13).

b) Irradiation Tests

There has been some interest at Chalk River in heterogeneous sinters such as those just described since they offer the prospect of recycling plutonium in a form which can be kept separate from the new plutonium generated within the UO_2 during irradiation. Two types of UO_2 - 2% PuO_2 pellets were recently irradiated, therefore, to compare their performance under irradiation. One type contained 1450 C-calcined coarse (-30+50 mesh) PuO_2 particles dispersed throughout the UO_2 sinter. The other type contained finely-divided PuO_2 prepared from the oxalate at 600 C. Microscopic examination indicated that it had been homogeneously distributed in the UO_2 matrix during sintering. All pellets were sintered for 2 h at 1450 C in Ar, then reduced for 0.5 h at 1450 C in H_2 .²²

Six Zircaloy-2 clad elements, three of each type, 0.8-inch diameter and 5-inches long were fabricated. They were irradiated for 2 months in the X-2 pressurized water-cooled loop of the NRX Reactor at power outputs up to 700 W/cm of fuel length. Only small changes in the external dimensions of the elements were produced by the irradiation but interesting changes occurred in the oxide cores.

8.14

FIGURE 11

Cross-Section of a Sintered UO_2 - 2% PuO_2 Pellet of Density 10.6 g/cm^3 Showing the Dispersal of 1450 C-Calcined PuO_2 Particles. X500

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FIGURE 12

Cross-Section of a PuO₂ Particle in a Dense UO₂ Matrix after Sintering for 2 h at 1450 C in Ar Followed by Reduction for 1/2 h at 1450 C in H₂. X 500

UO_2 PuO_2

FIGURE 13

Cross-Section Through a UO_2 - PuO_2 Junction After Sintering a UO_2 - 2% PuO_2 Pellet for 2 wk at 1500 C in Ar Followed by Reduction for 1/2 h at 1450 C in H_2 . X 500

The most highly-rated element (680-700 W/cm) containing the homogeneous pellets exhibited extensive columnar grain growth as shown in the stereoscopic cross-section in Figure 14. The comparably-rated element (660 W/cm) filled with heterogeneous pellets showed much less grain growth (Figure 15) and liberated only half as much fission gas during the irradiation.²³ The PuO_2 particles outside the region of grain growth in the latter element were still clearly visible. The particles on the original end faces of the pellets appeared to have lost plutonium from their centers since they were saucer-shaped (see Figure 16).

FIGURE 14

Stereoscopic End View of Zircaloy-Clad UO_2 - 2% PuO_2 Sintered Pellets After an Irradiation of 2 Months in the X-2 Loop at a Power Output of 680 - 700 W/cm. (The PuO_2 had been prepared from the oxalate at 600 °C and had been homogeneously distributed in the UO_2 matrix.)
X 4.2

FIGURE 15

Stereoscopic End View of Zircaloy-Clad UO_2 - 2% PuO_2 Sintered Pellets After an Irradiation of 2 Months in the X-2 Loop at a Power Output of 660 W/cm. (The PuO_2 had been prepared by crushing 1450 C-sintered PuO_2 pellets of density 10.5 g/cm³ and using the -30+50 mesh fraction. It was heterogeneously distributed in the UO_2 matrix.) X 4.2

FIGURE 16

Stereoscopic View of a PuO_2 Particle from the Outer Region of an Original End Face of a Heterogeneous UO_2 - 2% PuO_2 Pellet After Irradiation for 2 Months at an Element Power of 660 W/cm. X 40

Polished sections revealed more clearly the difference in the extent and nature of grain growth in the two types of fuel (Figure 17).²⁴ The PuO_2 particles within the region of columnar grain growth of the heterogeneous sample were no longer present. Only a few holes similar to that shown in Figure 18 remained. Even in the outer region of equiaxed grain growth the plutonium-containing particles had shown signs of break-up and dispersal into the UO_2 matrix (Figure 19). Nevertheless, one would estimate from the metallographic data that about 75% of the PuO_2 had not dissolved in the UO_2 . Chemical dissolution tests are underway to determine the actual amount that remains insoluble in hot nitric acid.

FIGURE 17 - Polished Cross-Sections of the UO_2 - 2% PuO_2 Pellets Shown in a) Figure 15 and b) Figure 16 Which Illustrate the Differences in Nature and Extent of Grain Growth and Plutonium Distribution. X 7.5

← Center of Element

FIGURE 18

Cross-Section at High Magnification of the Center Area of the Heterogeneous Pellet Shown in Figure 17 b) Illustrating that a 1450 C-Calcined PuO_2 Particle has been Dispersed into the UO_2 Matrix Leaving a Hole. X 125

Center of Element —→

FIGURE 19

Cross-Section at High Magnification of a Region from the Heterogeneous Pellet Shown in Figure 17 b) Where Equiaxed Grain Growth Occurred During Irradiation Illustrating Some Dispersal of a PuO_2 Particle into the Surrounding UO_2 Matrix. X 125

A microstructure of the columnar grains in the homogeneous specimen revealed the presence of lenticular voids similar to those observed in UO_2 subjected to a steep thermal gradient as illustrated in Figure 20. Such voids are believed to move up the thermal gradient leaving columnar-shaped grains in their wake.²⁵ An attempt is being made by Durham to determine if any plutonium redistribution has been produced in this sample by the irradiation. Early results indicate that some plutonium may have migrated to the hot center of the element since the Pu content of a 0.070-inch diameter trepanned sample from the center was 15% higher than that in an outside sample.²⁶

Center of Element —————>

FIGURE 20

Cross-Section at High Magnification from the Region of Columnar Grain Growth of the Homogeneous Pellet Shown in Figure 17 a) Illustrating Lenticular Voids.

The more extensive grain growth produced in the homogeneous pellets when compared with the heterogeneous ones is an interesting phenomenon and might be explained by:

- a) the 5% higher heat rating in the homogeneous fuel,
- b) a 4% lower density for the homogeneous pellets,
- c) a possible reduction in thermal conductivity produced by dissolving PuO_2 in the UO_2 lattice.

Whatever the causes, it has been interesting to observe that large refractory PuO_2 particles can be added to UO_2 without producing deleterious irradiation effects. In fact, there seems to be some advantage in adding the plutonium in such a form.

SUMMARY

Several studies on aluminum-plutonium alloys and UO_2 - PuO_2 ceramics are being carried out in a new plutonium laboratory at Chalk River.

The solid solubility of plutonium in aluminum has been determined and polymorphic transformations in Al-Pu intermetallic compounds are being investigated. The effect of spheroidizing the Al- PuAl_4 eutectic on properties such as corrosion resistance and thermal conductivity is being studied. Alloys containing ternary additions to improve the corrosion properties are being tested. New data from irradiation tests at core temperatures up to 570 C indicate that Al-Pu alloys should be satisfactory for use in high temperature water or organic coolant when sheathed with either Zircaloy-2 or stainless steel.

The sintering properties of UO_2 - PuO_2 pressed pellets have been studied in argon over a range of temperatures with both finely-divided and coarse, refractorized PuO_2 additions. Irradiation tests of the two types of pellets produced have revealed interesting differences in behaviour.

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PLUTONIUM FUEL DEVELOPMENT PROGRAMS
AT
NUCLEAR MATERIALS AND EQUIPMENT CORPORATION

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Nuclear Materials and Equipment Corporation has great confidence in the future of plutonium as a power reactor fuel. This confidence is so strong that NUMEC, which already has invested in plutonium facilities more than any other private organization, is now expanding these facilities still further.

In this paper, the NUMEC capabilities for plutonium fuel development are described. The current programs are discussed, and some recent analytical and experimental data are presented.

Figure 1 is a plan view of the facility as it will look in December 1962 after completion of construction of a 14,000 square foot addition. With this addition, the building will contain 38,000 square feet of floor space. In general terms, the building is divided into a front "clean" area and a rear working area with these areas separated by change rooms. The front "clean" area contains offices, a technical library, a digital computer facility, a drafting room, and a combination conference room-cafeteria with kitchen attached. Additional offices are located upstairs above the eastern end of the working area and within the working area proper. The working area is 60-feet wide by about 380-feet long and is functionally divided into a number of large rooms with smaller supporting laboratories all along the south face. Going from west to east, the working area is divided into a hot cell facility to allow for postirradiation examination of fuel materials, an oxide chemistry and ceramics area, a fuel element fabrication and metalworking area, an area for the development of technology on other refractory plutonium compounds, a plutonium-beryllium source manufacturing area and an area devoted entirely to transplutonium element or high alpha technology. The supporting laboratories are devoted to analytical and radiochemistry, experimental physics and electronics, non-plutonium powder characterization, metallography, electron microscopy, and health physics. Also included is a counting room, a plutonium storage vault, a decontamination room, and a small machine shop.

The hot cell facility contains a relatively large hot cell with walls composed of high density concrete. This cell is capable of handling up to 200,000 curies of cobalt-60 equivalent and is equipped with removable alpha enclosures to permit operation with alpha-active materials without contamination of the cell proper. Also included in this facility are two steel cells. These also contain alpha enclosures and are used for postirradiation examination of small specimens including metallography and fission gas release and burnout determinations.

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Figure 2 is a picture of the oxide chemistry and ceramics area showing the glove box line and the interconnecting transfer tunnel. This tunnel contains an electrically operated cart by which materials and tools can be readily transferred from one glove box to another. Experience has shown this tunnel to be most valuable in that time-consuming bagging operations and the attendant production of bagging waste materials are minimized. Since only one man is allowed to operate the transfer cart, this method of making transfers also provides convenient and strict administrative control of fissionable material within the boxes that are mass-limited for criticality control. It is seen that the glove boxes are built on a modular basis wherever possible; i.e., depending upon equipment space requirements, a glove box is made up of one, two, three, or more individual modular units. Of course, some equipment items are of such shape that this philosophy cannot be followed throughout.

This oxide chemistry and ceramics area contains both batch and continuous pilot scale precipitation equipment, controlled atmosphere calcining and reduction furnaces, powder characterization equipment such as a B.E.T. and an Innes apparatus for measuring surface area, a Whitby (Mine Safety Appliance) centrifuge, a Fisher sub-sieve sizer, and a Sharples micromerograph for measuring particle sizes, an Aminco and a Chevenard thermobalance, and a Deltatherm differential thermal analysis unit. The area also contains a variety of powder milling, grinding, and blending equipment, a laboratory press, an automatic Stokes pellet press, vacuum and controlled atmosphere sintering furnaces, and a Thermal Dynamics plasma torch. Equipment is also installed to allow determination of high temperature physical properties such as thermal conductivity, melting point, and thermal expansivity. Also included is a Phillips X-ray machine for X-ray fluorescence analysis and for diffraction analysis at both room and elevated temperature. Analytical chemistry equipment installed in this area includes a dual grating optical spectrograph and Leco induction heated gas analyzers for determination of carbon, oxygen, and hydrogen. Also included are facilities for electron microscope sample preparation and metallography. The metallography preparation equipment includes a NUMEC cathodic vacuum etcher. Autoclaves are also contained in glove boxes to allow corrosion testing. For scrap recycle and recovery, a series of boxes contains dissolvers and ion exchange and solvent extraction contactors.

The fuel element fabrication and metalworking area covers the range of equipment necessary for the fabrication of fuel elements by the sintered pellet-in-tube route, mechanical packing, swaging and rolling. Also included is an Instron testing machine with auxiliary glove box enclosed attachments to allow determination of mechanical properties.

The supporting laboratories contain a Phillips electron microscope, a Nuclear Data 256 channel analyzer, a controlled potential derivative polarograph, a recording spectrophotometer, a gas chromatograph, a coulometer, a potentiometric titrator, and many more standard analytical and radiochemistry equipment items.

FIGURE 2

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When the additional facilities are completed, NUMEC will have the capability to perform research and development and pilot scale production on essentially all plutonium compounds and alloys.

The purposes for which this equipment is being used can be ascertained from a listing of the various tasks included in our AEC sponsored program* as shown in Figure 3. The NUMEC plutonium fuels program is geared to the development of plutonium technology, in general, without investigation being limited by consideration of only one specific reactor concept. Assurance of emphasis in most promising areas is obtained through investigation of all phases of the fuel cycle including reactor physics and economics analysis. From the listing of the various tasks, it is seen that the end objective is refabrication of high burnup, reprocessed fuel into fuel elements and the subsequent performance evaluation under irradiation.

NUMEC PROGRAM
ON THE
DEVELOPMENT OF PLUTONIUM-BEARING
FUEL MATERIALS

REACTOR PHYSICS, ENGINEERING AND ECONOMICS STUDIES
PREPARATION AND CHARACTERIZATION OF FUEL MATERIALS
ANALYTICAL CHEMISTRY DEVELOPMENT
FABRICATION AND EVALUATION OF FUEL SHAPES
FUEL ELEMENT BUNDLE FABRICATION AND EVALUATION
RADIATION TESTING AND EVALUATION
FUEL REPROCESSING
SHIELDED-CELL FUEL AND FUEL ELEMENT REFABRICATION

FIGURE 3

*Under U.S. Atomic Energy Commission Contract No. AT(30-1)-2389.

Following closely the development of uranium technology, the initial effort has been concentrated primarily on the oxides PuO_2 and $\text{PuO}_2\text{-UO}_2$ mixtures ranging from 0-30 w/o PuO_2 ; i. e., compositions of interest for thermal as well as fast reactor systems. Similarly, fabrication effort has been concentrated on the cold press and sinter pellet-in-tube process. During the coming year, fusion by plasma torch techniques and fabrication by swaging, mechanical packing and rolling will be emphasized to a greater extent as will be the determination of physical and mechanical properties. Also, more basic studies will be initiated on cation and oxygen diffusion, and oxidation-reduction kinetics. In addition, synthesis, characterization, fabrication, and product evaluation will be initiated on oxide mixtures such as $\text{PuO}_2\text{-ZrO}_2$ and on other refractory compounds.

Confidence in plutonium as a power reactor fuel stems directly from the results of physics studies on near-thermal reactor loadings containing plutonium. Figure 4 shows typical curves of k_∞/ϵ , or k_{eff} if you like, as a function of plutonium-to-uranium ratio for a specific pressurized water reactor lattice. For this particular lattice, the results indicate that when plutonium is used in conjunction with natural uranium the reactivity first increases as plutonium is added and then decreases after the plutonium enrichment has exceeded about 3.5 a/o. This behavior is distinctly different from that obtained with slightly enriched uranium where the reactivity increases with increasing U-235 enrichment and approaches a maximum value asymptotically. This "negative resistance" behavior is due to the strong resonance absorption in Pu-240 which tends to increase appreciably, relative to absorption in other isotopes or elements, with the increasing neutron temperature that comes about when more absorber is added to a particular system. For the particular cases analyzed, the Pu-240 content was taken to be 12%. More generally, additional calculations indicate that such behavior can be obtained with plutonium having more than 10% Pu-240. This decrease in reactivity with increasing plutonium enrichment gives direct indication that plutonium reactors of this sort tend to have a relatively high obtainable core life (based on physics considerations alone) without appreciable associated changes in reactivity with time. Specifically, this can be illustrated by considering a reactor initially loaded with 6 a/o plutonium. Initially then, this reactor will have a k_∞/ϵ of about 1.06. As the reactor is operating and as we are not considering breeders, the plutonium-to-uranium ratio will decrease; hence, one may consider time as a continuous shifting towards lower plutonium enrichments. From the figure, it is therefore seen that the reactivity will tend to increase with time. This tendency, however, will be balanced out by the buildup of fission products which were not considered in the figure. The net result is that the reactivity remains nearly constant down to an enrichment of about 3.5 a/o. In studies that we have performed, results indicate that a core life of about 60,000 MWD/Tonne of heavy atoms yields optimum fuel cycle costs with plutonium.

FIGURE 4

Initial k_{∞}/ϵ for Various Lattice Parameters

Figure 5 shows these optimum fuel cycle costs and compares costs attainable with plutonium-natural uranium loadings to those attainable with slightly enriched uranium loadings. To develop this figure, calculations were performed for each lattice (denoted by the moderating power per heavy atom, $\xi \Sigma_s / \Sigma N_i$) using different enrichments, and then fuel cycle costs were calculated in order to arrive at an optimum for each point. These series of optima were then plotted to produce the two curves. In the economic analyses, the old UF_6 prices were used and the plutonium worth, both as a buy-back price and as a purchase price, was taken as \$12 per gram. The fuel costs were calculated for cores containing 20 tonnes of heavy elements which with a heat flux limitation of 100,000 BTU/hr-sq ft gives a core power output of about 500 MW thermal or 150 MW electrical. Further, to arrive at the optimum it was assumed that sufficient control rods, burnable poisons or solution poisons were available in each case to hold down the reactivity throughout the cycle. From the curves it is seen that the optimum fuel costs, on this basis, for the slightly enriched uranium system is about 2.52 mils/KWH and this occurs in a lattice having $\xi \Sigma_s / \Sigma N_i = 100$. Not shown on this curve is the fact that at this point the initial reactivity without burnable poisons is 1.3 and that this does not vary appreciably for off-optimum design points. Correspondingly, for the plutonium system, the optimum fuel cost is 2.2 mils/KWH, and this occurs for a lattice having a $\xi \Sigma_s / \Sigma N_i = 150$. The corresponding k_{eff} at this point for the plutonium

system is only 1.18. In fact, operation slightly away from the optimum fuel cost point, at $\xi \Sigma_s / \Sigma N_i = 130$ gives a corresponding initial k_{eff} of only 1.1. Based on the cost assumptions utilized, it is seen therefore that a plutonium fueled reactor is capable of achieving lower fuel costs than a corresponding uranium fueled reactor and that this can be achieved with less stringent control requirements. It is further seen, however, that the plutonium lattice must be somewhat looser than the corresponding uranium lattice in order to allow achievement of this cost advantage. Since a looser lattice generally means higher capital costs because the reactor pressure vessel must be larger for a specific heat output, calculations have also been performed to study the effects on fuel cycle costs of dilution of plutonium-natural uranium with relatively inert ZrO_2 . The results indicate that with such dilution plutonium can be utilized in lattices comparable to those giving optimum fuel costs for uranium without corresponding significant increases in fuel cycle costs. In fact, it was found that for a lattice equivalent to $\xi \Sigma_s / \Sigma N_i = 100$ the fuel cycle cost increased by only 2 percent over the optimum. From these calculations it is concluded that plutonium has a great potential as a power reactor fuel in near-thermal converter type systems.

FIGURE 5

Optimum Fuel Cycle Costs for Plutonium-Natural Uranium
 and Slightly Enriched Uranium Fuel Loadings
 in Near-Thermal Converter Reactors

Figure 6 shows the various preparation routes that we have used and are planning to explore for conversion of plutonium nitrate to plutonium dioxide. Since our interests to date have been in the preparation of PuO_2 for subsequent pelletizing and sintering, the major effort has been on the plutonium oxalate route for the preparation of ceramic grade powders having both intermediate and high surface areas. For the production of dense granular feed to be used, hopefully, directly for swaging and mechanical packing, the sol gel process and the homogeneous oxalate precipitation process will be studied.

FIGURE 6

Preparation Routes for Plutonium Dioxide

Preparation routes for PuO_2 - UO_2 are shown in Figure 7. Again, the initial interest has been on ceramic grade powders; hence, effort has been concentrated on the ADU-plutonium hydroxide routes using both dilute and concentrated ammonia. The other routes for the production of dense granular feeds as well as ceramic grade powders will be studied during the coming year.

FIGURE 7

Preparation Routes for Plutonium-Uranium Dioxide

Figure 8 shows typical conditions used for the preparation of PuO_2 and $\text{PuO}_2\text{-UO}_2$ ceramic grade powders. The resulting powders have been characterized in detail to show the effects of modifications in the various preparation steps.

	PuO_2	$\text{PuO}_2\text{-UO}_2$
PRECIPITATION METHOD	CONTINUOUS	CONTINUOUS
FEED SOLUTION COMPOSITION	(Pu) 0.4-0.5M (H^+) 3M (NO_3^-) 4.6-5M	(U+Pu) 0.4-0.5M (H^+) 1M (NO_3^-) 2.6-3M
PRECIPITANT COMPOSITION	$\text{H}_2\text{C}_2\text{O}_4$ - 1M H_2O_2 - 0.8M	NH_4OH - 14.5M
PRECIPITATION TEMPERATURE	35°C	55°C
PRECIPITATOR RESIDENCE TIME	20 Min.	30 Min.
INTERMEDIATE COMPOUND	PLUTONIUM IV OXALATE	AMMONIUM DI-URANATE - PLUTONIUM HYDROXIDE
DRYING TEMPERATURE OF INTERMEDIATE COMPOUND	180°C	180°C
FURNACE TEMPERATURE FOR CONVERSION TO OXIDE	740-840°C	740-840°C
FURNACE ATMOSPHERE COMPOSITION	AIR	H_2 - 6% H_2

FIGURE 8

Typical Preparation Conditions for PuO_2 and $\text{PuO}_2\text{-UO}_2$ Powder

Figure 9 shows the effect of heat treatment on the surface area and porosity of PuO_2 powders. It is seen that both these powder characteristics vary in similar fashion. In contrast, studies of the effect of conversion temperature have indicated that the porosity increases up to a temperature of 560 C, then drops off rapidly, while the surface area remains essentially constant up to a temperature of 490 C, then drops off linearly.

FIGURE 9

Effect of Heat Treatment on PuO_2 Properties

Figure 10 shows the effect of the reduction temperature on the surface area of UO_2 -20% PuO_2 for material precipitated with low and high concentration ammonia. It is seen that for the concentrated ammonia case, the surface area of the product can be adjusted by choosing the appropriate reduction temperature. In contrast, for the dilute ammonia case, a reduction temperature above 850 C is required to effect a change in surface area.

FIGURE 10

Effect of Reduction Temperature and Ammonia Concentration on Surface Area of Mixed Uranium-20% Plutonium Oxide (Continuous precipitation process, -70 minute reduction)

Figure 11 shows the effect of plutonium-to-uranium ratio on the surface area of mixed oxide powders produced with dilute ammonia and reduced at 740 C. In comparison, for the concentrated ammonia case, the surface area is less sensitive to plutonium concentrations, up to 20% plutonium content; for higher plutonium concentrations, the surface area decreases rapidly.

FIGURE 11

Effect of Composition on Surface Area
of Uranium/Plutonium Dioxide

Figure 12 shows the results of a number of pellet sintering trials using the powders derived from the preparation procedures discussed. The powders were pre-slugged and granulated, then finally pressed into pellets at 9-18 TSI without binder addition. The results show that all compositions can be sintered to better than 92% of theoretical density. Sintering PuO₂ in air generally results in higher final density than sintering in a reducing atmosphere; also, as will be discussed later, no Pu₂O₃ is formed during air sintering. For mechanically mixed component powders, an appreciably longer sintering time is required to attain density than for similar coprecipitated compositions. Even after 16 hours of sintering at 1600 C, the density of pellets made from mechanically mixed powders is lower than that achieved after one hour sintering of pellets made from coprecipitated feed.

COMPOSITION	PREPARATION	GREEN DENSITY gm/cc (I)	FIRING TIME HOURS	FIRING ATMOSPHERE	SINTERED DENSITY gm/cc(I)	PERCENT THEORETICAL DENSITY
PuO ₂	OXALATE	6.79	1	N ₂ -6%H ₂	10.60	92.6
PuO ₂	OXALATE	6.80	1	AIR	10.90	95.0
UO ₂ -5%PuO ₂	COPRECIPITATE	5.50	1	N ₂ -6%H ₂	10.55	96.1
UO ₂ -5%PuO ₂	MECHANICAL MIX	5.85	16	N ₂ -6%H ₂	10.20	92.9
UO ₂ -20%PuO ₂	COPRECIPITATE	5.10	1	N ₂ -6%H ₂	10.40	94.0
UO ₂ -20%PuO ₂	MECHANICAL MIX	6.05	16	N ₂ -6%H ₂	10.25	92.7
UO ₂	ADU	5.18	16	N ₂ -6%H ₂	10.56	96.2

(I) GEOMETRICAL DENSITY

FIGURE 12

Sintering Characteristics of UO₂-PuO₂
(Sintering temperature - 1600 C)

As a cost reduction feature, one of the objects of the program has been to compare and evaluate performance of coprecipitated mixed oxide feed and feed prepared by the mechanical blending of the component UO₂ and PuO₂ powders. In order to ascertain the degree of homogenization obtained with various blending procedures, autoradiographs of sintered pellets were taken. Figures 13a and 13b show these autoradiographs. These pictures were taken by placing pellets on mylar and then on an alpha sensitive film and exposing for 45 seconds. It is seen that overnight twin-shell blending is unsatisfactory for producing homogeneous material since the component powders tend to form spherical particles prior to mixing with the other component. What is seen in the picture are spheres having essentially PuO₂ cores, then a layer of UO₂, then a layer of mixed UO₂-PuO₂, or the reverse. Since these pictures are negatives, the light portions indicate PuO₂ rich zones, the dark portions, UO₂ rich areas. The next picture shows the effect of subsequent hammermilling in a Bantam micropulverizer. It is seen that the hammermilling tends to break up the spheres into odd shaped particles but that homogeneity is still not satisfactory when compared to the autoradiograph of the coprecipitated feed. Standard wet ballmilling for 4 hours following the twin-shell blending yields a homogeneity that is nearly equivalent to that obtained with coprecipitated material. This homogenization technique is now being used for mixing component powders and in-pile irradiation tests are planned to determine the difference in performance between the coprecipitated and blended material.

TWIN-SHELL BLENDED

TWIN-SHELL BLENDED
AND HAMMERMILLED

FIGURE 13a
Autoradiographs of Sintered UO_2 -20 w/o PuO_2 Pellets

TWIN-SHELL BLENDED
AND WET BALLMILLED

COPRECIPITATED FEED

FIGURE 13b

Autoradiographs of Sintered UO_2 -20 w/o PuO_2 Pellets

Capsule irradiations to be performed during the coming year will consist of both short-term screening tests and high-burnup performance tests. UO_2 - PuO_2 fuel compositions ranging from 0.5 to 30 w/o PuO_2 will be irradiated to allow assessment of performance as a function of composition. Also, as stated above, both coprecipitated and mechanically mixed materials will be utilized. Initial irradiations will be performed on samples prepared by the cold press, sintered pellet-in-tube route. Subsequent tests will be performed with mechanically packed and also swaged specimens. To allow for meaningful interpretation of results, thorough pre- and post-irradiation examination and evaluation is contemplated.

Figure 14 shows photomicrographs of a PuO_2 pellet sintered at 1600 C in a reducing N_2 -6% H_2 atmosphere for one hour. The light phase is PuO_2 . The striated area is a eutectoidal mixture of PuO_2 and alpha Pu_2O_3 . The pictures also illustrate the difference between acid etching and cathodic vacuum etching. It is seen that cathodic vacuum etching brings out the grain boundaries. Experiments have indicated that Pu_2O_3 is formed when sintering at temperatures above 1600 C in vacuum or in non-oxidizing atmospheres. When sintering in air or in inert atmospheres containing some moisture, only single phase PuO_2 has been observed. Since differential thermal analysis indicates a phase change at about 650 C in material that has been sintered in a reducing atmosphere, it is postulated that at this temperature there is a eutectoidal transformation of an oxygen deficient PuO_x phase into PuO_2 and alpha Pu_2O_3 .

In connection with the preparation of dense particulate feed for swaging and mechanical packing, a plasma torch has been utilized to fuse PuO_2 and UO_2 - PuO_2 powders. For PuO_2 , Figures 15a and 15b show the feed materials utilized as well as pictures of the final product. Crushed sintered pellets have been used as plasma torch feed; however, if a particular product size is desired, it was found that too much material is lost as fines. It, therefore, appears that this type of feed is not economical. To circumvent this material loss, feed was also prepared by mechanically preforming nearly spherical material and furnace sintering this prior to introduction to the torch. For both types of feed, the products were essentially identical, provided that slight modifications were made in torch conditions. It is seen that the product consists of almost perfect spheres. Sections of these spheres show that the density of most particles is close to the theoretical density with some particles exhibiting voids. Pycnometric density measurements indicate that a density of 95 percent of theoretical was obtained.

A paper of this limited length covering such a broad range of technology cannot go into satisfactory detail; however, I hope that I have conveyed to you the dynamism behind NUMEC's efforts on plutonium fuels development. We are confident that plutonium will be a significant power producer before the end of this decade and that through its use, competitive nuclear power will be assured without artificial incentives.

HNO₃ ETCHED

CATHODIC VACUUM ETCHED

FIGURE 14

Typical Photomicrographs Showing PuO₂ and PuO₂/ Pu₂O₃ Eutectoid Structure
Sintered at 1600 C for 1 Hour in N₂-6% H₂ (800 X)

FIGURE 15a
Plasma Torch Fusion and Spheroidization PuO_2

TYPICAL TORCH PRODUCT

FIGURE 15b
Plasma Torch Fusion and Spheroidization PuO_2

PREPARED DISCUSSION, SESSION IBATTELLE'S COMPOUND AND PLUTONIUM ALLOY
DEVELOPMENT PROGRAMS

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COMPOUND DEVELOPMENT PROGRAM

Battelle's effort in the non-metallic, or compound, phase of plutonium technology has been directed primarily at plutonium dioxide (PuO_2), and this presentation will be restricted to this subject.

Although the experimental work is overlapping in a sense, results will be categorized into two divisions: (1) the vapor pressure of PuO_2 in non-reducing atmospheres and (2) the compatibility of PuO_2 with various potential reactor construction materials. The compatibility results are reported in BMI-1580, "The Chemical Reaction of PuO_2 with Reactor Materials", while the vapor pressure information has just been released as BMI-1591, "The Volatility of PuO_2 in Non-Reducing Atmospheres".

The chemical reactivity of PuO_2 with various ceramic and metallic materials has been investigated by thermodynamic computations, metallographic examinations, and X-ray diffraction measurements after sintering abutted interfaces at temperatures representative for densification of the nonfissile material. It was found necessary to calcine plutonium oxalate at a minimum temperature of 600 C to completely eliminate hexagonal Pu_2O_3 and produce homogeneous PuO_2 . Sintering in dry hydrogen at 1750 C caused melting of the oxide containing Pu_2O_3 . Compatibility results showed that PuO_2 is compatible with MgO , austenitic stainless steels, Inconel, chromium, molybdenum, niobium, and vanadium. Maximum temperature investigated for these materials was 2500 F. ZrO_2 and PuO_2 are mutually soluble, while PuO_2 and Al_2O_3 react to form PuAlO_3 under reducing conditions. Titanium and tantalum are not compatible with PuO_2 and Zircaloy-2 reacts badly. Tungsten is apparently not completely inert in contact with PuO_2 .

With regards to the volatility of plutonium oxides, the vapor pressure was investigated in the temperature range 1450-1775 C in air, argon, and oxygen atmospheres by a transpiration technique. Data has been collected and thermodynamic calculations performed which show that PuO_2 can vaporize congruently or as a sub-oxide specie, probably PuO , depending on the atmosphere. ΔH_{298} for vaporization in one atmosphere oxygen is 153,800 cal/mole. The standard free energy of gaseous PuO_2 is $-121,000 + 10.7 T$ from 1500 to 2000 K. An estimated erosion rate at 1700 C is approximately 3 mils per year. The actual vapor pressure limits in the various atmospheres are given in Table I.

TABLE I
VAPOR PRESSURE OF PuO₂

<u>Atmosphere</u>	<u>Temperature, C</u>	<u>Vapor Pressure, atm</u>
Oxygen	1500	5.0×10^{-10}
Oxygen	1775	3.81×10^{-8}
Air	1450	1.12×10^{-9}
Air	1775	3.84×10^{-8}
Argon	1450	2.1×10^{-9}
Argon	1750	2.71×10^{-8}

These values are in themselves slightly ambiguous as experimental scatter and differences in temperature limits exist. However, when plotted with the other data obtained in the various carrier gases, they show a distinctly increasing volatility under progressively less oxidizing conditions.

PLUTONIUM ALLOY DEVELOPMENT PROGRAM

In the past year, plutonium alloy development efforts at Battelle have been mainly concerned with the iron-plutonium, niobium-plutonium-silicon, and uranium-niobium-plutonium alloy systems.

Iron-plutonium, iron-plutonium-chromium, and iron-plutonium-molybdenum alloys were studied in this phase of the program. All the alloys contained approximately 30 volume percent PuFe₂. Efforts to spheroidize the PuFe₂ phase by heat treating at 1000 C met with varied success. Some spheroidization occurred in the iron-plutonium and iron-plutonium-molybdenum alloys while very little was observed in iron-plutonium-chromium alloys. Fabrication tests of as-cast alloys indicated that the alloys are capable of sustaining reductions of about 25 percent in thickness by rolling at 900-950 C before cracks appear on the surface and edges. Tentative solidus temperatures of iron-27.6 w/o plutonium, iron-27.6 w/o plutonium-11.4 w/o chromium, and iron-27.6 w/o plutonium-2.1 w/o chromium alloys are 1162, 1148, and 1159 C, respectively.

Studies of the phase relationships in the niobium-plutonium-silicon alloy system indicated that niobium and PuSi₂ exist in equilibrium. Once this was established, a niobium-30 volume percent PuSi₂ alloy was selected for further investigation. Heat treatment studies indicated that the PuSi₂, present as a semi-continuous network in the as-cast alloy, could be spheroidized by a heat treatment of 16 hours at 1000 C. In preliminary fabrication studies, the heat treated alloy exhibited enough ductility at 550 C to indicate that substantial reductions could be made at higher temperatures.

Studies of this portion of the program have been confined to the effects of heat treatment on the structure and room temperature hardness of uranium-10 w/o niobium-5, 10, 15, and 20 w/o plutonium alloys. On the basis of metallographic examination of quenched alloys, approximately 20 w/o plutonium is soluble in gamma uranium-10 w/o niobium at 900 C. Aging the quenched alloys for 2 hours at 500 C resulted in microstructures consisting of gamma uranium solid solution and what is believed to be alpha uranium and/or zeta uranium-plutonium. The amount of gamma phase decreased with increasing plutonium. Structures of alloys furnace-cooled from 900 C consisted primarily of gamma solid solution and a small amount of transformation product at the grain boundaries. In general, the hardness of the alloys increased with increasing plutonium content in all conditions of heat treatment.

PREPARED DISCUSSION, SESSION IPLUTONIUM REACTOR FUELS RESEARCH PROGRAM
AT MOUND LABORATORY

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The Reactor Fuels and Materials Development Program at Mound Laboratory is directed to the determination of fundamental information relative to plutonium-bearing materials which are potential reactor fuels. Fuel materials for fast breeder power reactors which operate at high temperatures are emphasized. The investigations at Mound Laboratory are being accomplished in three major research areas, which are: the determination of plutonium alloy phase equilibria and structures; the measurement of physical and thermal properties of plutonium systems; and the formulation and characterization of plutonium-bearing ceramic systems.

PLUTONIUM ALLOY PHASE EQUILIBRIA STUDIES

The plutonium alloy phase equilibria studies are directed toward the development of complete phase diagrams of plutonium alloy systems which have application for both molten and solid reactor fuel studies. The phase relationships of binary and ternary systems investigated have been defined by the coordinated efforts of differential thermal analysis and metallographic studies with analytical support from X-ray diffraction techniques and the electron microprobe X-ray analyzer.

The differential thermal analysis apparatus developed at Mound Laboratory for the phase equilibria studies provides a rapid method of investigation with high sensitivity and requires only a small sample size, between 0.5 and 1.0 cc. The apparatus has the capability of detecting a heat unbalance of less than 0.1 C between sample and reference materials. Thus, the delta to delta-prime transformation in plutonium appears as a sharply defined transformation, and the alpha to beta transformation is observed to start at 112 C.

The present apparatus has a selection of heating or cooling rates from 1/4 C to 6 C per minute. The analyzing range is from room temperature to 1100 C. The accuracy of temperature measurement is limited by the ability to read the chart paper which is ± 1.5 C.

The samples for all plutonium alloy studies are contained in tantalum capsules which are sealed by inert gas arc welding. Each capsule is 3/8-inch diameter by 2-1/4-inches long with a 0.030-inch wall thickness and contains a thermowell which extends 1/2-inch into the base of the tube, Figure 1.

FIGURE 1

A differential thermal analysis apparatus which will extend the temperature range of operation to 1800 C is under development. This unit will have equivalent sensitivity and accuracy to the three current apparatus. With this additional temperature range for the differential thermal analysis apparatus, plutonium alloys with high melting metals as well as the reactions of plutonium alloys with possible container materials can be investigated. Adaptation of the apparatus for quantitative measurement of heats of transformation is being developed.

Specimens of plutonium alloys for metallographic studies are prepared routinely in a metallographic preparation area enclosed in an alpha glovebox train. The samples are obtained from either the differential thermal analysis tubes or from vacuum melted alloys. Annealing studies are conducted as required using vacuum encapsulated specimens. The combined effects of ultrasonic decontamination and surface passivation by etching permit limited handling of the metallographic specimens during examination and photography on the metallograph. Disposable plexi-glas stages on the metallograph prevent contamination of the instrument.

Similarly, microhardness testing is conducted with a disposable cover on the stage. An open exhaust duct near the stage of each instrument removes any small release of loose contamination.

A vacuum quenching furnace is used in the phase studies of low melting, alloy systems. A sample contained in a tantalum foil cup is equilibrated at a temperature at which solid and liquid exist together in equilibrium. A quench is effected when the cup is dropped into a chilled oil bath in the bottom of the vacuum furnace. On quenching, the liquid portion of the metal forms an extremely fine grained structure which is easily recognized as "quenched liquid". The large well formed crystals of the primary phases which were already present are effectively isolated for positive identification.

A vacuum hot-stage, useful to 1000 C, is also used in the study of plutonium and its alloys. The hot-stage microscope is equipped with a motion picture camera so that the solid state transformations of the specimens can be observed visually or filmed in time sequence.

X-ray diffraction methods are used for crystallographic classification and structure analysis of phases encountered in the phase equilibria studies. Both powder and single crystal methods are used, the former for convenience in routine work, the latter for uniqueness of crystallographic classification and for complete structural analyses of compounds.

The electron microprobe X-ray analyzer has been a valuable analytical instrument for the elemental analysis of very small volumes of low vapor pressure materials. In this apparatus, selected areas on polished samples are bombarded with a well defined, high-energy electron beam of preselected cross-section of 1 to 10 microns.

The instrument, as applied to the plutonium alloy phase studies, provides a rapid means of phase identification by composition analysis of a particular area in the metal matrix. Also, the limits of solid solubility and the shape of the solidus lines of a system are defined by direct analysis of annealed and quenched specimens. Uncoated plutonium metallographic samples placed in the instrument chamber have resulted in negligible radio-contamination. Other applications include identification of corrosion products, diffusion compositions and intergranular precipitates and deposits.

The investigation of phase equilibria in the plutonium-cerium-copper ternary system is currently in progress. Initially, all three binary systems were investigated. The major portion of the liquidus surface of the ternary system has been defined. A ternary eutectic at 419 C exists in the cerium-rich portion of the system and another ternary eutectic at 792 C appears in the copper-rich portion of the system. The primary phase field of the binary compound, PuCu_2 , occupies a large portion of the ternary system. A primary phase field for plutonium extends across the diagram to the 419 C ternary eutectic. A reasonably large area exists about this eutectic, extending to approximately twenty atomic percent

plutonium, in which the liquidus surface does not exceed 500 C. Compositions in this area may be particularly applicable as fuels for the molten plutonium fast breeder reactor.

The plutonium-yttrium binary system is of interest as a high temperature alloy since approximately twenty atom percent solid solubility of plutonium in yttrium has been observed. No binary compounds appear to exist in the system and there is no solid solubility of yttrium in plutonium. The ternary systems plutonium-thorium-yttrium and plutonium-thorium-cerium indicate the possibilities for a large amount of plutonium solid solubility at high temperature with only a slight decrease in the solubility at lower temperatures.

PHYSICAL PROPERTIES

The high temperature operation of fast reactors has focused interest on molten plutonium alloys as fuel materials as well as on liquid metal coolants. Increased understanding of the liquid state is required before a reactor design incorporating the circulation of liquid metal fuels can be made practicable. The scarcity of reliable literature data dictated that the required basic information on molten metals had to be determined experimentally. Apparatus have been developed at Mound Laboratory for the measurement of liquid density and viscosity and for the determination of specific heat and thermal conductivity of liquid alloys. A technique is being developed for the measurement of the interfacial tension between molten plutonium and immiscible liquids, such as sodium.

The oscillating cup viscosimeter was developed for the precise measurements of the viscosity of molten metals from their melting points to 1000 C. A second viscosimeter under construction will extend the range of operation to 1300 C. This method offers the advantages of being a simple system to operate, having the metal under investigation in a sealed container, and being an absolute method of measurement. In this type of apparatus the molten metal is contained in a right-circular cylindrical tantalum cup which is attached to a torsion fiber so that the system forms an oscillating pendulum, Figure 2. The damping effect of the molten metal upon the normal oscillations of the pendulum is a function of the viscosity of the liquid. Oscillating with the tantalum container is a four-inch diameter graduated aluminum disk. The motion of the disk is recorded photographically by a motion picture camera sighted through a telescope equipped with a vertical cross-hair. Using the projected picture film, measurements of the ratios of successive amplitudes of the oscillating disk, which can be determined with a precision of one part in ten-thousand, display the damping effect of the liquid upon the pendulum. The period of the oscillating system is determined by a beam of light, reflected from the suspension system, which actuates a photo-electrically-operated clock which determines the period to one-thousandth of a second.

FIGURE 2

The reliability of the viscosimeter was demonstrated when the expected functions of the viscosity of liquid lead, bismuth and zinc were determined over larger temperature ranges than previously reported. The viscosities of molten plutonium, plutonium alloys, lanthanum, cerium, cerium alloys and praseodymium have been measured. The plutonium alloys exhibit very high viscosity values as compared with other molten metals. Future studies will be directed toward comprehensive studies of such systems as: Pu-Zr, Pu-Cu, and the low melting point regions of the Pu-Ce-Co and Pu-Ce-Cu alloy systems. The change in viscosity with composition and temperature furnishes added insight into these alloy systems.

Reliable liquid density values of plutonium fissile materials are necessary for calculations of reactor criticality and for the mathematical solution of the oscillating cup viscosimeter. Accordingly, a vacuum pycnometric method for the density measurement of liquid metals from their melting points to 1000 C was developed. This temperature range of operation is being extended to 1300 C. The pycnometers, fabricated from 1/4-inch diameter tantalum rod, hold 0.5 cc of liquid. Each pycnometer is calibrated with a precision of ± 0.0002 cc.

The apparatus has been used to measure the liquid densities of plutonium, plutonium alloys, cerium, lanthanum, praseodymium, bismuth, lead and tin. The examined lanthanide elements display little change in density as a function of temperature. With an expanded temperature range the liquid densities of high-melting plutonium alloys will be investigated.

Investigations of methods for the measurement of the interfacial tensions of liquid metals have concentrated on modifying the maximum bubble pressure technique, which involves the measurement of the pressure required to force a drop of liquid metal out of a small orifice into a second liquid metal. The interfacial tensions of a number of organic-aqueous mixtures have been successfully measured by this technique. Thin-walled tantalum capillary tubes are being investigated for future use with the plutonium alloys at elevated temperatures.

The thermal properties, specific heats, latent heats of transformations and thermal conductivities, of plutonium metal and alloys are being determined up to 700 C in the high temperature calorimeter developed at Mound Laboratory. A knowledge of the thermal properties is essential not only for the successful operation of a fuel material, but also to obtain fundamental information on the plutonium and alloy phases.

The calorimeter consists of a sample and reference chamber located inside of a cylindrical nickel block. The chambers are connected differentially by means of a platinum resistance bridge network. The thermal properties of a sample are calculated from the observed response of the calorimeter to various thermal transient conditions imposed on the specimens in the sample chamber. Unlike many of the "drop" methods for measuring specific heats, the Mound Laboratory calorimeter maintains the sample at the test temperature.

A telescoped view of the sample chamber is shown in Figure 3. Because of its high electrical resistivity, the three inner forms have been fabricated from beryllium oxide. The innermost form contains a calibrating heater, used to determine the sensitivity of the chamber at a specific temperature. The bridge form, with its beryllia cover is placed over the heating element. The outside form is fabricated from bonded diatomaceous earth to give thermal insulation. Two of these identically telescoped units are placed in a nickel block, four-inches in diameter and six-inches high.

FIGURE 3

The block temperature is controlled to approximately ± 0.01 C by means of a servo-type system. In this system, a D. C. voltage across the temperature sensing element is modulated by the random temperature fluctuations in the block. This signal is amplified many times and returned to the block where it is dissipated as heat. With the control circuit in operation and the block at equilibrium, the observed temperature differential between the two chambers is ± 0.001 C in the lower temperature range.

The temperature inside the sample chamber can be determined to within ± 0.1 C from a measurement of the platinum resistance bridge in the sample chamber.

Under operating conditions, the estimated accuracy for specific heat values is $\pm 1\%$. The precision of the calorimeter, measured at 600 C, is $\pm 0.5\%$.

The specific heats of the alpha and beta phases, the latent heats of transformation of the five solid state transformations, and the heat of fusion for unalloyed plutonium have been determined. The values are in good agreement with the recent data from A.W.R.E., Aldermaston, United Kingdom.

Thermal conductivity measurements of plutonium materials have not been attempted, as yet, in the present calorimeter. Thermal conductivity measurements on related materials indicate an accuracy of $\pm 4\%$.

CERAMIC SYSTEMS

Ceramic fuels, because of their in-pile dimensional stability at high temperature and high burnup, have become of increasing interest. The investigations at Mound Laboratory have included the development and characterization of two types of material in this category: stable microspheres of plutonium oxide, and plutonium-bearing glass.

The method developed for the microspheroidization of high melting oxide particulates involves the use of a plasma torch in which temperatures in the range of 14,000 to 19,000 K are achieved. The unique feature of the method is the employment of an inductively coupled plasma torch. An oxygen-enriched argon plasma constitutes the "flame" established in the coil of a 10 kilowatt, five megacycle frequency, induction heater, Figure 4. The operation is performed in an alpha glovebox built about the plasma torch.

FIGURE 4

Plutonium oxide has been prepared in the form of thermally and chemically stable microspheres, Figure 5. These dense transparent spheroids exhibit a remarkable freedom from loose contamination. No airborne contamination is released. Surfaces which come in contact with the microspheres are not contaminated. This condition permits relatively simple handling of the oxide and makes possible many applications that were previously unattractive because of the hazards normally associated with the handling of plutonium oxide. The density of the microspheres was found to be 11.0 gm/cc or 96% of theoretical crystalline density. The microhardness was found to be 1163 Kg/mm². The chemical durability of the microspheres is so great that extensive leaching in strong mineral acids and caustic solutions did not result in contamination of the leach solutions. Analysis by X-ray diffraction characterized the microspheres as face-centered cubic PuO₂ with a unit cell dimension in close agreement with the accepted literature value for stoichiometric PuO₂. This result is significant since previous investigators have found an oxygen deficient species and/or the presence of more than one plutonium oxide phase in the material remaining after melting PuO₂. The microspheres exhibit excellent dimensional stability during thermal cycling. The higher crystallographic symmetry of stoichiometric PuO₂ is expected to result in better radiation stability than the expanded lattice of the oxygen deficient material.

FIGURE 5

The use of PuO_2 microspheres in dispersion type fuel elements is a potential application. For the production of microspheres covering a wide range of concentration of plutonium, the present investigations include the study of PuO_2 in combination with UO_2 , ZrO_2 and Y_2O_3 . From these components, a single phase solid-solution type material is expected containing a favorable concentration of plutonium oxide.

One program has been directed toward the formulation and characterization of plutonium-bearing glass systems. The studies have been concerned with the incorporation of plutonium oxide into glass, the measurement of the chemical, physical and nuclear properties of the plutonium-bearing glass, and the fabrication of these glasses into various forms.

The parent glass compositions and plutonium oxide are heated to temperatures as high as 2000 C in a platinum-rhodium crucible (Figure 6) for dissolution of the plutonium oxide. Up to twenty weight percent plutonium oxide has been dissolved into a silicate glass system. Five weight percent plutonium tetrafluoride has been dissolved in a fluoride glass system. These glasses are completely vitreous. The hazards involved in the handling of plutonium oxide are greatly reduced by the incorporation of the plutonium compounds into the silicate glass system. Leach tests which determine the chemical durability of the plutonium silicate glasses have shown that glasses can be formulated which resist attack by water and dilute mineral acids. The density and thermal coefficient of linear expansion have been measured on several plutonium glass compositions. Plutonium-bearing glasses exhibit only a slight loss in strength due to the alpha disintegration of the plutonium within the glass structure.

FIGURE 6

These glasses have been shaped into fibers, (Figure 7) rods, tubes, ribbons and microspheres. Plutonium glass microspheres are presently being evaluated by the Argonne National Laboratory. Plutonium glasses in the form of fibers are potential fuels for chemonuclear reactors. The results from one set of fibers irradiated in the Brookhaven Reactor showed that a low temperature irradiation of the plutonium-bearing glass was destructive. High temperature irradiation studies have not been completed. Rods, tubes, ribbons, plates and other massive forms are potential power reactor elements.

FIGURE 7

An investigation by X-ray analysis of non-crystalline structures has been initiated. X-ray intensity curves for plutonium oxide dissolved in a solvent glass have been obtained with monochromatic radiation. From these data, radial distribution curves as a function of interatomic distance will be determined. This Fourier analysis provides interaction peaks which can be interpreted as distances between nearest neighbors and second nearest neighbors. Areas under the peaks give the number of neighbors, or the coordination number.

The radial distribution function of the solvent glass, computed with the aid of the IBM 1620 computer, gave the average silicon-oxygen distance as 1.61 angstroms. The area under this peak indicated that four oxygen atoms are bonded tetrahedrally to each central silicon atom. The geometrical distribution of the plutonium oxide in the solvent glass will be obtained by removal of the electron density distribution function of the solvent glass.

This technique is an extremely useful method in the elucidation of the structure of liquid alloys as a function of temperature and composition since no a priori assumptions as to the nature of their structure are made. An X-ray structural investigation of liquid plutonium and its alloys is expected to give a better understanding of their physical and transport properties, such as viscosity, thermal conductivity and diffusion.

PREPARED DISCUSSION, SESSION IPLUTONIUM FUEL PROGRAMS
OF THE UNITED NUCLEAR CORPORATION

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1. SCOPE

The United Nuclear Corporation is developing uranium-plutonium carbides for fast and thermal reactors. The programs are sponsored by the USAEC and jointly by the USAEC and EURATOM. The United Nuclear Corporation is working in conjunction with their subcontractor, the Carborundum Co. Carborundum Co. is fabricating the fuel and United Nuclear is performing the out-of-pile and in-pile evaluation.

The advantages of the carbides are their high thermal conductivity, high melting point and high fuel density. The high thermal conductivity and melting point should permit a higher, central temperature limited, specific power than either metal or oxide fuel. The advantage of higher specific power is either a smaller core and lower fuel inventory charges. Or the advantage may be utilized as a higher heat generation per unit length, minimizing the degree of subdivision of the fuel (fewer but thicker rods or plates per subassembly), resulting in lower fabrication costs per unit weight of fuel. The higher fuel density of the carbides compared to the oxides leads to higher neutron economy and higher breeding gains.

2. MAJOR ACCOMPLISHMENTS

Initially, an evaluation was made of the effect of the direct substitution of carbide fuels into existing fast reactors (EBR II and Enrico Fermi) on the heat transfer, physics, and fuel cycle cost. The results of this study were presented previously.¹ Since then, progress has been made in the fabrication and evaluation of the carbide fuel.

The purpose of the fabrication program has been to produce high density monocarbide pellets, with reproducible structure and composition. The irradiation program will evaluate the behavior of the 80% UC-20% PuC composition for high burnups at high power levels. The tests will be capsule irradiations in thermal test reactors. The out-of-pile evaluation program will measure the melting point, coefficient of expansion, thermal stability (vapor pressure), thermal conductivity, and compatibility with clad materials of the 80% UC-20% PuC, and the 95% UC-5% PuC compositions.

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1. G. Sofer, J. Sapir, "Potential of Carbide Fuels in Fast Breeder Reactors". Trans. of ANS Meeting, Vol. 3, No. 1. June, 1960.

To date about 1 Kg of solid solution carbide powder, and several hundred high density solid solution carbide pellets have been produced. The powder is produced by the reaction of a pelletized mechanical mixture of UO_2 , PuO_2 and carbon. The reaction is carried out at about 1670 C. The resulting clinker is crushed and ball milled. The powder is cold pressed and sintered into pellets either with or without a nickel sintering aid. A small amount of nickel, such as 0.1 w/o, has a marked effect on the final density and the required sintering temperature. The effect is similar to that of nickel on pure UC, found in our previous investigations. The results of densities obtained are summarized below:

Material	Sintering, 1 Hour	Appr. No. of Pellets Made	Densities g/cc, (% of Theoretical)	
			Average	Maximum
$(\text{U}_{.8}\text{Pu}_{.2})\text{C}_{.95}$	1965 C	200	12.54(92.4)	12.83(94.4)
$(\text{U}_{.8}\text{Pu}_{.2})\text{C}_{.95}$ +0.1 w/o Ni	1590 C	200	13.15(96.8)	13.30(97.8)

Figure 1 shows the essentially single phase microstructure of the carbides made without sintering aid. Traces of Widmanstatten structure are present. Figure 2 shows the two phase microstructure of the material with nickel sintering aid; the second phase is $(\text{U}, \text{Pu})_2\text{C}_3$. The nickel is in solid solution; it appears to promote the formation of sesquicarbide.

FIGURE 1

80% UC-20% PuC, 600 X,
Nitric Acid-Acetic Acid-
Water Etch

Sintered 1990 C, 1 hour in
helium, 12.8 g/cm³, 4.5%
carbon X-ray analysis:
single phase (U, Pu)C,

$a_0 = 4.961 \pm 0.003$ angstrom.

FIGURE 2

80% UC - 20% PuC - 0.1% Ni,
600 X, Nitric Acid-Acetic
Acid-Water Etch

Sintered 1590 C, 1 hour in
helium, 12.91 g/cm³, 4.77%
carbon X-ray analysis:
major (U, Pu)C, faint (U, Pu)₂C₃,
 $a_o = 4.9644 \pm 0.0002$ angstrom.

Pellets have been fabricated for property measurements and irradiation tests. The latter have been ground to tolerances of ± 0.0007 ".

Carbides with and without sintering aid will be irradiated in the near future to burnups of 17,000, 34,000, and 68,000 MWD/T at power levels of 14 and 20 KW/ft. The irradiation specimens are clad fuel rods, about 1/4" OD with a 3" fueled length. The void space within the cladding is filled with helium. The assembly of all the irradiation capsules is currently being completed.

Equipment to measure the out-of-pile properties of solid solution uranium-plutonium carbides has been calibrated with standards and will operate with plutonium in the near future.

Coefficient of expansion is measured with an alumina differential dilatometer to about 1500 C.

The melting point is measured in a tantalum resistance furnace.

Vapor pressure is measured by the Knudsen effusion method in the tantalum resistance furnace. A carburized tantalum cell will be used with a water cooled collector.

Compatibility tests between fuel and cladding will be made in individual capsules. Tests of several thousand hours, up to 820 C are planned.

3. FACILITIES

Two plutonium facilities were designed, constructed and put into operation to carry out the above program. Both facilities operate on the principle of complete plutonium containment, within helium mass spectrometer leak tight boxes. The facilities have operated satisfactorily at zero

level of contamination for about a year and a half. The plutonium handling problem is aggravated by the pyrophoricity and poor oxidation resistance of UC-PuC.

Pure, inert atmospheres are required for the handling of carbides, and many of the initial problems were concerned with the supply of sufficiently inert atmospheres to the glove boxes. These problems were solved by the use of zirconium-titanium gettered, recirculated helium at Carborundum, and once-through helium and nitrogen at United Nuclear. The recirculating helium system which is used for a furnace atmosphere in synthesis and sintering, as well as handling of fine powders, maintains a purity of 3 ppm oxygen and less than 1 ppm moisture. The once through systems are used for the room temperature handling of solid fuel pieces only, and the purity requirement is lower. The once through helium systems maintain less than 100 ppm oxygen and less than 300 ppm moisture. The once through nitrogen systems maintain less than 500 ppm oxygen, but has several thousand ppm moisture because of some of the operations performed in them. Leak tight systems are an absolute requirement for high purity atmospheres.

The Carborundum facility has a work space of 15 by 37 feet, which houses six aluminum frame glove boxes (Figure 3). Three boxes have a helium atmosphere, and three have an air atmosphere. The facility is capable of synthesizing the carbide powder and fabricating it into shapes by pressing and sintering. It is also set up to do carbon analyses, X-ray diffraction analyses, and grinding of parts to close tolerances.

FIGURE 3

Glove Boxes in Carborundum's Plutonium Facility

The United Nuclear facility has a work space of 28 by 32 feet, which houses ten carbon steel glove boxes coated with a corrosion resistant lining and two hoods (Figure 4). One box has a helium atmosphere, six boxes a nitrogen atmosphere, and three boxes an air atmosphere. The facility is set up to do chemical analyses (plutonium, uranium, carbon, iron, nitrogen), metallography, cutting-drilling-grinding of specimens, assembly of fuel rods, and determination of properties such as melting point, vapor pressure, coefficient of expansion, and fuel-clad compatibility.

FIGURE 4

Glove Boxes in United Nuclear's Plutonium Facility

PHYSICS OF PLUTONIUM IN THERMAL REACTORS

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INTRODUCTION

In this topical meeting devoted to "Plutonium as a Power Reactor Fuel" the general background of the subject will be covered by other authors and we will here proceed directly to consider the narrower range implied by the title of this paper.

In considering the use of plutonium in thermal reactors, two general types of fuel are of interest: a uniform mixture of plutonium and uranium oxides in which the plutonium plays the same general role as U^{235} in partially enriched uranium; and plutonium alloyed with some diluent (e. g., aluminum, MgO) to provide fuel for the seed in seed-blanket cores or as a fuel in plutonium burner reactors for special applications. Mixtures of plutonium and thorium may also be of eventual interest for a mixed cycle type of operation.

The isotopic compositions of interest for thermal reactors using mixed oxide fuels are indicated by a study by Eschbach¹ and others to lie in the following ranges:

Pu^{239}	70 - 30 percent
Pu^{240}	15 - 40 percent
Pu^{241}	8 - 16 percent
Pu^{242}	Balance

Concentrations in the fuel will range from a fraction of a percent up to a few percent depending upon the uranium enrichment used for makeup fuel. The lower concentrations would apply for strict recycle type of operation. In alloy fuels the concentration would be similar to U^{235} concentrations for the same applications, i. e., a few atom percent, and the isotopic compositions would fall in the same ranges as indicated above.

The physics of plutonium as a thermal reactor fuel is more complex than that of U^{235} because of the plutonium isotopic chain:

which provides two fissionable isotopes and one fertile isotope of high cross section, Pu^{240} ; and because of the low lying and prominent resonances in the cross sections of Pu^{239} and Pu^{241} and the large resonance in Pu^{240} at 1 ev.

In assessing the current state of the knowledge of the physics of plutonium as a thermal reactor fuel, it may be helpful to draw a comparison with U^{235} . As a starting point for this comparison, we take the experimental

work that has been done with lattices using plutonium as a fuel. Such experiments have been performed with plutonium aluminum alloy, which for physics purposes can be regarded as the same type of fuel as highly enriched uranium. Data on this type of fuel have been obtained for light and heavy water moderation in subcritical and exponential experiments and for graphite moderation in a series of experiments in the PCTR. These experiments have used only one size of fuel rod and have been directed toward providing some preliminary assessment of the physics of such assemblies. All of the experiments were performed with plutonium whose isotopic composition is not representative of material likely to be available from power reactor operation. The experiments in graphite have, however, lately been supplemented by work with plutonium containing higher percentages of Pu^{240} . In the area of fuels consisting of mixtures of plutonium and uranium oxide and which correspond to the case of partially enriched uranium fuels, experimental work is only now beginning.

This collection of experimental data which has been so briefly enumerated here is to be contrasted with the wealth of data obtained by experiments with uranium fuels over a period of nearly 20 years, and such a comparison would not give a misleading impression of the comparative state of our knowledge of the two types of fuels.

On the calculational side, the picture is very much the same. In contrast to the wide variety of computer programs and theoretical methods that have been developed for treating uranium fueled thermal reactors, there have been relatively few programs and methods applied to the fueling of reactors with plutonium. It is not simply a matter of taking the methods developed for uranium and substituting a set of numbers representing the plutonium cross sections for those representing the uranium cross sections. The behavior of the plutonium isotopes in thermal reactors is far more sensitive to the energy distribution of the neutrons than is the case with uranium. This requires a more sophisticated and rigorous approach to many of the problems of reactor analysis.

With respect to the basic nuclear data, moderately satisfactory information now exists on the cross sections of Pu^{239} , Pu^{240} , and Pu^{241} , but these data do not display as high an internal consistency as the corresponding data for U^{235} or even U^{233} and some further improvements in this area are also desirable.

Physics knowledge for use by the reactor designer and operator is today most often presented in the form of computational methods, whether these be formal computer codes or the provision of recipes and equations to be applied in prescribed situations. The work of the reactor physicist is thus to provide such methods, the basic constants which are used in them, and evidence of their efficacy by comparison of their predictions with results obtained in an appropriately broad range of experimental situations. We will follow this order in reviewing the present state of knowledge of the physics of plutonium in thermal reactors.

COMPUTATIONAL METHODS

An important aspect of the use of plutonium as a thermal reactor fuel is the change of concentration of the various isotopes of the fuel as exposure proceeds. The computational methods developed to date have understandably addressed themselves principally to this type of problem. A practical computing method is a compromise between the desire to give a thorough treatment to the neutron economy at each particular stage in the burnup process and the need to progress from stage to stage in following the burnup. Here, as in many other cases, a proper balance is needed between thoroughness and computing time, effort and cost. From a physics standpoint, the burnup calculation itself is reasonably straightforward and is handled in similar fashion in all of the computational methods. Concentration of the various isotopes of interest is known at a given time and their changes are computed over a short additional time interval by taking into account the effects of neutron capture and of radioactive decay, and thus the concentrations at the beginning of the next interval are determined.

To compute the neutron capture the reaction rate for each isotope must be determined and this, in turn, depends upon the distribution of the neutron flux both in energy and in space. The crux of the physics problem lies in this latter determination, and it has been treated in different ways by different workers in the field. At the Hanford Laboratories a code called MELEAGER has been developed for the IBM 7090.² A primary consideration in development of this code was the ability to handle rather sizable numbers of isotopes in order to explore many different kinds of fuel compositions. In this scheme reaction rates are determined by the use of modified Westcott cross sections, the cross sections being given by the usual expression:

$$\sigma(r, T) = \sigma_0 [g(T) + r s(T)]$$

where the first term on the right hand side arises from an average over a Maxwellian neutron energy distribution of temperature T and the second term arises from a $1/E$ slowing down spectrum whose magnitude is determined by the parameter " r ". In computing these cross sections, provision is made for self-shielding in the resonances by use of an expression for the effective resonance integral of the following form:

$$I_{\text{eff}} = \frac{I_{\infty}}{\sqrt{1 + \frac{NZ}{\Sigma_s + S/4V}}}$$

in which S is the surface area of the fuel region possibly reduced by a Dancoff factor and V is the volume and N is the isotopic concentration, and Z is a parameter which for an isotope with a single resonance would formally be the reaction cross section at the resonance energy. This formulation is useful for materials such as Pu^{240} where the resonance capture occurs primarily

in a single resonance. In the case of materials such as U^{238} , thorium, or their oxides, the value of Z is chosen so that the expression approximates the experimentally determined effective resonance integrals. The same type of expression is used in the low energy resonances of Pu^{239} and Pu^{241} and in addition a thermal flux disadvantage factor of semi-empirical form is introduced in the calculations. It is also possible to use for the temperature of the Maxwellian portion of the neutron spectrum a temperature higher than that of the moderator and determined by any one of a number of prescriptions. Arrangements are also made so that a fast fission factor can be introduced into the calculation. Results obtained from the use of this code, in studying plutonium recycle in various reactors, will be presented by Eschbach in another paper in this Topical Meeting.³ Recent studies on the performance of this code physics-wise and some resulting modifications will be discussed further below.

Puechl⁴ has used a somewhat similar approach except that his thermal cross sections were determined by calculations of a Wigner-Wilkins spectrum up to 0.625 ev and subtraction from this of an integral over a $1/E$ spectrum above 5 kT. He has handled the problem of resonance self-shielding by using an empirically determined expression for the lumped resonance integral of U^{238} . Self-shielding for plutonium isotopes is determined by scaling the effective integral for U^{238} up or down in proportion as the infinite dilution resonance integral of the plutonium isotope is higher or lower than that of U^{238} . A thermal disadvantage factor is used as obtained from a semi-empirical formula. In addition, a resonance flux depression factor is introduced by using different values of r for different isotopes.

A difficulty of the Westcott type of approximation is occasioned by the fact that the $1/E$ tail cannot be modified in shape to allow for the effects of resonance absorption. Since this $1/E$ tail supplies the neutrons that are captured in the system, the introduction of a strong resonance absorber requires an increase in the magnitude of this tail, which in turn leads to more resonance absorption, with the result that for sufficiently high concentrations of the absorber a divergence arises and the effective cross section becomes infinite. In Puechl's work this problem is handled by using only the absorptions up to 0.625 ev in determining the magnitude of the $1/E$ tail. In the case of MELEAGER a study was conducted⁵ in which reaction rates for homogeneous mixtures of fuel and water were computed by the use of the SPECTRUM V code.⁶ These computations used a Nelkin kernel and the calculation was carried up to 4.2 ev. MELEAGER was then modified so that r is now determined by an empirical fit to the SPECTRUM results. Table I gives a comparison of the ratios of the reaction rates of several isotopes as computed by the modified MELEAGER and by SPECTRUM V. In this same study determination was made of a prescription for the neutron temperature to be used in MELEAGER for a best fit to the spectrum results. This prescription is given as follows:

$$T_n = T_m \left(1 + 0.7 \frac{\sum_a^*}{\xi \sum_s^*} \right)$$

Where T_m is the moderator temperature, E_a^* is the total thermal absorption cross section divided by the thermal nonleakage probability, and $\xi \Sigma_s^*$ is the slowing down power of the moderator normalized to the fuel.

The results given in the table are obtained with the use of this temperature prescription as well as the modified handling of the epithermal portion of the spectrum.

A more sophisticated approach than either of these was taken by Greebler.^{7,8} In his approach the thermal disadvantage factor of the fuel was obtained from blackness theory and was energy dependent. Fuel cross sections corrected in this way were then used in a calculation of a Wilkins spectrum for the determination of thermal reaction rates. Resonance capture was handled by a 5-group method which included resonance self-shielding and Dancoff corrections. The results of Greebler's work pointed up especially the importance of a proper treatment of the thermal disadvantage factors in calculating the neutron spectrum and also the sensitivity of the spectrum in the region around 0.3 ev to the scattering kernels used in the spectrum calculation. It is just in this region that the lowest resonances of Pu²³⁹ and Pu²⁴¹ occur.

TABLE I
COMPARISON OF MODIFIED MELEAGER REACTION RATES WITH
SPECTRUM V REACTION RATES FOR VARYING ENRICHMENT
IN HOMOGENEOUS 260 C H₂O

Assumed Isotope Concentrations, grams/cc:

U ²³⁵	0.028	0.056	0.140	0.28	0.56
Pu ²³⁹	0.028	0.056	0.140	0.28	0.56
Pu ²⁴⁰	0.012	0.024	0.060	0.12	0.24

Computed Spectral Index:

Modified MELEAGER	0.1465	0.251	0.491	0.672	1.03
SPECTRUM V	0.1405	0.243	0.458	0.623	0.730

Computed Reaction Rate Ratio:

Pu²³⁹ to U²³⁵ per Atom Present

Modified MELEAGER	2.822	2.856	2.46	1.65	---
SPECTRUM V	2.902	3.124	3.39	3.35	2.97

Computed Reaction Rate Ratio:

Pu²⁴⁰ to Pu²³⁹ per Atom Present

Modified MELEAGER	0.717	0.883	1.339	2.15	---
SPECTRUM V	0.6534	0.782	1.052	1.88	1.736

Computed Pu²³⁹ Alpha:

Modified MELEAGER	0.548	0.561	0.584	0.581	---
SPECTRUM V	0.5515	0.565	0.581	0.581	0.5648

At Hanford Laboratories work is now in progress along two separate lines to improve the methods for carrying out such calculations. In one line, further modifications to MELEAGER are being planned.⁵ These consist in the computation of the thermal disadvantage factors by a P-3 method and in the determination of two-group constants from MELEAGER which can then be used in the examination of overall reactor effects such as zoned loadings or the occurrence of spectral variations due to the presence of a reflector. The second line of development is tackling the construction of an entirely new multi-group code for the handling of the problem.⁹ In this approach the GAM code¹⁰ is used to calculate effective cross sections in 68 energy groups extending from 0.682 ev to 10 Mev, including the affects of resonance self-shielding and Doppler broadening. The results of this calculation with 68 energy groups are then combined into eight groups to furnish the constants for the epithermal portion of a lattice cell calculation. Below 0.682 ev the problem is handled by TEMPEST,¹¹ a Fortran version of SOFOCATE, which uses a gas kernel model to calculate the neutron spectrum in this range. Flux disadvantage factors must be introduced from outside the code and incorporated into a homogenization process to furnish a composition upon which the spectrum calculation is based. Reaction rates are then computed from the nine-group cross sections and are used in carrying through a burnup step. After which, the new compositions can be used in connection with the nine-group cross sections to determine the reaction rates for the subsequent burnup step. It is intended that the GAM calculation be carried through only once in each burnup problem, based upon an average composition initially supplied to it. This code will be able to handle 15 fuel isotopes and 100 non-fuel isotopes in any particular calculation and will have a cross section library tape containing data for 40 fuel isotopes and 150 others, including fission products. The combined overall code has been given the name CALX.

NUCLEAR DATA

There is a voluminous literature on the low energy cross sections of the fissile nuclides. The information on U²³³, U²³⁵ and Pu²³⁹ was reviewed in 1961 by Leonard¹² and his review includes a bibliography of the original sources. Leonard concluded that the 2200 m/sec values of the cross sections and eta of Pu²³⁹ were somewhat less consistently known than those for the other two isotopes. This problem was recently reinvestigated by Sher and Felberbaum¹³ with the inclusion of data reported by Leonard, plus some additional information. The problem was treated by a least square analysis. Their best values were somewhat different from those obtained by Leonard and in particular the value of eta was lower by 0.07 from the value that he obtained. The differences arise primarily because of the inclusion of measured values of η by Sher. The values suggested by Leonard disregarded the measured η values as those most likely to suffer systematic error. The information on all of the isotopes of plutonium has recently been reviewed again by Leonard¹⁴ and he has again provided an extensive bibliography of original sources. Interested persons are referred to that review for details. We are reproducing in Table II the 2200 m/sec values for the various isotopes and have also included for Pu²³⁹ the values obtained by Sher.

TABLE II

2200 M/S CROSS SECTIONS OF THE PLUTONIUM ISOTOPES

Isotope	σ_t	σ_a	σ_f	σ_γ	ν	η	α
Pu ²³⁸			16.8 ± 0.3	403 ± 10			
Pu ²³⁹ (Leonard)	1018 ± 5	1008 ± 6	754 ± 9	254 ± 11	2.89 ± 0.05	2.16 ± 0.05	0.337 ± 0.017
Pu ²³⁹ (Sher)		1030 ± 7.4	748.2 ± 4.9		2.882 ± 0.016	2.093 ± 0.014	0.377 ± 0.011
Pu ²⁴⁰	283 ± 10	281 ± 10	0.030 ± 0.45	281 ± 10			
Pu ²⁴¹	1380 ± 15	1370 ± 15	1030 ± 8	340 ± 16	3.03 ± 0.05		
Pu ²⁴²		30 ± 2	<0.2	30 ± 2			

Resonance parameters are available¹⁴ for the 20 lowest resonances of Pu²³⁹, for 11 resonances of Pu²⁴⁰, and two resonances for Pu²⁴². For Pu²⁴¹ total cross section resonances have been clearly resolved up to about 20 ev, and measurements extend to about 2 kev at lower resolution. More recently measurements extending to 100 ev have been made at the MTR of the fission cross section of Pu²⁴¹ with greatly improved resolution.¹⁵ A multi-level analysis of the total and fission cross section measurements has been made up to 12 ev.

The dilute resonance fission integrals of Pu²³⁹ and Pu²⁴¹ have been measured by Hardy, et al.¹⁶ and values of the capture integrals of Pu²⁴⁰ and Pu²⁴² have been given by Harvey and Schwartz.¹⁷ Clark¹⁸ has calculated the integrals in the homogeneous case as a function of scattering cross section per plutonium atom. The experimental results are given in Table III while Clark's results are reproduced in Table IV.

TABLE III
MEASURED DILUTE RESONANCE
INTEGRALS OF PLUTONIUM

<u>Isotope</u>	<u>I-Fission (Barns)</u>	<u>I-Capture (Barns)</u>
Pu ²³⁹	327 ± 22(a)	545 ^(b)
Pu ²⁴⁰	---	8700 ± 800
Pu ²⁴¹	557 ± 33(a)	
Pu ²⁴²	---	1275 ± 30

(a) Above 0.5 ev.

(b) Obtained from I-fission and Clark's calculated value of α .

TABLE IV
RESONANCE INTEGRALS OF PLUTONIUM ISOTOPES

The integrals are over the range 0.625 to 9100 ev. Capture and fission integrals are denoted by I_c and I_f and their ratio by α . The scattering cross section per plutonium atom is denoted by σ_s .

$\text{Log } \sigma_s$	Pu^{239}		Pu^{240}	Pu^{241}		Pu^{242}
	I_f	α	I_c	I_f	α	I_c
2.5	85	0.41	77	55	0.3	19
3.5	117	0.52	86			29
4.5	151	0.57	134			58
5.5	186	0.60	258			86
6.5	220	0.62	460			140
7.5	249	0.64	815			250
8.5	268	0.66	1400			420
9.5	277	0.665	2350			630
10.5	281	0.67	3650			860
11.5	283	0.67	5290			1070
12.5	284	0.67	6680			1190
13.5			7490			1250
14.5			7900			1260
15.5			8040			1270
16.5			8120			1275
17.5			8140			

Data are available on abundances and periods of the delayed neutrons from Pu^{239} and Pu^{241} and these are reproduced in Table V. The fission product yields for fission of Pu^{239} differ in important respects from those of U^{235} . The yield of Xe^{135} is about 7.1 percent as compared to 6.4 percent for U^{235} fission. The yield for Pu^{241} is not well-known but is expected to be about the same as that for Pu^{239} .

The energy released in the fission of Pu^{239} is 5 percent greater than that in U^{235} and there is expected to be little difference between Pu^{241} and Pu^{239} in this regard.

TABLE V
ABUNDANCES AND PERIODS OF DELAYED NEUTRONS
FROM FISSION OF PLUTONIUM ISOTOPES

Nuclide Period	Abundances [(neutrons/fission) $\times 10^4$]			
	$\text{Pu}^{239} + n_{\text{th}}$ (a)	$\text{Pu}^{239} + n_{\text{f}}$ (a)	$\text{Pu}^{240} + n_{\text{f}}$ (a)	$\text{Pu}^{241} + n_{\text{th}}$ (b)
1	2.1 ± 0.6	2.4 ± 0.2	2.2 ± 0.3	1.54 ± 0.4
2	18.2 ± 2.3	17.6 ± 0.9	23.8 ± 1.6	36.5 ± 1.0
3	12.9 ± 3.0	13.6 ± 1.3	16.2 ± 0.4	27.5 ± 4.0
4	19.9 ± 2.2	20.7 ± 1.2	31.5 ± 2.7	62.0 ± 8.0
5	5.2 ± 1.8	6.5 ± 0.7	11.9 ± 1.8	29.0 ± 3.0
6	2.7 ± 1.0	2.2 ± 0.3	2.4 ± 0.5	
Total Yield	61 ± 3	63 ± 3	88 ± 6	154 ± 15

Period	Delayed Neutron Periods (sec)			
	$\text{Pu}^{239} + n_{\text{th}}$ (a)	$\text{Pu}^{239} + n_{\text{f}}$ (a)	$\text{Pu}^{240} + n_{\text{f}}$ (a)	$\text{Pu}^{241} + n_{\text{th}}$ (b)
1	54.28 ± 2.34	53.75 ± 0.95	53.56 ± 1.21	54.0 ± 1.0
2	23.04 ± 1.67	22.29 ± 0.36	22.14 ± 0.38	23.2 ± 0.5
3	5.60 ± 0.40	5.19 ± 0.12	5.14 ± 0.42	5.6 ± 0.6
4	2.13 ± 0.24	2.09 ± 0.08	2.08 ± 0.19	1.97 ± 0.1
5	0.618 ± 0.213	0.549 ± 0.049	0.511 ± 0.077	0.43 ± 0.04
6	0.257 ± 0.045	0.216 ± 0.017	0.172 ± 0.033	

(a) Keepin, G. R., T. F. Wimett and R. K. Zeigler, Reference 19.

(b) Cox, S. A., Reference 20.

INTEGRAL EXPERIMENTS

Some integral experiment results are now available for lattices of plutonium fuels in light water, heavy water, and graphite although they are not extensive. In the case of light water some comparison has been made with calculated k_{eff} obtained from the MELEAGER code and certain additional refinements needed in treating critical experiments. Comparisons in the case of the other two moderators are not yet complete.

Light Water

The experiments on light water lattices fueled with Pu-Al alloy rods conducted at the Hanford Laboratories^{21, 22, 23} were carried out with two plutonium concentrations in the alloy: 5 percent and 1.8 percent. The rods were of 1/2-inch diameter and were clad in Zircaloy-2. Lengths of the rods were 24 inches in the 5 percent case and 44 inches in the 1.8 percent case. The Pu²⁴⁰ content was low.

The experiments were carried out in a 4-foot diameter by 5-foot deep tank of light water. The lattices were hexagonal in shape and placed so as to provide effectively infinite water reflection on all sides. The lattice plates were constructed from lucite. Safety circuits, neutron sources, and BF₃ counters for monitoring the neutron flux were adapted from previous experiments of this type on 3.1 percent enriched uranium systems.²⁴

The neutron multiplication measurements were carried to within 96 percent of the critical mass as determined from the inverse multiplication plots. The critical number of rods, N_c , was found from a least squares fit to the plot of the number of rods divided by the counting rate (N/ϕ) versus the number of rods in the region between 85 to 96 percent of criticality. It has been shown that a straight line fit to this section of the plot is a very good approximation.

In order to conduct the exponential experiments, it was necessary to reduce the pile size from the near critical condition.

Cylindrical geometry was approximated in the hexagonal lattice using the equation,

$$R_{\text{cylinder}}^2 = \frac{NL^2 \sqrt{3}}{2\pi}$$

where

N = number of fuel rods, and

L = center to center fuel rod spacing.

The extrapolation length was determined by equating the buckling expression for the exponential measurements with the critical buckling as determined from neutron multiplication measurements,

$$B_{\text{critical buckling}}^2 = \left(\frac{2.4048}{R_{\text{critical size}} + \lambda} \right)^2 + \left(\frac{\pi}{h + 2\lambda} \right)^2 = \left(\frac{2.4048}{R_{\text{exponential}} + \lambda} \right)^2 - \left(\frac{1}{B_{11}} \right)^2$$

Results of the experiments are presented in Tables VI and VII.

TABLE VI

SUMMARY OF CRITICAL APPROACH AND EXPONENTIAL MEASUREMENTS
WITH Al-5 wt % Pu ALLOY RODS IN LIGHT WATER

Type of Experiment	Lattice Spacing (Inches)	H/Pu ^(a) (Atom Ratio)	H ₂ O/V Pu-Al Alloy	Extrapolation Length (λ)	Critical No. of 24-in. Rods (Cyl. Geometry)	Critical Mass ^(a) (Spherical Geometry)	Buckling (Meters ⁻²)
Experiment	0.75	217.8	1.17	8.10 cm ^(b)	356.1	3.31 Kg Pu	96.05
C. A. and Exp.	0.85	354.7	1.86	7.92 cm	230.2	2.07 Kg Pu	108.39
C. A. and Exp.	0.90	426.8	2.23	7.93 cm	192.0	1.70 Kg Pu	112.61
C. A. and Exp.	1.00	582.6	3.06	7.73 cm	170.1	1.53 Kg Pu	108.53
C. A. and Exp.	1.10	755.1	3.96	7.34 cm	166.5	1.53 Kg Pu	101.07
C. A. and Exp.	1.20	944.0	4.95	7.03 cm	181.1	1.73 Kg Pu	88.40
C. A. and Exp.	1.30	1149.3	6.03	6.04 cm	215.5	2.12 Kg Pu	76.81
Experiment	1.50	1609.2	8.44	5.80 cm ^(b)	307.8	3.10 Kg Pu	53.32
Experiment	1.80	2422.3	12.70	5.20 cm ^(b)			-1.97

(a) Total plutonium including all isotopes; the plutonium contains approximately 5 percent Pu²⁴⁰.

(b) Assumed values of λ extrapolated from curve.

For those cases in which the exponential experiment (Exp.) only was conducted the critical number of rods and critical mass were calculated from the measured buckling.

TABLE VII

SUMMARY OF CRITICAL APPROACH AND EXPONENTIAL MEASUREMENTS
WITH Al-1.8 wt % Pu ALLOY RODS IN LIGHT WATER

<u>Type of Experiment</u>	<u>Lattice Spacing (Inches)</u>	<u>H/Pu (Atom Ratio)</u>	<u>H₂O/Rod Volume Ratio</u>	<u>Extrapolation Length (λ) (cm)</u>	<u>Critical No. of 43.83 in. Rods (Cyl. Geometry)</u>	<u>Critical Mass (Spherical Geometry)(gm)</u>	<u>Buckling (Meters⁻²)</u>
C.A. and Exp.	0.75	630	0.9361	8.89 ± 0.05	563.4 ± 0.9	4011 ± 6.4	60.24 ± 0.20
C.A. and Exp.	0.80	810	1.203	8.26 ± 0.07	510.3 ± 0.9	3633 ± 6.4	61.27 ± 0.28
C.A. and Exp.	0.85	1001	1.487	7.56 ± 0.06	493.9 ± 1.0	3517 ± 7.1	60.09 ± 0.26
C.A. and Exp.	0.90	1204	1.788	7.96 ± 0.12	515.8 ± 1.0	3672 ± 7.1	52.72 ± 0.32
C.A. and Exp.	0.95	1418	2.106	7.07 ± 0.06	577.7 ± 1.2	4113 ± 8.5	47.34 ± 0.23

10.13
 HW-75007

The MELEAGER code has been used to calculate k_{eff} for the experimental arrangement using 5 percent Pu-Al fuel.²⁵ Since MELEAGER calculates k_{∞} an auxiliary calculation of the ages and thermal diffusion lengths was made and these were used with the measured bucklings to obtain the leakage. This was combined with the k_{∞} value to obtain k_{eff} . Some additional changes were also made in MELEAGER itself for the purposes of these calculations as follows:

- a) The neutron temperature was obtained from the Deutsch formula.²⁶

$$T_N = T_M \left(1 + 0.75 \frac{\Sigma_a (kT)}{\xi \Sigma_s} \right)^2$$

- b) A Bell correction²⁷ was introduced into the effective resonance integrals which were now obtained from the formula:

$$I_{\text{eff}} = \frac{I_{\infty}}{\sqrt{1 + \frac{N \sigma_{\text{max}}}{\Sigma_s + \tau_o}}}$$

where:

$$\tau_o = \frac{s_o \Sigma_M}{\Sigma_M + s_o \frac{V_o}{V_1}}$$

$$\Sigma_M = \text{moderator total cross section}$$

$$V_o/V_1 = \text{fuel lump to moderator volume ratio}$$

$$s_o = S/4V_o$$

- c) Thermal disadvantage factors were obtained from a diffusion cell calculation.
- d) Thermal leakage was included with absorption in calculating the Westcott "r".
- e) Variation in "r" due to reflector effects in these small assemblies was taken into account by a one dimensional, two-group calculation. The modified method of obtaining "r" by fit to a more detailed spectrum calculation, described above, was used as the base.

A three-group analysis using the SWAP code has also been carried out for both plutonium concentrations.²⁸ In this work the fuel cross sections for the thermal and epithermal groups were taken from the Westcott formation with resonance self-shielding and Bell corrections as in the preceding analysis. All other cross sections were obtained from APEX-515.²⁹ The thermal fuel

cross sections were taken with an upper limit of 3.681 kT and as above the Deutsch prescription was used to compute neutron temperature. A ν value of 2.91 was used. The cell was homogenized by a SWAP calculation to obtain volume and flux weighted values of the cross sections. Adjustments for spectral changes near the reflector were required. The results expressed in terms of k_{eff} are given in Table VIII along with the results of the MELEAGER based analysis.

TABLE VIII
CALCULATED k_{eff} FOR LIGHT WATER Pu-Al LATTICES

H/Pu	5 Percent Pu		1.8 Percent Pu	
	$k_{\text{eff}}^{(a)}$ (MELEAGER)	k_{eff} 3-Group	H/Pu	$k_{\text{eff}}^{(b)}$ 3-Group
218	0.979	1.019		
355	0.992	0.995		
427	0.987	0.979		
583	1.004	0.983	630	1.002
755	1.010	0.989	810	0.997
944	1.015	1.003	1001	0.993
1149	1.011	1.013	1204	0.992
1609	0.984	0.990	1418	0.994
2422	1.000	---		

(a) Errors on k_{eff} arising from errors in the measured bucklings are
~ 1/2 percent in the mid-range and larger at the extremes.

(b) Errors in k_{eff} arising from error in the measured bucklings are
~ 1/4 percent.

It should be noted in this connection that there is a considerable body of data available on criticality of aqueous solutions of plutonium nitrate^{30, 31} which may be used as a check on computational methods. The data are available in terms of spherical critical masses. However, arrangements are being made to obtain spectral indices such as Pu239/U235 fission ratios, lutetium activations, cadmium ratios, etc., in the near future at the Hanford Laboratories, Plutonium Critical Mass Laboratory; and we understand that detailed spectral measurements with a chopper are being planned on solution assemblies in the UK.³²

Heavy Water

At the time of the initial loading of the PRTR the critical size of a lattice of 19-rod Pu-Al clusters on 8-inch centers in D₂O was determined from sub-critical extrapolations.³³ These fuel elements were composed of Pu-Al rods containing 1.8 percent plutonium of one-half diameter clad in Zircaloy-2. The loadings were essentially cylindrical and five different heights were obtained by varying the moderator level. Table IX presents the critical sizes obtained.

TABLE IX
EXTRAPOLATED CRITICALS FOR Pu-Al
CLUSTERS IN D₂O

<u>Moderator Level</u> <u>(Inches)</u>	<u>Critical Number</u> <u>of Clusters</u>
55	19 ± 2
75	14 ± 1
83	13.6 ± 0.5
95	12.2 ± 0.2
109	11.9 ± 0.1

Triplett and Peterson³⁴ have analyzed this data to obtain a value for the critical buckling of

$$B^2 = 14.9 \pm 0.3 \text{ meters}^{-2}.$$

Graphite

Experiments have been done in the PCTR on reactor lattices fueled with 19-rod clusters of 0.5-inch diameter rods of low exposure Pu-Al.³⁵ The lattices were graphite moderated and air cooled. Measurements were made for lattice spacings of 6.500, 8.375, and 10.500 inches, all with a square pitch.

The mass of copper required to poison each lattice to unit multiplication was determined. Foil activation data were obtained at various positions throughout the lattices with gold, copper, lutetium, U²³⁵-Al, and Pu-Al detectors. Ratios of fission rates of Pu²³⁹ and U²³⁵ in the lattice spectra were measured relative to the ratio of the same fission rates in a Maxwellian spectrum.

The analysis of the data to obtain traditional reactivity parameters: k_{∞} , f , p , encountered difficulties because of the grossly non- $1/v$ character of the Pu²³⁹ and Pu²⁴¹ cross sections in and very near the thermal region.³⁶

This gives rise to distortions of the thermal and near-thermal flux distributions which are difficult to calculate or monitor. The distortions cannot be monitored because neutron capture in Pu^{239} or Pu^{241} quickly leads to Pu^{240} or Pu^{242} in the ground state. Pu^{239} fission cannot be relied upon because of the rather rapid changes in $\eta^{49}(E)$ with energy E . The unpoisoned lattices have very high neutron leakage rates. It became apparent during the first measurements that these high leakage rates caused sizeable changes in the fast-to-slow flux ratios as given by the cadmium ratios, and therefore, a sizeable change in values of η . Under these circumstances it is obvious that our previous interpretations of PCTR data, and probably the use of k_∞ becomes difficult.

Another difficulty arises because the neutron capture rate in Pu^{240} and Pu^{242} cannot be monitored. Capture in Pu^{240} leads quickly to the ground state of Pu^{241} and capture in Pu^{242} leads to a five hour activity (of Pu^{243}) which is impossible to count in the background of fission product gammas unless rather pure samples of Pu^{242} were available.

Finally, the location of the 0.3 ev resonances in Pu^{239} and Pu^{241} at the joining region of the thermal and epithermal spectra, but below the cadmium cutoff, makes it difficult to describe absorption in the resonance as either a thermal or a resonance absorption process.

The data may, however, be used as a test of computer codes by noting how closely the code gives the experimental result in the poisoned lattice as expressed by

$$k_\infty = \frac{\int_0^R \int_0^\infty \nu \Sigma_f(E) \varphi(E) dE dr}{\int_0^R \int_0^\infty \Sigma_a(E) \varphi(E) dE dr} = 1$$

Additional tests of the codes may be obtained by calculation of activation ratios of the foils of various materials inserted into the test lattice. The reader is referred to the references cited for details.

The lumped resonance integral of Pu^{240} in Pu-Al rods was measured in graphite moderator in the PCTR.³⁷ The results were obtained by a step-wise comparison of pairs of rods which differed only in Pu^{240} content. Eight such pairs were made up such that the rod with the more Pu^{240} in one pair had the same content as the rod with the less Pu^{240} in the next pair. To calibrate the method the resonance integral for the four lowest Pu^{240} concentrations was calculated. Results are presented in Table X.

TABLE X

LUMPED RESONANCE INTEGRALS OF Pu²⁴⁰

<u>Pu²⁴⁰ Concentration</u> <u>(10¹⁸ Atoms/cc)</u>	<u>Resonance Integral</u> <u>(Barns)</u>
0.050	8612(a)
0.142	8572(a)
0.407	8466(a)
1.17	8203(a)
3.35	7068 ± 364
9.59	5347 ± 378
27.41	3482 ± 272
78.05	2096 ± 172
219.6	1237 ± 106

(a) Calculated value.

A measurement of the reactivity temperature coefficient of Pu-Al alloy fuel has been made with plutonium containing 6 percent Pu²⁴⁰ and with plutonium containing 16 percent Pu²⁴⁰.³⁸ The plutonium composition of the alloy in the two cases was adjusted to obtain approximately the same Pu²³⁹ content in both sets of fuel. The fuel was in the form of 19-rod clusters of Pu-Al alloy containing approximately 2 percent plutonium and of 1/2-inch rod diameter. Measurements were conducted in the Physical Constants Testing Reactor (PCTR) over a temperature range from 20 C to above 400 C. Techniques used were similar to those previously used with uranium fuels.³⁹ Electrical heating was used. The composition of the two types of fuel was as follows:

<u>Isotope</u>	<u>Atom Percent (LX)</u>	<u>Atom Percent (HX)</u>
Pu ²³⁹	93.434	80.727
Pu ²⁴⁰	6.062 ± 0.005	15.944 ± 0.011
Pu ²⁴¹	0.448 ± 0.001	2.828 ± 0.004
Pu ²⁴²	0.0159 ± 0.001	0.501 ± 0.001
wt % Pu in Al	1.878	2.078

A measurement was also made with dummy aluminum rods to enable correction for the positive reactivity effect caused by the increase of neutron temperature due to scattering from the heated aluminum. The low Pu²⁴⁰ fuel yielded a negative coefficient; the positive effect of the aluminum being overcome by the Doppler effect in plutonium. With the higher Pu²⁴⁰ fuel a more

negative coefficient was obtained probably due to the Doppler broadening of the Pu^{240} resonance. The temperature coefficients:

$$1/k_{\infty} \frac{dk_{\infty}}{dT}, \text{ are}$$

6 percent Pu^{240} fuel	$-0.7 \pm 0.3 \times 10^{-5}/^{\circ}\text{C}$
16 percent Pu^{240} fuel	$-1.4 \pm 0.3 \times 10^{-5}/^{\circ}\text{C}.$

Burnup Experiments

While data on the neutron economy of various lattices under static conditions is essential to test computation methods and determine the correctness and completeness of our understanding of the physical effects involved, the final definitive tests will be given by experiments in which the changes in isotopic composition resulting from the operation of fuels in reactors are determined.

The first of a series of such experiments is now in progress in the PRTR. Fuel elements of a selected set of Pu-Al alloy fuels provided with flux monitoring devices are being carried to six different exposure levels, the highest being 50 percent burnup of the original Pu^{239} . A similar experiment is in progress with natural UO_2 fuels. Similar experiments with mixed oxide fuels are planned.

A burnup experiment in light water which will involve loading at least a substantial fraction of the core of the EBWR at Argonne with mixed oxide fuel is in the planning stages.

A satisfactory understanding of the physics of plutonium as a thermal reactor fuel will only be achieved by continuing to push forward in the four areas of code development, basic data, zero power integral experiments, and burnup experiments.

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EXPERIENCE WITH THE PLUTONIUM RECYCLE TEST REACTOR

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INTRODUCTION

The Plutonium Recycle Test Reactor (PRTR), located at the Hanford Laboratories, is an experimental facility designed to support the technical development of plutonium fuel cycles in thermal power reactors. The reactor design is a compromise aimed at providing a versatile test facility with prototypical power reactor operating conditions, in which to demonstrate the feasibility of recycling plutonium fuels. The reactor first went critical in November, 1960, and reached design power in July, 1961. It has operated since that time in the intermittent manner typical of a test reactor.

REACTOR DESCRIPTION¹

Figure 1 shows a cut-away view of the PRTR. The reactor is moderated and cooled with D_2O , and produces 70 megawatts of thermal energy at design power. Eighty-five vertical pressure tubes, made of Zircaloy-2, pass through the aluminum calandria that contains the moderator. The pressure tubes, which are located in an 8-inch triangular lattice, contain the fuel elements and the primary coolant at 1050 psig and 530 F outlet temperature. The moderator is thermally insulated from the pressure tubes by means of shroud tubes and a helium gas gap. A radial reflector of D_2O surrounds the calandria.

Reactor control is achieved by raising or lowering the moderator level through use of a gas balance system. This system provides an excellent vernier control for normal operation and also provides a rapid shutdown mechanism in that the moderator is dumped from the core in case of a scram.

Additional reactivity control is provided through the use of 36 "gray" shim rods. These rods provide the necessary compensation for fission-product buildup and fuel depletion during an operating cycle.

REACTOR FUEL

The basic fuel element design for the PRTR is a 19-rod cluster of 0.5-inch diameter fuel cores that are clad in Zircaloy. Fuel core materials which have been used are natural UO_2 , plutonium-aluminum alloy, and natural UO_2 enriched with PuO_2 . Many other fuel materials and designs will be utilized on a test basis during the course of the fuels development program. Table I illustrates the variety of special fuel elements that have been irradiated in PRTR during the past year, in addition to the standard elements of cold swaged natural UO_2 and low Pu^{240} plutonium-aluminum alloy (Lx Pu-Al).

FIGURE 1
PRTR Cutaway View

TABLE I
SPECIAL TEST FUEL ELEMENTS

<u>Fuel Type</u>	<u>Fabrication Method</u>	<u>Fuel Geometry</u>	<u>Number of Elements</u>	<u>Average Exposure (September 1, 1962)</u>
UO ₂	Vipac	Tubular	1	1338 Mwd/T _U
UO ₂	Vipac	19-Rod	1	1129 Mwd/T _U
UO ₂	Hot Swage	19-Rod	1	1010 Mwd/T _U
Pu-Al(HX) ^(a)	Extrusion	19-Rod	3	34.8 Mwd
UO ₂ -PuO ₂ (HX)	Cold Swage	19-Rod	2	16.6 Mwd
UO ₂ -PuO ₂ (HX)	Vipac	19-Rod	2	17.0 Mwd
MgO-PuO ₂ (HX)	Cold Swage	19-Rod	1	7.5 Mwd

Maximum Exposure to Date - UO₂ Element - 2650 Mwd/T_U

Total Reactor Exposure to Date - 8550 Mwd

(a) (HX) = high exposure plutonium, 10 to 17 atomic percent Pu²⁴⁰

REACTOR FUELING HISTORY

The initial reactor fuel loading for power operation, illustrated in Figure 2, consisted of a central region of 25 natural UO₂ elements, an annular enrichment ring of 30 plutonium-aluminum elements that contained 1.8 weight percent plutonium in the alloy, and an outer annulus of 30 additional natural UO₂ elements. As the average exposure of the fuel increased, additional plutonium-aluminum elements were charged in the enrichment ring to provide needed reactivity, increasing the total number to 39. Considerable flexibility in fuel scheduling and in attainable power distributions has been possible since all fuel elements are physically interchangeable. A satisfactory balance between necessary excess reactivity and desirable power distributions has been achieved by "outside-in" refueling. In this method, fresh enrichment elements are charged around the outer periphery of the enrichment ring and the irradiated elements are moved inward as their exposure increases until they are finally discharged at a nominal goal exposure of 75 Mwd/element. This technique improves the power flattening across the core since the fission cross section of the plutonium-aluminum elements decreases steadily with exposure. Figure 3 illustrates the relative power distribution produced by the initial core configuration and several subsequent distributions that were produced during the transition from a "green" core to a core that had a uniformly-graded exposure on the enrichment fuel. The plotted data are the observed ratios of the average tube power in each of the nine concentric rings of tubes in the core to the average tube power of the reactor. The band beneath each power distribution curve illustrates the

FIGURE 2
PRTR Core Configuration
Startup Loading

FIGURE 3
Relative Power Distributions

fuel composition in the various rings of tubes. The numbers in the composition bands represent the average fuel exposure in the ring whose relative power is plotted directly above.

The reactivity worth and the heat output of the uranium oxide fuel underwent relatively small changes during this transition period. The buildup of Pu^{239} in the UO_2 fuel is sufficient to compensate for the burnup of U^{235} , for exposures up to about 2500 Mwd/ T_U . Therefore, it was possible to attribute most of the changes which were observed to occur in several of the reactor parameters to burnup of the plutonium-aluminum fuel.

One of the parameters that changed, as the exposure of the enrichment fuel increased, was the rate of reactivity burnup. This quantity is a significant factor in determining the length of an operating cycle. Figure 4 shows how the observed rate of reactivity burnup has varied as the fraction of fissions occurring in plutonium increased. The first seven points are from the three-zone loadings of UO_2 and Pu-Al. The last three points are from later loadings that contained increasing numbers of mixed-oxide fuel elements. Shown also, for comparison, are the results of three-group diffusion theory perturbation calculations of the rate of reactivity burnup.

The addition of the mixed-oxide fuel to the core has made an obvious and significant improvement in the rate of reactivity burnup and makes possible operating cycles of increased length. The reason for this improvement can be seen in Figure 5, which presents the results of Meleager's² calculations of k_∞ for cells containing the three basic PRTR fuel types. The increased useful reactivity lifetime of the mixed-oxide fuel is quite apparent.

REACTOR KINETICS

The increase in the fraction of fissions occurring in plutonium affected another important reactor parameter, $\bar{\beta}$, the effective delayed neutron yield fraction. This quantity governs the speed of response of the reactor to changes in multiplication and thereby largely determines the minimum response time required of the reactor's control system. Because β for Pu^{239} is only about one-third as large as β for U^{235} , the value of $\bar{\beta}$ for the reactor tends to decrease as a larger percentage of the total fissions take place in plutonium. Figure 6 presents the results of calculations of $\bar{\beta}$ for the range of plutonium fission fractions that have been encountered in the PRTR so far. The delayed yield fractions for U^{235} and Pu^{239} reported by Keepin, et al.³ and the γ -n yield fractions reported by Bernstein,⁴ renormalized to Keepin's results, were used in the calculations. An effectiveness factor of 0.2 for the transmission of γ -rays into the D_2O was assumed. A systematic error, believed to be less than 2 percent, has been introduced into the calculations by neglecting the difference in the age-to-thermal of the delayed groups compared with the prompt groups.

FIGURE 4

Reactivity Burnup Per Mwd Reactor Operation Versus
Fraction of Total Fissions Occurring in Plutonium

FIGURE 5

Cell Multiplication as a Function of Exposure

FIGURE 6

Effective Delayed Neutron Fraction Versus Fraction of Total Fissions Occurring in Plutonium

The calculated delayed fraction that would be characteristic of the reactor without D_2O is also shown in Figure 6. It can be seen that the γ -n reaction in D_2O causes a significant increase in the effective delayed fraction and thereby eases the problem of control system speed in a plutonium-fuel reactor.

Also shown in Figure 6 are the preliminary results⁵ of several measurements in the PRTR of the ratio $\bar{\beta}/l^*$, where l^* is the prompt neutron lifetime in the reactor. These measurements employed the technique of analyzing the frequency spectrum of the reactor noise.⁶ Experimental difficulties, due chiefly to moderator level fluctuations, have made interpretation of the data very difficult and these results are included only to indicate the qualitative agreement with calculation.

TEMPERATURE COEFFICIENTS

The observed temperature coefficients of reactivity^{7, 8} associated with the fuel plus coolant system and with the moderator system are presented in Table II. It can be seen that both coefficients have decreased in magnitude as the core composition changed from the initial loading to the graded-exposure loading.

TABLE II
TEMPERATURE COEFFICIENTS OF REACTIVITY

<u>Reactor Parameter</u>	<u>Initial Loading</u>	<u>Graded-Exposure Loading</u>
Fuel plus coolant coefficient	$-3.5 \times 10^{-5} \frac{\Delta k}{k}/C$	$-1.7 \times 10^{-5} \frac{\Delta k}{k}/C$
Moderator Coefficient	$-5.3 \times 10^{-5} \frac{\Delta k}{k}/C$	$-4.3 \times 10^{-5} \frac{\Delta k}{k}/C$

The large change in the fuel plus coolant coefficient can be attributed to several causes:

1. The removal of U^{238} from a region of high statistical weight and the reduction in the total amount of U^{238} in the core due to the replacement of UO_2 elements with plutonium-aluminum elements; and
2. The degradation of the primary coolant from 99.25% D_2O to about 95% D_2O .

The smaller decrease in the moderator coefficient can be attributed to a decreased migration area caused by the increased amount of plutonium in the core and by the buildup of fission products in the fuel.

It is expected that the negative fuel plus coolant coefficient will increase in magnitude during the coming months as the plutonium-aluminum fuel elements are replaced with mixed-oxide elements.

CONCLUSIONS

The first year's operation of the PRTR has demonstrated that substantial quantities of plutonium can be utilized as fuel in a thermal reactor conditions, together with natural uranium, under power reactor conditions, without creating any undesirable or unsafe control characteristics.

The plutonium-aluminum spike enrichment elements, as presently constituted, have relatively short reactivity lifetimes and therefore have caused short operating cycles and a high through-put of elements. The conversion to mixed-oxide fuel, presently under way, has shown promise of increasing the reactivity lifetime of the elements by a factor of three or four. This increased lifetime will be reflected directly in longer operating cycles and reduced through-put of elements.

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A SURVEY OF THE ECONOMICS OF PLUTONIUM AS A FUEL IN THERMAL REACTORS

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Comprehensive, but not detailed, survey studies of the utilization of plutonium in thermal reactors are nearing completion as part of the Hanford Laboratories Plutonium Recycle Program included in the US-AEC Contract AT(45-1)-1350 with the General Electric Company. More specific studies are under way which will be supplemented with experimental data as they become available.

The survey results indicate that the value of plutonium in thermal reactors is greater than earlier, less sophisticated studies have predicted. Furthermore, several interesting special applications of plutonium have been identified.

The studies include the following major facets:

1. Successive recycle of plutonium with the maximum quantity of plutonium recycled being limited to the quantity produced in the previous cycle.
2. Successive recycle of plutonium without any limitations as to the maximum quantity of plutonium recycled.
3. Plutonium of fixed isotopic composition used as fuel in a broad variety of reactors.
4. Reuse of fuel between fast and thermal reactors with and without intermediary separations steps.
5. Plutonium enrichment of thorium and U^{233} enrichment of U^{238} , i. e., "crossed progeny".
6. Economic optimum fertile fuel density, especially when large amounts of Pu^{240} are involved.
7. Special burner cycles involving only Pu^{240} as the fertile fuel.
8. Multidimensional studies of the relative importance of alpha and cross sections of bred fuels.
9. Multidimensional studies of the importance of reactor parameters, particularly lattice spacing, parasitic absorption, and neutron leakage.

Uranium Price Schedule Influence

A significant, but unappreciated, asset of plutonium is that it is chemically separable from U^{238} , while U^{235} is not. This, coupled with the structure of the uranium cascade price schedule, leads to the rather surprising

result that regardless of the enrichment level or the degree of fissile burn-up, the alternative value of plutonium as U^{238} enrichment is proportional to the cost of fully enriched U^{235} and not to the cost of U^{235} burnup. For plutonium to recover the enrichment cost, it is only necessary that the plutonium be mixed with uranium whose U^{235} content is at least that of the optimum tails composition and that a chemical separations step be performed to recover the spent fuel.

A study to be published as HW-72219 considers many combinations of price schedules and fueling systems. Included are the results of detailed plutonium value calculations with different uranium price schedules as shown in Table I. The constant proportionality of plutonium value to the cost of fully enriched uranium is apparent. Computed U^{233} values exhibit the same characteristics.

TABLE I
COMPUTED VALUES OF PLUTONIUM AND U^{233} IN THE APWR
FOR THREE DIFFERENT URANIUM PRICE SCHEDULES
(Values Calculated by the Successive Recycle
Method Described in HW-72217)

AEC Uranium Price Schedule	Price of 90% Enriched U \$/gram U^{235}	U^{238} Fertile			Thorium ²³² Fertile		
		Fuel Cost mills/kwh _e	Corresponding Value of Pu \$/gr (Fissile)	Ratio of Pu Value to Price of 90% Enriched U	Fuel Cost mills/kwh _e	Corresponding Value of U^{233} \$/gram	Ratio Of U^{233} Value To Price Of 90% Enriched U
Pre-July 1, 1961 ⁽¹⁾	17.04	2.76	17.34	1.018	3.38	19.50	1.144
Post-July 1, 1961 ⁽²⁾	13.65	2.31	13.95	1.022	2.88	15.67	1.148
Post-July 1, 1962 ⁽³⁾	12.01	2.15	12.14	1.011	2.63	13.87	1.155

	Cost of Separative Duty, \$/kg	Cost of Natural Uranium Feed, \$/kg	U^{235} Composition of Optimum Tails Weight Fraction in U^{238}	Minimum Priced U^{235} Composition Weight Fraction in U^{238}
(1)	37.29	39.27	0.002214	0.00340
(2)	37.29	23.50	0.002770	0.00403
(3)	30.00	23.50	0.002531	0.00380

Plutonium Concentration and Alpha Values

Comparison of plutonium and U^{235} fueling systems is complicated by the unique low energy plutonium resonances. The 0.3 ev Pu^{239} and the 1.0 ev Pu^{240} resonances are particularly important. The Pu^{239} resonance is predominantly a capture resonance and the corresponding alpha value is

approximately 0.75, and the value of η is 1.67; while the α value for 2200 meter per second neutrons is about 0.39 and the value of η is 2.08²-the same as for U²³⁵. It would appear that as the thermal flux is hardened, or elevated in temperature, the effective α value for Pu²³⁹ would increase well beyond the 2200 meter figure, and the associated fuel value would decrease. It is of extreme importance that the resonance self-shields so that as the concentration of plutonium is increased, the 0.3 resonance absorptions tend to decrease per atom of Pu²³⁹ present. This effect is often encountered, but it is usually overshadowed by the corresponding spectral hardening due to the increased blackness of the fuel as the plutonium concentration is increased. This effect is clearly shown by considering addition of increasing amounts of plutonium to a lattice cell already well hardened by $1/v$ absorber. The resulting fluxes are illustrated in Figure 1 in which the relative decreased flux in the resonance region is apparent as the concentration is increased. From this, one would suspect that irradiation of a small concentration of plutonium in a relatively hard flux would be less productive than irradiation of a significantly greater amount. Results of such computations are shown in Figure 2 for two computer models. The upper curve is for the simpler model which considers the reactor to be homogeneous, with the fuel and moderator intimately mixed. The lower curve is from a more realistic model which considers the reactor to be heterogeneous with lumped fuel in a moderator medium. The increased self-shielding of the heterogeneous case is apparent.

FIGURE 1

Neutron Flux Spectrum for Different Pu²³⁹ Densities

*Recent analysis of data by B. R. Leonard, Hanford Laboratories, indicate a higher value of η for Pu²³⁹ than the 2.08 used here.

$\alpha, (\alpha = \bar{\sigma}_c / \bar{\sigma}_f)$ Determined with Flux Weighted Cross Sections
Computed for 75 Energy Groups

FIGURE 2

Pu^{239} Alpha as a Function of Concentration of Pu^{239} Comparing Homogeneous and Heterogeneous Loading

As indicated in Dr. Gast's paper, the plutonium cross sections in the MELEAGER computer code are calibrated against the upper curve which is for the homogeneous case. the MELEAGER code is adjusted to have higher values for Pu^{239} 's alpha than is shown by the upper curve of Figure 2 and, therefore, is conservative.

The possibility of lowering the alpha of Pu^{239} by the amounts shown in the figure is intriguing. However, it should be noted that large concentrations are involved. By further concentrating the fuel in heterogeneous reactors, the same result can be achieved with substantially less plutonium. This is indicated in Figure 3 for which a more detailed computer model allows comparison of the alpha values for (1) U^{238} uniformly enriched with plutonium, (2) the same amount of plutonium concentrated on the surface of the fuel, and (3) this plutonium concentrated in the center. The alpha value of 0.532 for Pu^{239} concentration in this heterogeneous case is significantly lower than the value of 0.575 for the corresponding homogeneous case in Figure 2. It is for this reason that the MELEAGER code is actually doubly

conservative since its alpha values are greater than the homogeneous case, and MELEAGER's value of alpha for Pu²³⁹ is about 0.59 for the concentration corresponding to Figure 3. It is of significance that the alpha value of plutonium is reduced further by placing the plutonium in the central rod as shown on Figure 3. This indicates that the in situ burning of plutonium as it is formed in an enriched U²³⁸ fueled reactor is not the most favorable with respect to minimizing the alpha of plutonium. A larger amount of plutonium concentrated in central rods gains significantly more shielding, and results for other concentrations await computations which involve transport calculations of self-shielding factors and slowing down diffusion calculations of reaction rates. The self-shielding factors are determined by Doppler broadened cross sections in fine energy detail.

PLUTONIUM α^* FOR THREE GEOMETRICAL CONFIGURATIONS

$$*\alpha = \frac{\sigma_c}{\sigma_f}$$

Plutonium Composition 95% Pu-239, 5% Pu-240

1. Mixed Fuel Case

Plutonium Distributed Uniformly in the Fuel Region - 2% Enrichment of Uranium

Plutonium 239 $\alpha = .5323$

Total Pu $\alpha = .6463$

2. Annulus

Plutonium From Case 1 Concentrated in a Thin Annulus Surrounding the Fuel Region

Plutonium 239 $\alpha = .5323$

Total Plutonium $\alpha = .6463$

3. Central Rod

Plutonium From Case 1 Concentrated in a Slender Rod in the Center of the Fuel Region

Plutonium 239 $\alpha = .5120$

Total Plutonium $\alpha = .5795$

FIGURE 3

The reduction of alpha for Pu^{239} is accompanied, of course, by reduction in its absorption cross section. The Pu^{240} cross section is reduced even more, as indicated in Figure 3, by the greater reduction in alpha for the plutonium batch than for the Pu^{239} alone. While adjustment of the effective alpha value of Pu^{239} by concentration is appealing, the increased inventory charges and the reductions of cross section must be balanced against the gains in neutrons and heat. Sensitivity to inventory charges is also introduced with plutonium fueling because of the fertility of Pu^{240} . In essence, a fertile absorption capacity is added above that of the U^{238} in the fuel as one increases its plutonium enrichment because of the high effective Pu^{240} cross section. Thus enrichment to a higher level to attain a given k may be necessary. Of course, an associated bonus of longer exposure may prevail, but it still must be weighed against the additional inventory charges.

Plutonium Value Determination

In the course of our studies of plutonium value, we have considered many formulations to determine the value. It is often particularly tempting to choose a highly simplified formulation when one is interested in the impact of only one variable--particularly an economic variable--as these always seem less important to the scientist or engineer than a nuclear process factor. We have found that the results of such brief analyses are often inconsistent with results of other simplified analyses as well as with the results of analyses with more elaborate models. The differences usually stem from incomplete knowledge of the mutual interdependence of economics and the nuclear process. Much of this is eliminated by adhering to comparison of costs at operating points based upon minimized fuel costs, and to ascertaining values considering clean sale and purchase of materials in marketable form to and from second and third parties. While this methodical approach appears tedious and may be irritating to the physical scientist, we are convinced it assists us in maintaining a sound business logic.

The cost coding system used by HLO is named QUICK and is described in HW-71812. This code is used to indicate directly the impact upon fuel costs of using plutonium valued at some fixed price. Solutions for theoretical market prices for plutonium yielding the same fuel cost as that achieved by an alternative standard reactor fueling scheme can be made for a limited number of alternatives by coupling adjoint solutions of QUICK to PUVÉ, a value analysis code described in HW-71811. In practice, PUVÉ is limited to consideration of successive recycle and cannot consider alternative fueling interchange of fuel between reactors having different nuclear properties. To treat this larger problem, a limited number of typical plutonium compositions will be considered as fuel for variously sized reactor groups having different nuclear properties. By considering the rate at which stockpiles and inventories should be accrued, it is hoped that a manageable solution method can be developed to yield reasonably accurate results.

Plutonium Fuel Fabrication Costs

Common to all plutonium fueling systems is the possible complication of its toxicity. Because of this toxicity, plutonium requires special handling and higher costs may result for fuel element fabrication and jacketing costs for plutonium than for enriched uranium. The magnitude of the incremental cost so incurred is uncertain at present, but is calculated for reasonable production quantities to be no greater than \$5 to \$10 per pound of U^{238} contained in the fuel for fuel element designs amenable to remote fabrication. The higher the enrichment of plutonium in the fuel, the lower is the incremental cost per gram of plutonium involved. Thus, if three percent fissile plutonium enrichment of U^{238} is being used, the increment amounts to from \$0.50 to \$1.00 per gram of fissile plutonium; but if 0.3 percent fissile plutonium enrichment is involved, the incremental cost amounts to from \$5 to \$10 per gram of fissile plutonium for fuel element fabrication and jacketing cost increments of \$5 to \$10 per pound of uranium. In instances of plutonium recycle wherein the amount of plutonium is limited to the plutonium available from the previous cycle, the average plutonium enrichment level of the reactor is in the range of 0.3 to 0.5 percent. However, if the reactor loading is zoned and plutonium enrichment is confined to depleted uranium, enrichment levels of 2 to 3 percent are typical and an incremental fabrication charge as high as \$20 per pound of fuel is spread to from \$1 to \$2 per gram of plutonium. Thus, zoning of the reactor makes the problem of incremental fabrication costs upon plutonium value one of minor concern if these costs are below \$15 per pound of uranium jacketed. This is shown in Figure 4. It should be recognized that there is a slight cost of zoning plutonium because of the nonlinearity of the uranium price schedule, and if the incremental fuel element fabricating and jacketing cost were zero, zoning would be undesirable. However, zoning of plutonium has virtues in its own right in that buildup of Pu^{242} in the freshly recycled plutonium can be minimized by sale or even discard of the highly burned material. The flexibility of zoned plutonium loads with and without U^{235} -enriched zones allow maximization of many other important facets, among which are reactor power flattening and adjustment of flux peaking. By virtue of the high cross sections of plutonium, substantial power flattening can be achieved without increases in leakage that could override the productivity of power flattening.

FIGURE 4

The Value of Plutonium as Affected by the Incremental Fuel Element Fabricating and Jacketing Cost for Plutonium Bearing Fuels

Fuel Survey Codes

Much of our activity has been devoted toward development of reactor physics and economics codes that are sufficiently rapid to enable survey of the possible systems and still be sufficiently accurate to take into account the pertinent factors as to direction and relative magnitude. To investigate the more promising systems identified by the survey, several successively more rigorous stages of investigation are planned.

The rapid surveys are performed with two major code systems, each developed around the MELEAGER burn-up code and a standard computer linkage routine known as CHAIN that couples separate computer

codes. These codes are prosaically named RECYCLE CHAIN and COMBINED CYCLE CHAIN. The CHAIN calls each code as needed into the computer's rapid access memory from standby magnetic tapes.

The current RECYCLE MELEAGER CHAIN sequence consists of the following major programs:

MELEAGER to supply the reactor fuel burn-up data ("physics")

QUICK to supply the cost data by operating on MELEAGER results

MINIMIZER to determine the minimum fuel cost

PUVE to establish the plutonium value resulting from an appropriate recycle sequence.

The plutonium recycle sequence considered in this model starts with the uranium enrichment of the reactor. The plutonium ashes from this first step are used to re-enrich the reactor for the next cycle and so on with further cycles until enough data can be developed to solve for the plutonium values. Due to the changes in isotopic composition with irradiation, the ash values are generally not equal for all cycles and PUVE takes this into detailed account.

The plutonium values resulting from PUVE are then compared with the estimated prices originally assigned for each step in the RECYCLE CHAIN sequence so that the minimum cost points can be selected. If too much difference exists, new plutonium price estimates are prepared and the chain sequence is rerun until a sufficiently small difference is achieved.

Results RECYCLE CHAIN Studies

The types of reactors considered are shown below:

APWR - Simulated Advanced Pressurized Water Reactor
(Based upon Report TID-8502)

BWR - Simulated Boiling Water Reactor
(A generalized description, with zirconium jacketing
but not of specific design)

OMR - Simulated Organic Moderated Reactor
(Based upon Report TID-8501)

HWR - Simulated Deuterium Oxide Moderated Reactor
(With zirconium tubes, but not a specific design)

GCR - Simulated Gas Cooled Graphite Reactor
(A low temperature moderator type patterned
after "Bradwell").

A number of fueling strategies and economic climates were studied for these five reactors. As indicated, two general successive recycle schemes have been studied for plutonium. One considers a classical single reactor recycle in which the amount of plutonium available for recycle is limited to the amount produced in the previous cycle (with supplementary

U^{235} enrichment as needed) and the other considers a hypothetical stockpile from which plutonium of the same isotopic composition as the previous cycle can be purchased so that no limitations exist as to the amount of plutonium available to enrich natural uranium for the next cycle.

The general decrease in bred fuel value as successive recycle proceeds is typically demonstrated in Figure 5 for the two general recycle schemes and for U^{233} recycle with U^{235} makeup. The fuel costs associated with each of these systems are shown adjacent to the curves. Notice that for these reactors, U^{233} has apparent higher values than plutonium, but correspondingly higher fuel costs, namely 2.63 mills/kwh_e compared with 2.15-2.16 mills/kwh_e for the plutonium U^{238} systems. This reflects the basically higher cost of the first step of the thorium cycle which is enriched with nearly pure U^{235} from the uranium cascade, while the first step of the U^{238} cycle uses lower priced, slightly enriched U^{238} from the uranium cascade. Reactors designed for higher conversion ratios with U^{233} can make back more of the original U^{235} investment incurred which should be reflected in lower costs. The apparent anomaly of high bred fuel values not necessarily assuring low fuel costs (or power costs) is manifested throughout such studies. As used here, plutonium value is defined as the price of plutonium that yields the same fuel cost whether a given reactor is fueled with plutonium or with uranium. Interactions between reactors, ore sources, and stockpiles are not considered. It is important then, that the term "value" should not be confused with the term "price", a market term, beyond the scope of this study. Of course, the plutonium values computed in these studies are basic to future consideration of the interchange of plutonium among different reactors because those reactors for which the computed values of plutonium are high would probably be users of plutonium, and those reactors for which the values are low would probably be the producers.

FIGURE 5

Bred Fuel Value in Successive Recycle at the Enrichment for Minimum Fuel Cost

It should be noted in Figure 5 that as the recycle sequence proceeds, the value of plutonium is lowered more than the value of U^{233} , particularly for the second recycle system involving hypothetical stockpiles. In this case, the isotopic compositions of plutonium contain far more Pu^{242} than the classical recycle scheme. In fact, at Step 6, Pu^{242} makes up 72.5 percent of the plutonium for the hypothetical stockpile system and only 18 percent of the plutonium for the classical recycle system. The basic reason for the decrease in performance of this plutonium is shown in Figure 6 to be the Pu^{242} . The lower curve in this figure relates the fuel exposure that can be achieved using the high Pu^{242} content plutonium to enrich natural uranium. The upper curve results when the Pu^{242} is completely removed from the plutonium source. The isotopic composition of the plutonium for the lower curve is 5 percent Pu^{239} , 18.0 percent Pu^{240} , 4.5 percent Pu^{241} , and 72.5 percent Pu^{242} , while the composition that results by omitting Pu^{242} and used to develop the upper curve is 18 percent Pu^{239} , 66 percent Pu^{240} , and 16 percent Pu^{241} . The intermediate curve is for the high Pu^{242} content plutonium, but operated at twice the specific power to reduce the formation of Am^{241} by the decay of Pu^{241} . This shows that the decrease in productivity is not due to parasitic neutron capture in Am^{241} but that the absorptions in Pu^{242} appear to be the dominant effect. It is believed that in actuality, the effective absorption cross section of Pu^{242} will be substantially less than used in our computation model, thereby elevating the value of such highly burned plutonium. In HW-72217, it was shown that the decrease in plutonium value with successive recycle for the classical recycle case could be closely correlated with the build up of Pu^{242} . With this in mind, relative plutonium values can be indicated by the value of plutonium for the first recycle step only. Such a summary for the majority of the studies completed is shown in Table II.

TABLE II
COMPUTED RESULTS FOR SUCCESSIVE RECYCLE OF BRED FUEL
Listing Bred Fuel Values in \$/gram⁽¹⁾ (Fissile) and Corresponding Fuel Costs in mills/kwh_e⁽²⁾ for Five Simulated Reactors and Six Operating Modes

Simulated Reactor Type	Plutonium Successively Recycled for Minimum Fuel Cost				U^{233} Successively Recycled for Minimum Fuel Cost	
	Self-Produced Pu Recycled with Optimum U^{235} Enrichment of U^{238}		Optimum Pu Quantity Recycled in Natural Uranium		Self-Produced U^{233} Recycled with Optimum U^{235} Enrichment of Th	
	Batch Loading	Graded Loading	Batch Loading	Graded Loading	Batch Loading	Graded Loading
APWR	12.10 (2.15)	11.80 (1.73)	11.10 (2.16)	(Not to be Calculated)	13.90 (2.63)	(Not to be Calculated)
BWR	11.90 (1.74)	10.20 (1.28)	10.30 (1.77)	(Incomplete)	14.00 (2.00)	(Incomplete)
HWR	11.50 (1.71)	10.30 (1.15)	10.20 (1.72)	(Incomplete)	14.00 (1.89)	14.20 (1.57)
GCR	12.70 (1.82)	11.80 (1.20)	11.20 (1.98)	11.20 (1.20)	13.60 (2.35)	13.40 (1.79)
OMR	12.50 (1.96)	(Incomplete)	12.80 (1.75)	(Not to be Calculated)	13.10 (2.30)	(Not to be Calculated)

Note: (1) Shown as the Top Figure for each Instance

(2) Shown in Parenthesis for each Instance

As shown in Table II, two major fuel management schemes have been examined--batch and graded loading. In batch fueling, all fuel is placed in the reactor at the same time and discharged at the same time; but it may be shuffled periodically within the reactor. In graded fueling, the fuel is charged and discharged more or less continuously. For a given enrichment level, graded operation achieves approximately twice the fuel exposure of batch operation, but over-all costs are not necessarily lower because there are charges for startup and shutdown to establish the equilibrium cycle and there are also capital outlays for the necessary reactor apparatus. Many reactor fuel cycles are a combination of the two systems. It is observed that the plutonium values tabulated here are remarkably close to one another and only a few trends from uniformity can be noted.

Plutonium values are somewhat less and fuel costs are significantly less for the graded cycle than for the batch cycle. Plutonium values are the least for the heavy water reactor. Although not shown on Table II, the lowest plutonium values are for the graded cycle using a hypothetical stockpile of plutonium to enrich natural uranium. As indicated on Figure 6, this latter effect is due to the rapid build up of Pu^{242} , which is most severe for the graded cycle in which the fuel exposure for each step is nearly twice that for the batch cycle. When the values are normalized to Pu^{242} content rather than to the step number, the values are more nearly the same for the batch and graded cycles. It should be noted that the value of U^{233} is very nearly the same for all reactors. This reflects the insensitivity of U^{233} 's properties to reactor spectrum. Furthermore, the U^{233} values as well as associated fuel costs are both greater than those of plutonium.

FIGURE 6

Exposure Characteristics of Highly Burned Plutonium

1. Effect of Plutonium Fabrication Costs (Zoned and Nonzoned Reactors)
2. Effect of Uranium Price Schedules and Fuel Use Charges
3. Effect of Uranium Supply Limited to Natural Uranium
4. Effect of Eliminating Uranium Cascade After First and Third Steps
5. Effect of Limiting Control System to Value Below Optimum
6. Effect of BWR and APWR Lattice Variations
7. Effect of Key Generalized Reactor Parameters
8. Effect of Key Bred Fuel Nuclear Properties

FIGURE 7

Special RECYCLE CHAIN Studies

Examination of the possible sensitivity of plutonium values with variations in reactor parameters cannot be done here because too many variables are altered in the five simulations. Accordingly, supplementary special studies of limited recycle modes in a wider range of reactors have been conducted, including many variations of the economic environment. A partial listing of these is shown in Figure 7. The relatively minor influence upon plutonium values of the expected incremental fabrication charges associated with plutonium fuels has been discussed as has the second item, the influence of uranium price schedules. Item 3, which deals with limiting the fuel sources to natural uranium, indicates that without plutonium recycle, the reactor types having reasonable fuel costs are limited to those of extremely high nuclear economy such as D₂O types with zirconium jacketing and to those with extremely high fast effects, each operating on the graded fuel cycle. If plutonium recycle with natural uranium is considered, a wider range of reactor types will have reasonable fuel costs. For these recycle cases, the apparent plutonium values can be very high because of the dramatic increase in fuel exposure available when natural uranium enrichment is supplemented with recycled plutonium. For recycle not to be of potential benefit to natural uranium cases on a cost basis, the neutron economy of the reactor must be very high, the cost of jacketing fuel must be relatively low, and the cost of spent fuel separations must be high.

A wider set of reactor conditions can be accommodated under Item 4 which deals with the circumstance of having the slightly enriched uranium source interrupted and natural uranium becoming the sole supply. Depending upon the size of the slightly enriched uranium inventory in the pipeline, practically any of the thermal reactor and associated fuel element systems studied to date can be operated with plutonium recycle at reasonable fuel costs for periods of 5 to 10 years.

The fifth item deals with the fact that the high cross section of Pu^{240} also enables it to serve well as a burnable, fertile poison, and reactors having limited control systems can substantially extend their reactivity life-time by using it. This is not brought out in the RECYCLE CHAIN studies because optimum cycle operating points are chosen at minimum fuel cost without any constraints by a control system. If such constraints are applied, the U235-enriched U238 step usually has increased costs because reactivity-peaking limits are reached before minimum cost exposures are developed, whereas plutonium-enriched systems do not encounter this limit because of Pu^{240} . Under this circumstance, the productivity of plutonium fueling was increased by substantial amounts in preliminary examples computed for the APWR with an arbitrarily limited control system. The long-term productivity of this characteristic is largely dependent upon the relative costs of control systems and until these are known, the values computed will be somewhat tenuous. It must also be remembered that this cycle must compete against the graded cycle which is usually of lower cost and slightly lower plutonium values.

The lattice variations noted in Item 6 are made to ascertain the sensitivity of plutonium values to the jacketing materials, moderator-to-fuel volume, moderator temperatures, and other factors in view of the simplifications necessary to employ the MELEAGER code. Part of this examination involved substituting the parasitic neutron absorption term used for the APWR in the BWR. This elevated the computed plutonium value for the first cycle from \$11.90/gram (fissile) to \$12.20/gram (fissile) for which the corresponding APWR value was \$12.10. The moderator index for the APWR was substituted in the BWR, which lowered values from \$11.90 to \$11.40. These minor variations result in productivity changes that appear correct as to direction, but correctness of the relative magnitude of the effect cannot be assured. It appears that the slight difference in computed values of the first cycle plutonium for the five reactor types could be reversed by slightly altering two variables or by changing the associated analytical treatment. This may not be the case for the more highly burned plutonium.

Items 7 and 8 are designed to gain more insight by making larger changes in the key reactor and key isotope properties. To reduce confounding of the many variables only one term is being altered at a time even though such experiments could not be duplicated in practice. An example is shown in Figure 8 which relates the computed indifference value and associated costs as the moderator index is altered. A definite peak in value and minimum in cost are shown at an index slightly above two, which closely corresponds

to the APWR figures. Another dimension of this study is the parasitic absorption factor which was arbitrarily held constant for calculations making up the report in Figure 8, because this factor tends to confound the issue as does moderator temperature.

FIGURE 8

Values of Recycled Plutonium and Corresponding Fuel Costs as Affected by the Amount of Moderation

Item 8 of Figure 7 is addressed to the more general problem that the use of models of reactor physics and economics systems on computers does not alter the uncertainties in the facts upon which the computations are based. A major purpose of the Plutonium Recycle Program is, of course, to supply authoritative data for such studies. In the meantime, plutonium value computations are being made which would be considered precarious extrapolations of known facts unless a concurrent investigative program such as Plutonium Recycle were not well enough along to afford any necessary recalibration. Except for Pu^{242} , the bred fuel nuclear parameters are most important in the first steps of a plutonium value analysis wherein plutonium compositions are changing most rapidly. These can be measured by perturbation of the physics parameters and observation of the effect on the relative fuel costs for an appreciated recycle chain sequence involving the first cycle (which is uranium enriched) and two plutonium recycle steps. Figure 9 shows the results of such calculations for the APWR in which the ν value for Pu^{239} was varied to effect a change in η . The absorption cross section of Pu^{239} was also varied. These data indicate that the high neutron absorption cross section of plutonium is an asset especially in view of the lower η values in the APWR neutron spectrum. Similar studies indicate that the value of this cross section asset is not so great in a reactor such as the HWR which has very little parasitic absorption. Figure 9 also indicates that the plutonium values being computed are not precarious in the sense that a slight change in effective η or cross-section figures will not greatly alter the values. The associated fuel costs, of course, do vary.

FIGURE 9

Variation in the Value of Plutonium in the APWR as a Function of η .
The Neutron Reproduction Coefficient

COMBINED CYCLE CHAIN Studies

The foregoing results of the successive recycle analysis, while informative, are highly restrictive because of the limitation of plutonium enrichment to the reactor in which they were produced. Except by coincidence, this does not allow cross comparison of the value of a specific plutonium batch in the various reactors because plutonium compositions developed in successive recycle are unique to the parent reactor spectrum and the values of a specific step as computed by PUVe are dependent upon the values of the preceding as well as following steps. To provide such insight into the exchange of fuels among reactors, computations are under way examining the use of selected plutonium batches of different isotopic compositions as alternatives to U^{233} or U^{235} enrichment of U^{238} and thorium in selected reactor concepts. To complete this picture, the potential of enriching U^{238} with U^{233} is also being studied. Thus, the term "crossed progeny" has been used to describe this portion of the more generally termed "Combined Cycle Study" summarized in Figure 10. The scope of this study is so great that limitation of the specific cases examined is necessary. To facilitate this selection, a broad survey was completed using generalized reactor characteristics from which the more promising systems could be identified. The fueling systems are limited to U^{238} and Th^{232} , each enriched with five alternatives: U^{233} , U^{235} , and three different isotopic compositions of plutonium.

OUTLINE OF COMBINED CYCLE STUDY

FIGURE 10

The fuel geometry is being limited initially to the classical shapes, such as large single rounds, clusters of rounds, and coaxial tubes. Cladding material, average fuel density, and isotopic distribution of enrichment are varied only for those fueling systems whose values can be enhanced by altering them. The reactor types are currently limited to three general moderators: H_2O , D_2O , and graphite. The lattice spacing is varied, and an optimum is chosen for each enrichment level. The reactor size is varied

by use of adjoint solutions. The interactions with fast reactors are limited to fuel reuse considering the insertion of fast reactor blanket rods into thermal reactors without an intermediary separations step and to the determination of the general productivity of the three standard plutonium isotope batches as enrichment for selected fast reactors. Burner reactors involving specific plutonium fuels are being examined in a separate program with specific reactor design concepts. The fuel management schemes being considered are necessarily approximations. The idealization of these approximations is greater the smaller the reactor and is the least for large central station reactors of current specific power. A wide variety of economic variables is examined as shown in Table III. This is possible because of the use of the MINIMIZER code which selects a new minimum operating point for each economic circumstance. The selection of economic parameters anticipates future reduced jacketing costs, possible increases in fuel use charges, and alterations of the uranium price schedules. A more distant future anchor point involving an increase in uranium costs by a factor of three is also used for which the minimum fuel cost often coincides with maximum uranium conservation. This seldom occurs with the current economic environment of low priced uranium and relatively high jacketing costs.

TABLE III
STANDARD COMBINATIONS OF ECONOMICS PARAMETERS
USED IN THE COMBINED CYCLES STUDY

(Combinations are Shown as Variations from the Standard Case Number 1)

Economics Numbers	Economic Interest Rate %	AEC Use Charges %	Fuel Fabrication Charges \$/# U	Spent Fuel Separations Charges \$/# U	Factor Plutonium Cost To Fully Enriched U Price	Thermal To Electrical Conversion Efficiency	^{U235} Price Schedule (% Of Present Schedule)	Ratio: Price of Pu In Price of Pu Out
STANDARD CASE 1								
1	12.5	4.75	\$30	\$10	.85	.333	1.00	1.00
2	.	.	10					
3	.	.	20					
4	.	.	40					
5	.	.	60					
6	.	.	00	20				
7	.	.	.	.	.425			
8	.	.	.	.	1.700			
9	.	.	.	.	.	.	.750	
10	.	7.0	.	.	.	.	.	
11	.	10.0	.	.	.	.	.	
12	.	12.5	.	.	.	.	.	
13	.	12.5	.	.	.	.	.500	
14	.	12.5	.	.	.	.	.750	
15	.	.	.	.	.	.	3.000	
16	.	12.5	.	.	.	.	3.000	
17	.	.	.	.	.	.	.	.85
18	.	.	.	.	.	.	.	1.15

The COMBINED CYCLE CHAIN code is the major computation tool employed. Data with respect to the basic lattice cell and associated temperature specifications are fed to a transport theory code using the P-3 approximation. The results are fitted against cell blackness and passed to MELEAGER. Other parameters such as reactor size, neutron leakage, and the fast effect are evaluated with appropriate subroutines. The MELEAGER code then computes the burn-up relationships. The data are then sent directly to the cost code QUICK to determine the fuel costs for each enrichment level. MINIMIZER then computes the condition for minimizing cost for each set of economic circumstances. The code repeats these calculations to determine minimum fuel costs for as many lattice spacings as desired and then automatically computes the moderator conditions yielding the lowest fuel cost minimized against enrichment. Thus, a double minimization is performed. Figure 11 shows the minimum cost selection as a function of enrichment for a fixed moderator spacing. The lower the fuel fabrication cost, the lower are the fuel costs and necessary fuel exposures. The optimum lattice spacing is then determined by examining the minimum fuel costs as a function of moderator spacing. This is shown in Figure 12 for the batch and graded cycles.* It should be observed that the optimum cost circumstance for batch and graded cycles occurs at substantially different moderator indexes. On this basis, one would not expect that plutonium values in a single reactor design would be the same for the batch and graded cycles. In the cases studied to date, the plutonium value with graded cycles is usually less, which would be expected on the basis of the Figure 12. For each moderator and fuel type, a surface is generated to determine the optimum lattice spacing. Such a surface is shown in Figure 13. A new surface and optimum point are developed for each of the eighteen economic environments. These and other associated data are now being analyzed and only selected cases are shown here for purposes of illustration. The difference in optimum moderator index for plutonium of different isotopic composition is shown in Figure 14. The optimum moderator indexes are distinctly different for batch and graded loadings. For each loading scheme, the optimum for each enrichment material is quite close. However, it should be noted that plutonium Batch No. 3 which is high in Pu^{240} is the most sensitive to moderator index variations; and for low index values, it will not go critical as indicated by the dotted lines. Figure 15 shows that the optimum moderator index is different for each moderator type. A large factor contributing to this is that graphite and H_2O each has significant capture cross sections. As a consequence, the more moderator, the more parasitic absorptions.

Selection of the optimum moderator index for D_2O moderated reactors involves consideration of the interest charges and losses for the moderating material in addition to the usual fuel cost considerations. Adding the D_2O expense to the fuel cycle costs as has been done in Figure 15 allows inclusion of this factor in moderator index optimization. This type of charge is negligible for the H_2O and graphite moderated cases in comparison to the rest of the fuel cost components.

*Unusual start-up charges for the graded cycle are approximated by amortizing over a 30-year life one-half the cost of the first load. No shut-down charges are levied and the graded costs reported here may be more or less than specific instances may develop.

FIGURE 11

Minimized Total Fuel Cost (FEFJ Varied) and Fuel Exposure as Functions of Fissile Enrichment for a Plutonium Enriched Reactor

FIGURE 12

Minimized Total Fuel Cost as a Function of Moderator Index Comparing Batch and Graded Fuel Cycles

Note, Fuel Natural Uranium Enriched with Plutonium Composition
95% Pu-239
5% Pu-240
Heavy Water Moderated

FIGURE 13

Total Fuel Cost Optimization
(Varying Fissile Enrichment and Moderator Index for
a Plutonium Enriched Reactor)

Type	Uranium Source	Isotopic Weight Fraction of Source Plutonium *			
		Pu-239	Pu-240	Pu-241	Pu-242
U-1	Cascade	0.0	0.0	0.0	0.0
Pu 1	Natural	95.0	5.0		
Pu 2	Natural	70.0	18.0	11.0	1.0
Pu-3	Natural	31.3	33.6	25.0	10.1

* For Plutonium Fuel Types Sufficient Plutonium of the Indicated Composition is Added to Natural Uranium to Yield Minimum Fuel Costs. The Fuel Cost of the Uranium Fuel Types are Similarly Minimized by Varying the U²³⁵ Enrichment Level

FIGURE 14

Minimized Fuel Costs as a Function of Moderator Index
for Various Enrichment Materials

* Moderator Index is the Slowing Down Power Normalized to Unit Fuel Volume

Note: Fuel; Natural Uranium Enriched with Plutonium Composition
 95% Pu-239
 5% Pu-240

FIGURE 15

The Effect of Moderator Type on the Optimization of Moderator Index with Pu Enrichment

It can be argued that the interest charge of 12-1/2 percent, as applied in Figure 15, is excessive although it should be kept in mind that a lower interest rate would have to have an expense for obtaining surety since there is a finite possibility of catastrophic losses. The impact of the D_2O costs is considerable even at reduced interest rates as shown in Table IV. The data are for a 200 megawatt electrical plant operating the fuel at an average specific power of 15 megawatts per ton of contained U in UO_2 . Losses are set at 2 percent per year of the total D_2O inventory at \$28.50 per pound for interest rates of 12-1/2 and 5-3/4 percent. The losses are assumed to be zero and the cost of D_2O is assumed to be reduced to \$17.50 per pound in the right-hand columns of Table IV. In all cases depicted in Figure 15, the minimum fuel cost is achieved by adding plutonium to natural uranium. The

jacketing cost used is \$30/pound uranium. At substantially lower jacketing costs, minimum fuel cost with natural uranium enrichment may be observed for high moderator index values.

TABLE IV

ESTIMATED D₂O CAPITAL CHARGES IN MILLS PER KILOWATT HOUR
ELECTRICAL AS AFFECTED BY INTEREST RATES AND MODERATOR INDEX

Moderator Index	12-1/2% Int. \$28. 50/lb D ₂ O,	5-3/4% Int. \$28. 50/lb D ₂ O,	12-1/2% Int. \$17. 50/lb D ₂ O,	5-3/4% Int. \$17. 50/lb D ₂ O
	2% Loss	2% Loss	0% Loss	0% Loss
0. 84	0. 507	0. 271	0. 269	0. 124
1. 05	0. 530	0. 283	0. 280	0. 128
1. 31	0. 583	0. 312	0. 309	0. 142
1. 57	0. 635	0. 340	0. 336	0. 155
2. 09	0. 738	0. 394	0. 390	0. 180
3. 13	0. 934	0. 499	0. 494	0. 227
5. 22	1. 309	0. 700	0. 693	0. 319

The potential productivity of plutonium's distinctively larger thermal reactor cross sections is shown in Figure 16 which relates the increase in optimized fuel costs for U²³⁵ and plutonium enriched UO₂ fuels as the amount of stainless steel jacketing is increased. This points up the fact that for very small amounts of stainless steel jacketing, the high absorption cross sections of plutonium are of only nominal importance. At more substantial thicknesses, such as 0.020-inch, the increased plutonium cross sections are of benefit. It is important to realize that this comparison is for fully optimized systems, both with respect to moderator index and enrichment. Greater or lesser benefits may exist for fixed lattices in which only the enrichment level is altered for each jacketing situation. It also appears that the fast effect in U²³⁸ will be of particular benefit to some plutonium batches. The observed productivity is so large that the calculations are being refined because the conversion ratios encountered are often near or greater than unity. Conversion ratios substantially greater than unity are encountered with U²³³ enrichment of U²³⁸ arranged for a high fast effect. This latter cycle requires additional examination as to how to appropriately close it and as to what costs may be involved to prevent irreversibly mixing of U²³³ in U²³⁸.

FIGURE 16

Incremental Fuel Cost for Stainless Steel Clad Fuel
Comparing U^{235} and Pu Enrichment (Optimized Lattices)

Reduced Concentrations of U^{238}

The basic Combined Cycle Study data are not all in hand yet and until they are available, reporting must be limited to noting trends as the relative values of each system can be established only after the major alternatives have been evaluated. One facet of the study involving reducing the density of U^{238} has been separated and pursued independently. This appears important when enriching with plutonium batches containing significant quantities of Pu^{240} because with full U^{238} density more fertility is developed than can be economically supported. In the extreme, such a system may have conversion ratio of 1-1/2 and reactivity below 1 and, hence, will not function. As noted in Figure 14 by the-----lines, this was encountered with plutonium Batch No. 3 in closely packed lattices.

The effect of varying the U^{238} spatial concentration is illustrated in Figure 17 which is a plot of the initial reactivity (k_{∞}) as a function of the spatial concentration of U^{238} in the fuel (the jacketed fuel size and the plutonium spatial concentration being held constant). Note that the reactivity rises as the U^{238} average spatial concentration is reduced. However, this does not mean that the optimum operating point corresponds to the near maximum reactivity point since the conversion ratio drops as the U^{238} spatial concentration is reduced. These data are for a plutonium isotopic composition 45 atom percent Pu^{239} , 40 atom percent Pu^{240} , 10 atom percent Pu^{241} , and 5 atom percent Pu^{242} in a relatively closely packed lattice with UO_2 fuel rods approximately 1/4-inch in diameter. The corresponding impact on minimized fuel costs is shown in Figure 18 in which slight sensitivity to the jacketing cost is indicated. The minimized fuel costs for two jacketing costs and two fuel use charges are shown as a function of U^{238} spatial concentration. The jacketing charges are in terms of \$/cubic centimeter of fuel. For orientation, it is noted that \$1.22 per cubic centimeter of fuel corresponds to \$60/pound uranium if the fuel is composed of 100 percent dense UO_2 . Similarly, \$0.61 per cubic centimeter corresponds to \$30/pound uranium at full UO_2 density. Without combinations of composition and lattice as used here such economic minimums are not as marked and are not observed for U^{233} , U^{235} , and 95 percent Pu^{239} -5 percent Pu^{240} .

FIGURE 17

Conversion Ratio and k_{∞} as a Function
of Spatial Concentration of the Fuel
Plutonium Enrichment of Tails Uranium

FIGURE 18

Minimized Fuel Costs as a Function of Spatial
Concentration of the Fuel
Plutonium in Tails Uranium
(AEC Interest Charges and FEFJ Parameterized)

These data indicate that reduction in fertile concentration of the fuel is of benefit to some plutonium batches if certain lattice configurations are inherited. A more general question involves whether or not an optimum lattice spacing exists for which full density UO_2 yields the lowest fuel costs when high Pu^{240} content plutonium is used for enrichment. Some results of such a study are shown in Figure 19. For several levels of moderator index bracketing current power reactor designs, it appears that the lowest costs for this plutonium composition do exist for reduced density fuel. The top three curves are for the same resonance shielding index corresponding to 1/2-inch diameter. The lowest curve is for 1/4-inch diameter rods with correspondingly less

resonance shielding, but has the same moderator index as the curve immediately above it. The bottom curve represents the lowest fuel costs observed for all the reactor perturbations made for this plutonium composition. It should be noted that by use of certain geometrical configurations, these conclusions may be altered. Attention is drawn to Figure 3 which considered the alternative of concentrating the plutonium in the center of the fuel as opposed to uniform distribution. The decrease in the alpha value of plutonium by such concentration is accompanied by a decrease in effective cross sections. The Pu^{240} cross section is most directly altered and in that example was reduced by a factor of two. With this value for the Pu^{240} cross section, it is doubtful that a reduction in U^{238} spatial concentration would be of benefit. This does not mean that such a fuel is necessarily more economical than uniformly enriched reduced density fuels as the specific power is lower and the relative flux depressions are less favorable than uniformly enriched examples. Obviously additional study is warranted.

FIGURE 19

Minimized Fuel Costs as a Function of Spatial
 Concentration of the Fuel
 Plutonium in Tails Uranium

Phoenix Fuels

If the U^{238} spatial concentration is reduced essentially to zero and the reactor is fueled completely with plutonium, certain combinations of Pu^{239} , Pu^{240} , and Pu^{241} lead to extraordinarily long reactivity lifetimes in view of the total weight of the fuel loading. These fuels have been named "Phoenix Fuels". Phoenix fuel conditions result when the Pu^{240} decreases and the Pu^{241} grows appropriately at just the right rates to maintain the reactivity, which would otherwise drop off because of buildup of fission products and burnout of Pu^{239} . Phoenix fuels use the batch cycle and can have an unusually constant reactivity without use of parasitic control systems. They are not breeders and do not have high conversion ratios throughout life. Figure 20 shows the reactivity as a function of exposure of such a fuel at various fuel concentration levels. Attainment of satisfactory Phoenix fuel performance is highly dependent upon achieving a large effective neutron absorption cross section for Pu^{240} so as to grow Pu^{241} at an adequate rate. While an initially satisfactory Pu^{241} growth rate can be achieved with a low effective Pu^{240} cross section by arbitrarily increasing the Pu^{240} concentration, the ultimate lifetime of such fuel could be curtailed because as exposure proceeds, there may be too much Pu^{240} left in proportion to the remaining concentration of Pu^{239} and Pu^{241} and the reactivity would drop sharply.

FIGURE 20

Reactivity as a Function of Exposure for a Phoenix Fuel
Plutonium/cc Paramaterized

Long fuel exposures without poison control can be achieved with reactors using the graded, variable spectrum, or multiregion fueling systems. However, tonnage quantities of fertile U^{238} or thorium are involved. This is distinctly different from the Phoenix fuels in which control is an inherent fuel characteristic. Since the total amount of fertile fuel is limited to the Pu^{240} content, fuel costs for Phoenix fuels are very sensitive to inventory charges and hence to specific power as shown in Figure 21. The upper curve is for a likely unreal jacketing cost of \$2.44/cc of fuel which corresponds to \$120/lb cost for 100 percent dense UO_2 . This high jacketing cost example was used to emphasize the effect of specific power. The lower curves are for an equivalent jacketing cost of \$30/lb of uranium at 100 percent UO_2 density. A specific power of one kw/cc corresponds to 90 megawatts/ton at the conventional U^{238} density of 10 grams/cc. Phoenix fuel costs are sensitive to specific power not only because of the valuable inventory, but also because of the radioactive decay of Pu^{241} . In view of Pu^{241} 's 13-year half-life, this latter effect is most marked and can limit the maximum lifetime of a Phoenix fuel. This is illustrated in Figure 22 which is a plot of Phoenix fuel life in years as a function of reactor specific power. The maximum life of 15 years corresponds to a specific power about six times the average of current water cooled central station power reactors. The decrease in life is due to the diminished concentration of Pu^{241} as decay proceeds and also due to neutron capture in americium-241, the decay product. The latter has several low lying resonances which are coincident with those of Pu^{239} and Pu^{241} and mutually shield one another. The maximum expected life will likely be increased when this is more accurately taken into account. The loss of Pu^{241} per unit of heat can be made insignificant at extremely high specific powers, but the life is then simply limited by the fact that the fuel is completely burned in a short time. For the many applications in which maximum fuel life-time is of no concern, the Phoenix fuels may have considerable overall economic merit. Fuel costs for Phoenix fuels are minimized by selecting the optimum fissile concentration in the fuel rather than altering the ratios of fissile plutonium to fertile plutonium. This is shown in Figure 23. This indicates that minimum fuel cost operating points are sensitive to interest charges and jacketing costs. The minimum fuel cost of 1.4 mills/kwh_e corresponds to 10 percent fissile concentration in 100 percent dense UO_2 and yields a lifetime of five to six years. The possible augmentation of the pure Phoenix fuels by addition of U^{238} or thorium offers many avenues for investigation. Such studies are being interwoven with the reduced spatial concentration and geometrical shielding studies.

FIGURE 21

Optimized* Fuel Costs as a Function of Specific Power
for a Plutonium Phoenix Fuel

*Optimized fuel cost is the minimum fuel cost at optimum fuel density.

FIGURE 22

Phoenix Fuel Lifetime as a Function of Specific Power

FIGURE 23

Minimized Fuel Costs for a Phoenix Fuel
Specific Power 330 W/cc*

*330 watts/cc $\approx$ 30 megawatts/ton at 10 gram/cc fuel density.

Concluding Remarks

In summary, the following observations are noted. It appears that incremental fabrication charges associated with plutonium toxicity will not greatly reduce plutonium values because the reactor fuel loadings can be designed either to concentrate all of the plutonium in a part of the fuel elements or to employ a large total amount of plutonium throughout the reactor. Either design distributes the modest incremental fabrication costs (believed to be \$5 to \$10 per pound at most) over a sufficiently large number of grams of plutonium that the reduction in plutonium value need be no more than fifty cents to a dollar per gram.

It is reported that plutonium values are proportional to the cost of fully-enriched uranium for the AEC uranium price schedule in question. Conversely, the cost of using plutonium priced relative to fully-enriched uranium will follow the changes in the cost of fully-enriched uranium from the cascade and not just the price of separative duty or natural uranium feed. In the analyses reported, plutonium value has been defined as the price of plutonium that yields the same fuel cost whether a given reactor is fueled with plutonium or with uranium. This definition of value should not be confused with price, a marketing term involving the possibility of exchange of plutonium among many parties which has not been considered quantitatively in this report. The computed plutonium values for the successive recycle mode for thermal reactors range between \$10 and \$12 per gram of fissile plutonium with the post-July 1, 1962 uranium price schedule as a standard.

In the thermal spectrum, the neutrons released and the energy liberated per atom of Pu^{239} destroyed are somewhat less than that of U^{235} . This is counter-balanced if the large effective cross sections and unique plutonium progeny are of adequate benefit to the reactor in question. In reactors having few parasitic absorbers such as the D_2O reactor with zirconium hardware, relatively large cross sections are of little benefit and this is reflected by the lower plutonium values computed for such machines. However, by using plutonium enrichment as small diameter rods, geometrical shielding can raise the value of η and lower the effective cross sections. This may be a more valuable characteristic for reactors having few parasitic absorptions. Since the higher plutonium cross sections allow greater use of stainless steel for jacketing, for process tube liners, and for other hardware, various fuel loadings and reactor designs could exploit this feature. In addition, the high nuclear cross sections can permit the use of smaller cores.

Under some circumstances, the high Pu^{240} cross section allows reduction in the average fuel concentration and may be useful for matrix fuel plates in a special zone of the reactor. The relatively constant reactivity Phoenix fuels further exploit the high cross section of Pu^{240} and may permit the use of long-lived compact reactor cores (total active life likely limited to 15 years, or so, by virtue of Pu^{241} decay).

Of great interest is the possibility of lowering costs and of integrating fuel resources through the use of fast and thermal reactor combinations together with crossed progeny of the fertile species of U^{238} and thorium-232.

PREPARED DISCUSSION, SESSION II

PLUTONIUM IN THE SWEDISH POWER REACTOR PROGRAM

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I. INTRODUCTION

To understand the Swedish standpoint on the use of plutonium in power reactors the following should be appreciated.

- (i) The Swedish power reactor program is characterized by a concentration on heavy water moderated reactors of pressure vessel type designed for natural or slightly enriched uranium. The first reactor of this type - Ågesta 65 MWth - is expected to go critical towards the end of this year; it is cooled by pressurized heavy water confined to shrouds surrounding clusters of fuel rods. The development is along two main lines; the first an advanced pressurized water design, where the coolant shrouds have been eliminated and the fuel rods are uniformly distributed over the core; the second a boiling and integral superheat design.
 - (ii) The present rate of development of the power reactor program is relatively slow, since hydro power is expected to meet the increased demand of electricity for another ten years. Present prognosis shows that an installed nuclear power of 1000 MW is required in 1975 and 4000 MW in 1980. The present power reactor program calls for a full scale reactor Marviken (100 - 200 MWe) in operation by 1967 - 1968, a further (400 MW) in 1972 and from 1975 one or two large (600 - 1000 MW) stations a year.
 - (iii) Sweden has great reserves of low grade uranium ores but no thorium has as yet been found. An uranium plant with a capacity of 120 t/year is under construction at Ranstad and will be in operation in 1965.
 - (iv) No uranium isotope separation plant is available or planned.
- The problem regarding the use of Pu may be stated as follows:
- 1. What types of fuels and fuel cycles are of interest?
 - 2. What is the economical potential of Pu?
 - 3. What are the technical problems in developing and using Pu and how can these be solved?

After reviewing briefly the present thinking and state of affairs in Sweden a short description will be given of how answers are sought on these questions.

The most efficient use of Pu ultimately may very well be in fast reactors. This reactor type is also included in the long range development reactor program in Sweden. The present work is, however, confined to basic physics in connection with the zero energy facility being constructed for operation in 1963. In the following mainly thermal reactors will be discussed.

II. FUEL AND FUEL CYCLES

Disregarding pure burning of plutonium, which may find only special application, plutonium may be used in connection with U or Th as breeding material. The U-cycle is considered to be of major interest, mostly because the early development will be based on this cycle and no significant Th-sources are known. Furthermore, the Pu-Th-cycle does not appear to have too attractive nuclear characteristics and requires larger investments of Pu than that of the U-cycle.

Although single fuel cycles hitherto have been used almost exclusively, combined fuel cycles are believed to be more advantageous and should find wider use in the future. By a combination of different fuels and types of reactors it appears easier to meet different and sometimes contradicting requirements, e.g., good overall economy, high conversion ratios and short doubling-times in breeders. In this way some of the disadvantages of the Th-U²³³ cycle, e.g., high parasitic absorption and low specific power, may be eliminated by the introduction of Pu. Also, plutonium may be formed in one reactor optimized for production and used in another optimized for this purpose.

The fuel material has to fulfil a great number of requirements, metallurgical stability, compatibility, neutron economy, low cost etc. Taking all these into account only one type of fuel appears feasible for present Swedish circumstances, viz, mixed Pu and U-oxides.

No acceptable Pu alloy seems available. Alloys with Fe and Ni have good metallurgical properties but give poor neutron economy. Pu-Al alloys are of doubtful value in water reactor systems, especially if high temperatures are aimed at. Pu-Zr offers uncertain expectations, the most serious difficulty being irradiation stability due to the nonisotropic properties of Zr.

Of ceramic fuels carbide is excluded at present in water cooled reactors due to the instability in hot water. If this problem could be overcome, it has good potential in many respects.

Fuels of the cermet type, e.g., plutonium oxide in stainless steel, may find use in certain instances as in superheater elements or fast reactors.

Regarding the fuel element, designs similar to those developed for uranium are of prime interest. Mixtures of Pu and U are preferred to those containing U and Pu separately. Considering burnup requirements and heat ratings, the plutonium enrichment will be confined to a maximum of a few percent.

III. ECONOMICAL POTENTIAL OF PLUTONIUM

An attempt has been made to determine the value of plutonium as a fuel for the Marviken type of reactor. Some of the main characteristics of this reactor are summarized in Table I. Some of the cost assumptions used are given in Table II.

TABLE I

MAIN CHARACTERISTICS OF THE MARVIKEN 100 MWe REACTOR

Thermal Power	366 MW
Fuel	27-Rod Cluster UO_2
Fuel Rod, OD	13.5 mm
Sheath	0.6 mm Zr-2
Shroud, ID	108 mm
Thickness	1.0 mm
Lattice Pitch	28.1 cm square
Moderator	222 C D_2O

Coolant	234 C D_2O

Core Radius	211.9 cm
Core Height	442.0 cm
Extrapolated Core Radius	244.2 cm
Extrapolated Core Height	515.1 cm

TABLE II
COST ASSUMPTIONS

Natural Uranium Ore 8 \$/lb U_3O_8	19.3 \$/kg U
Refining and Production of UO_2 Powder	7.7 \$/kg U
Manufacture of Complete Fuel Element	53.0 \$/kg U
Storage Spent Fuel, Shipping Pre-Reactor	4.4 \$/kg U
Total Cost Once-Through Natural	84.4 \$/kg U
Reprocessing Base Charge	17000 \$/day
Shipping Post-Reactor	8.8 \$/kg U
Conversion of Pu	1.5 \$/gm
Conversion of U	5.6 \$/kg U
Waste Disposal	2.2 \$/kg U
Interest Rate U, Pu and Manufacture	7%/annum
Plant Load Factor	0.685

A great number of cost parameters are inadequately known at present. To study the effect on the plutonium value, the following four were chosen as being both important and uncertain:

- U = Cost of uranium (natural or enriched)
- X = Excess plutonium manufacturing cost (expressed relative to that of uranium)
- Y = Market value of plutonium
- Z = Cost of reprocessing including shipping and storage.

In Figures 1 to 6 are given examples of results from this study. Two main cycles were studied, viz, selfsustained recycling (Figures 1 to 4) and free optimization, with arbitrary plutonium compositions (Figures 5 and 6). In the first case comparison is made with the natural uranium cycle and the potential of plutonium is expressed as permissible excess plutonium manufacturing cost for equal fuel costs (Figure 2). For free plutonium enrichment a reactor value of plutonium is determined by equalizing the fuel costs for the plutonium and uranium cycles, which are both optimized with regard to enrichment (Figure 5).

FIGURE 1

Selfsustained Recycle Burnup Versus Cycle Number

FIGURE 2

Selfsustained Recycling. Fuel Cost Versus Excess Pu Fabrication

FIGURE 3

Selfsustained Recycling Break-Even Excess Plutonium Fabrication
Versus Integrated Burnup

FIGURE 4

Selfsustained Recycling, Effect of Plutonium Market Value Upon
the Break-Even Excess Fabrication Cost

FIGURE 5

Free Optimization of Plutonium Enrichment, Determination of Reactor Value from Variation of Minimum Fuel Costs Versus Plutonium Value

FIGURE 6

Plutonium Value Versus Quality of Pu
for Different Uranium Prices and Excess Manufacturing Costs

The following remarks may be made on the results:

- (i) Selfsustained recycling is competitive with the once-through natural uranium cycle, provided that the excess plutonium manufacturing cost is approximately equal to the uranium manufacturing cost (estimated to be 53 \$/kg U excluding fuel material).
- (ii) Free optimization gives reactor values of plutonium that depend strongly upon the excess plutonium manufacturing cost, and the cost of enriched uranium. This is particularly true in a system with a high conversion ratio, where more plutonium is achieved than is burned. (Note the shape of the plutonium curves in Figure 5.)
- (iii) The reactor value of plutonium does not vary significantly with an increase in higher isotopes until larger amounts of Pu^{242} is formed (Figure 6).
- (iv) With spiked or zoned cores, rather than homogeneous as assumed, considerably higher break-even plutonium manufacturing costs can be expected.

The conclusion is that plutonium will not find wide use in the Swedish reactor program before at least 1975. The reason for this is primarily not a lack of long range economical potential but rather that separation of plutonium from spent fuel and manufacture of plutonium bearing elements both require to become economical, large capacity plants which are not justified in the present program during the aforementioned period.

IV. PROBLEMS CONNECTED WITH THE USE OF PLUTONIUM IN REACTORS

Certain complications can arise in connection with the design or operation of reactors fueled with plutonium. The contribution of delayed neutrons is decreased to one-third of that of U^{235} , thereby causing more restrictive limitations in the choice of certain design parameters or necessitating greater reliance on the control system. Reactivity coefficients are not markedly different for water moderated reactors. The moderator coefficient may in some cases tend to more positive values. For fuel elements containing plutonium only, there is some uncertainty regarding the fuel temperature coefficient; although in all probability negative it is certainly significantly lower in magnitude than the corresponding quantity for uranium. This can imply a reduction in self stabilizing properties.

Other problems that may be aggravated by the use of plutonium, is associated with the power distribution and its changes with burnup. This would be particularly apparent with spiked cores. On the other hand there are also some potential operating advantages. The change of reactivity with burnup may for instance be decreased by using Pu^{240} as a fertile burnable poison. The xenon poisoning is lower and thus also the transients following changes in power.

V. THE SWEDISH DEVELOPMENT PROGRAM

In view of the points discussed above the plutonium work will be of long term nature and characterized by an initial concentration to thermal water moderated systems. Work on fast reactors, now mainly limited to basic physics problems, may later be initiated.

Much of the work is preliminary and might be subject to revision as experience or information is obtained. Four main categories of work are briefly outlined.

1. Fuel and Fuel Element Development

Work up to now has primarily been devoted to the development of the laboratory techniques for handling plutonium. The effort will be concentrated on the study of PuO_2 - UO_2 -system and the preparation of sintered mixed oxide. Chemical co-precipitation and mechanical mixing are both of interest. The phase diagram in the region of low concentration of PuO_2 will be studied.

Problems and economics involved in the manufacture of specified types of plutonium bearing elements will be studied.

2. Reprocessing and Separation of Plutonium

Sweden participates in the Eurochemic project and cooperates besides with Norway on a reprocessing pilot plant. A detailed pre-project study for a plant with a relatively limited capacity has been undertaken. The effect of plant capacity is being studied.

Alternative plutonium separation methods and problems of special Swedish interest, e.g., Zircaloy-canned elements will be studied.

3. Basic Physics and Economics Studies

Some of the problems that will be studied are:

- distribution of plutonium in reactor core, uniformly, in zones, or in spikes,
- rejection of plutonium containing larger amounts of higher isotopes,
- effect of various design parameters (type and temperature of moderator, etc.),
- different basic fuel cycles and combinations thereof.

Development of calculational methods and measurements on plutonium bearing elements are needed to support this.

4. Design Studies

To investigate the problems associated with the operation on the plutonium cycle design studies for specific reactors will be undertaken. Fuel elements design, core arrangements and control problems should particularly be looked into.

PREPARED DISCUSSION, SESSION IITHE EURATOM-AEC PLUTONIUM RECYCLE PROGRAM
AT VALLECITOS ATOMIC LABORATORY

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I. INTRODUCTION

It is generally recognized that the most effective use of plutonium as a power reactor fuel will require a thorough understanding of the fuel-cycle physics of plutonium, and that one principal avenue to such understanding is the irradiation of plutonium-bearing fuels to high burnups in appropriately selected power reactor lattices. Such long-term irradiations, accompanied by thorough assays of the fuel, are considered necessary to provide final confirmation of analytical models designed to describe the detailed course of fuel burnup. In the Vallecitos "Program for the Development of Plutonium Recycle for Use in Light Water Moderated Reactors", a long-term burnup study is in progress in which (a) burnup is accomplished in an operating light-water-moderated power reactor, and (b) the fuel is enriched in plutonium of an isotopic composition intermediate between "young" plutonium and mature recycle fuel of near-equilibrium composition.

The importance of obtaining burnup data in light water (in addition to data being obtained in other moderators) is suggested by the cross section curves shown in Figure 1. These curves emphasize the complications which the large plutonium resonances create for the analyst who attempts to apply non-hydrogenous burnup experience to fuel cycle analysis in light water. These cross section curves further suggest that individual isotopic reaction rates (and, consequently, the course of evolution of the isotopic composition) are quite sensitive to the local slow neutron spectrum. This spectrum dependence is given particularly close attention in the Program.

II. IRRADIATION PROGRAM

The plutonium-enriched fuel is being burned in the bottom quarter of one of the fuel elements in the Vallecitos Boiling Water Reactor, in an environment which is indicated in the plan view in Figure 2. The four sides of this element are adjacent to uranium fuel which is identical to that in its own upper three-quarters: 2.7 percent enriched fuel, Zircaloy clad, 16 fuel rods per element. Figure 3 is an elevation drawing of the fuel element, showing the plutonium enriched segments which were fabricated by the Plutonium Metallurgy Operation at Hanford. This fuel consists of 1.5 weight percent PuO_2 in natural UO_2 , and absorbs thermal neutrons at

FIGURE 1**IRRADIATION POSITION** FIGURE 2

approximately the same rate as the surrounding uranium fuel. The plutonium contains about 17 percent Pu^{240} and 3 percent Pu^{241} , which is similar to typical compositions expected in recycled plutonium which is still remote from a pseudo steady-state composition. Preliminary analysis indicates that the isotopic composition of this fuel will change substantially during the 10,000 MWD/ton irradiation planned.

PLUTONIUM RECYCLE FUEL ELEMENT

FIGURE 3

The mathematical analysis of the experiment is simplified by the fact that all of the plutonium-bearing fuel is located well below the boiling boundary in this region of the reactor.

III. CONSIDERATIONS OF SPECTRUM

Reference has been made to the sensitivity of reaction rates and burnup behavior to the precise shape of the slow neutron spectrum. On the experimental side, concern for this problem led to the inclusion within the fuel element of guide tubes for flux-wire thimbles, as illustrated schematically in Figure 4. By means of thimbles which penetrate the reactor pressure vessel, flux wires may be inserted into and withdrawn from the fuel element during full-power operation of the reactor.

FIGURE 4

At the present time, wires containing Lu, Eu, Ir, In, Au, W, Se, and La are being activated within the fuel element, and since each of these materials possesses a single, dominant, low-energy activation resonance, the activation rates in these wires will provide a check on the calculated neutron spectrum.

Some program theoretical effort has been devoted to a study of the possible effects on spectrum of inelastic scattering of slow neutrons by the molecular levels of UO_2 . The fact that no significant effects are apparent in criticality measurements on pure uranium oxide fuel, is not necessarily adequate grounds for dismissing the effect here, especially in view of the close approximation to "1/V" of the U^{235} absorption cross section.

IV. SUMMARY OF PRESENT STATUS

The center of the plutonium-enriched fuel has attained a burnup estimated at about 2,000 MWD/ton, and the first fuel sample has been removed within the past month. Burnup measurements and mass-spectrographic assays will begin soon on specimens taken at several radial locations within the fuel rod (pellet). Sets of resonance wires have been activated in three of the five locations within the fuel element. The raw data are qualitatively reasonable. Further activations are planned for the near future, together with detailed comparisons between observed and predicted activation rates in the resonance wires.

THE PHYSICS OF PLUTONIUM IN FAST REACTORS

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INTRODUCTION

The term "fast reactor" is a broad one, covering a host of reactor types and sizes. Their common attribute is the absence of moderating material deliberately introduced to lower the neutron energy. The sodium, oxygen or carbon present do affect the neutron energy spectrum, but their moderating power is too small to reduce neutron energies to thermal values before capture or leakage occurs. In a very small, high metal density fast reactor, inelastic scattering will reduce the average fission neutron energy of about 2 Mev only to several hundred kilovolts before fission or leakage occurs. In a very large, ceramic-fueled, fast reactor, on the other hand, most fission events in the Pu^{239} may occur at energies between 1 and 100 kilovolts, and some neutrons will be found in the few electron volt range.

All these systems differ from thermal reactors in that the cross-sections for fission at these energies are ~ 100 times smaller than at thermal energies. But, the high concentration of fissionable atoms needed to maintain criticality is what permits the use of large quantities of say, steel, in the core with only slight effect on the breeding ratio. Per atom the ratio of capture in iron to fission in plutonium is similar for fast and thermal reactors. The effect on breeding then depends primarily on the relative numbers of atoms of steel and plutonium; fifty times as much plutonium per cc of fast reactor core permits the use of fifty times as much steel.

Generally speaking, fission and capture cross sections are slowly varying in this energy range. There are no huge capture resonances, hence no special reactivity effects associated with xenon and samarium. Excepting the Doppler effect, the spectrum is not directly affected by changes in temperature, if densities remain essentially constant. Reactors are relatively small, and most changes in reactivity with rising temperature are associated with changes in leakage probability, due to sodium and fuel expansion, or small motions of the fuel.

The objective of this paper is to review the physics of the behavior of plutonium in medium and large fast power reactors. First, the relative reactivity worth and breeding potential of the various plutonium isotopes will be deduced from an examination of the pertinent cross sections. This will lead into an examination of the performance of a broad range of fast reactors as a function of size, fuel type, structural material and plutonium isotopic composition. Following a perusal of long term reactivity effects, the two crucial dynamic parameters, Doppler effect and sodium reactivity

coefficient, will be discussed. The transition from initial to equilibrium plutonium composition on core recycle will be treated next, followed by some aspects of blanket design. The review of steady state characteristics will close with a discussion of the uncertainties inherent in theoretical prediction of fast reactor performance, today.

The last section of the paper will deal with selected kinetics and safety considerations in plutonium-fueled fast power reactors. Delayed neutron questions and the variation in safety problem with fuel element type will receive primary attention.

BASIC CROSS SECTIONS

The fission cross sections for Pu^{239} and Pu^{241} are plotted in Figure 1 as a function of neutron energy. The cross sections are rather flat at high energies, rising slowly below 100 kev. Pu^{241} is seen to have a considerably higher cross section than Pu^{239} at low energies. The fission cross sections for Th^{232} , U^{238} , Pu^{240} , and Pu^{242} are compared in Figure 2. The two plutonium isotopes both have a lower threshold energy for fission and a much higher cross section than U^{238} . Notice that thorium has a very high threshold and a very small cross section, so that fission in thorium will be small.

FIGURE 1

Fission Cross Section of Pu^{239} and Pu^{241}

FIGURE 2

Fission Cross Section of Th^{232} , U^{238} , Pu^{240} , and Pu^{242}

The capture cross section of Pu^{239} is presented in the form of alpha ($= \sigma_{\text{capture}} / \sigma_{\text{fission}}$) versus neutron energy in Figure 3. Alpha, which has a major effect on breeding potential, is seen to fall off considerably around 100 kev. Hence, one can expect considerable differences in breeding ratio for reactors with fluxes lying primarily above or below this energy. Essentially no data exists on neutron capture in the higher plutonium isotopes, and performance calculations must rely on rather shaky estimates¹ for these parameters.

FIGURE 3

Ratio of Capture to Fission in Pu^{239}

The number of neutrons emitted per fission, ν , rises with energy, as is demonstrated for Pu^{239} in Figure 4. A similar behavior is expected for all fissionable isotopes. But the number of neutrons emitted per fission varies from isotope to isotope, as is illustrated in Table I for a specific neutron energy, 1.5 Mev. Pu^{241} gives slightly more neutrons than Pu^{239} , while Pu^{240} and Pu^{242} exceed U^{238} considerably.

FIGURE 4

Neutrons Emitted per Fission for Pu^{239}

TABLE I

NEUTRONS PER FISSION FOR VARIOUS ISOTOPES AT 1.5 Mev

 $\bar{\nu}$ AT 1.5 Mev

U^{238}	2.65
Pu^{239}	3.07
Pu^{240}	2.93
Pu^{241}	3.16
Pu^{242}	2.93

A direct consequence of these cross sections is that Pu^{241} is more reactive than Pu^{239} , and that Pu^{240} and Pu^{242} are more reactive than U^{238} . The parameter $(\nu\sigma_f - \sigma_f - \sigma_c)$, which provides a partial measure of the relative reactivity worths of these isotopes, has been plotted for the two thermally fissionable isotopes in Figure 5, and for the three threshold-fissionable isotopes in Figure 6. Of course, below the threshold, the parameter is negative ($= -\sigma_{\text{capture}}$). The same parameter has been averaged over the core spectrum for four reactors having widely different spectra. The results listed in Table II again demonstrate the greater reactivity of the higher isotopes. The rapid variation with reactor spectrum for threshold fissionable materials is obvious. The apparent large increase in reactivity of Pu^{241} with lower neutron energies may not be real; capture cross sections are entirely hypothetical for this isotope.

FIGURE 5

Reactivity Parameter $(\nu\sigma_f - \sigma_f - \sigma_c)$, for Pu^{239} and Pu^{241}

FIGURE 6

Reactivity Parameter $(\nu\sigma_f - \sigma_f - \sigma_c)$, for U^{238} , Pu^{240} , and Pu^{242}

TABLE II

AVERAGE REACTIVITY PARAMETER $(\nu\sigma_f - \sigma_f - \sigma_c)$
FOR VARIOUS ISOTOPES IN DIFFERING SPECTRA

Reactor Type Spectrum	U^{238}	Pu^{239}	Pu^{240}	Pu^{241}	Pu^{242}
EBR-II	.033	3.467	1.045	3.995	.931
FERMI	-.064	3.391	.667	4.251	.581
800L METAL	-.188	3.441	.264	5.136	.184
1500L OXIDE	-.190	3.442	.215	5.24	.147

The breeding ratio of any particular reactor can be expressed by the formula

$$\text{Breeding Ratio} = \frac{\nu - 1 - \alpha - A - L + (\nu^1 - 1) F}{1 + \alpha}$$

where the numerator measures the number of neutrons left for breeding purposes (by capture in fertile material) per absorption (fission plus capture) in the thermally fissionable isotope.

A = parasitic capture in non-fissile and non-fertile materials

L = leakage of neutrons from the blanket

F = fissions in fertile material, giving off ν^1 neutrons per fission.

The term $\frac{\nu - 1 - \alpha}{1 + \alpha}$ then measures the breeding potential of the primary fissionable material. The term $(\nu^1 - 1) \sigma_{\text{fission}}$ measures the "fast fission bonus" to breeding from the fertile material. These terms, averaged over the spectra of four different reactors, are compared for the appropriate isotopes in Table III. One sees that Pu²³⁹ and Pu²⁴¹ have a considerably greater breeding potential than U²³³ and that U²³⁵ is well below the latter. All the isotopes show some loss in breeding potential as the neutron spectrum is lowered. Among the fertile materials, Pu²⁴⁰ and Pu²⁴² can provide a very much greater "fast fission bonus" than U²³⁸, while that for thorium is tiny.

TABLE III
BREEDING POTENTIAL OF VARIOUS ISOTOPES
IN DIFFERING SPECTRA

Reactor Type Spectrum	$(\nu - 1 - \alpha) / 1 + \alpha$				$(\nu^1 - 1) \sigma_f$			
	Pu ²³⁹	Pu ²⁴¹	U ²³³	U ²³⁵	U ²³⁸	Pu ²⁴⁰	Pu ²⁴²	Th
EBR-II	1.785	1.862	1.438	1.173	.187	1.349	1.302	.034
FERMI	1.707	1.810	1.389	1.123	.116	1.007	.978	.021
800L METAL	1.559	1.755	1.329	1.041	.081	.745	.724	.014
1500L OXIDE	1.546	1.748	1.321	1.032	.087	.709	.689	.015

PERFORMANCE CHARACTERISTICS

The performance of a broad range of large fast power reactors was computed by Yiftah and Okrent² in 1960, using a newly developed, multi-group cross section set.¹ Some representative results are reproduced herein to demonstrate performance trends. The calculations were all made in spherical geometry on reactor systems having typical compositions. The breeding ratio has been defined as the ratio of thermally fissionable isotopes made to those used, and the breeding ratio and critical mass of thermally fissionable isotopes are for the new, clean just-critical reactor. The details of geometry, composition and density are given in Table IV.

TABLE IV
REACTOR SPECIFICATIONS

<u>Core Volume Fractions:</u>	Fuel and Fertile Material	0.25
	Structural Material	0.25
	Sodium	0.5
<u>Fuels:</u>	Density, g/cc	
Pu-U ²³⁸	19	
PuO ₂ -UO ₂	8.4	
PuC-UC	11.39	
<u>Plutonium Composition:</u>	<u>Atom Percent</u>	
Type	Pu ²³⁹	Pu ²⁴⁰ Pu ²⁴¹ Pu ²⁴²
A	100	0 0 0
B	74.7	10.2 12.4 2.7
C	40	10 25 25
<u>Blanket Thickness:</u>	45 cm	
<u>Blanket Volume Fractions:</u>	U ²³⁸	0.6
	Steel	0.2
	Sodium	0.2

The critical mass, breeding ratio, internal breeding ratio and average α Pu²³⁹ are listed for metal, oxide and carbide reactors as a function of core volume in Table V. All reactors are assumed to have 25% by volume fuel mixture and 25% stainless steel in the core. The plutonium is pure Pu²³⁹. The critical masses rise with reactor size, while the breeding ratios fall off slightly due to a rise in α Pu²³⁹ with a degradation in average neutron energy. The absence of a moderator like oxygen or carbon and the higher concentration of a strong absorber like U²³⁸ both cause the metal-fueled reactor to have fewer neutrons in the low energy range. As a consequence, the average α is lower for the metal fuel. This factor, plus the larger fast fission bonus which accompanies a higher concentration of U²³⁸, leads to a considerably higher breeding ratio for metal-fuels, other things being equal.

TABLE V
PERFORMANCE CHARACTERISTICS FOR REACTORS
USING STEEL STRUCTURE, SODIUM COOLANT AND PURE Pu²³⁹

Reactor Core Volume (liters)	Fuel	Critical Mass (kg)	Breeding Ratio	Internal Breeding Ratio	α Pu-239 ($\frac{\text{Capture}}{\text{Fission}}$)
800	Metal	431	1.82	0.73	0.19
	Oxide	372	1.55	0.31	0.23
	Carbide	396	1.62	0.46	0.22
1500	Metal	686	1.79	0.91	0.20
	Oxide	562	1.47	0.44	0.25
	Carbide	613	1.56	0.61	0.24
	U ²³³ -C, Th-C	616	1.24	0.52	0.10
2500	Metal	1025	1.76	1.04	0.21
	Oxide	806	1.42	0.54	0.27
	Carbide	897	1.52	0.73	0.25

For purposes of illustration, one U^{233} -Th-C reactor of 1500 liters volume has been included in the table. The critical mass is about the same as for the corresponding Pu- U^{238} -C reactor, but the breeding ratio is much lower. This drop in breeding can be attributed only in part to the somewhat reduced breeding potential of U^{233} . The major cause lies in the sharp reduction in fast fission bonus obtained with thorium. A typical plutonium-fueled reactor in which thorium is substituted for the U^{238} can be expected to suffer a reduction in breeding ratio of 0.2 or more.³

The effect on performance of higher isotopes of plutonium is illustrated in Table VI. Reactors having a core volume of 1500 liters are compared for three hypothetical plutonium fuels: Pu-A, which is pure Pu^{239} ; Pu-B, which has 10% Pu^{240} , 12% Pu^{241} , and 2.7% Pu^{242} ; and Pu-C, whose composition is 40-10-25-25. As we were led to expect from the examination of basic cross sections above, the breeding ratio rises as the amount of higher isotopes is increased and the critical mass, measured in terms of thermally fissionable isotopes, decreases.

TABLE VI
PERFORMANCE CHARACTERISTICS FOR 1500 LITER REACTORS
USING Pu-A, Pu-B OR Pu-C

	Pu-A		Pu-B		Pu-C	
Reactor Fuel	Critical Mass	Breeding Ratio	Critical Mass	Breeding Ratio	Critical Mass	Breeding Ratio
Metal	686	1.79	635	1.90	552	2.08
Oxide	562	1.47	514	1.57	442	1.72
Carbide	613	1.56	562	1.67	483	1.82

Stainless steel was taken as the cladding and structure in all the preceding examples. While it is true that there are no large capture resonances in the fast neutron energy range, cross sections do vary, and in a fairly systematic fashion from element to element. Usually, elements having a larger atomic number capture more neutrons. The capture cross sections of three important structural elements, iron, niobium and tantalum, are plotted in Figure 7. They substantiate this trend (in a limited fashion). The capture cross section of U^{238} has been included for comparison purposes.

FIGURE 7

Capture Cross Section for Fe, Nb, Ta, and U²³⁸

The detailed effects of various structural materials on reactor performance are illustrated in Table VII for an 800 liter core volume, metal-fuel reactor. Steel, titanium, zirconium, and vanadium can generally be grouped together. They have the least deleterious effect on performance. Niobium and molybdenum form a middle category, while tantalum is very bad. To maintain good breeding in the presence of large amounts of tantalum, one must use small reactors having a high concentration of fissionable isotopes. In this way the parasitic capture per fission is reduced.

TABLE VII

**PERFORMANCE CHARACTERISTICS FOR 800 LITER METAL-FUEL
REACTORS WITH VARIOUS STRUCTURAL MATERIALS**

<u>Structural Material</u>	<u>Critical Mass</u>	<u>Breeding Ratio</u>
Stainless Steel	431	1.82
Ti	425	1.92
V	456	1.68
Zr	415	1.89
Nb	494	1.51
Mo	502	1.46
T	716	1.02

The detailed capture cross sections of all the fission products have not been measured, but one can make theoretical estimates thereof. The effect of fission products on breeding ratio and critical mass⁴ are listed in Table VIII for typical metal, oxide and carbide fueled reactors of 1500 liter core volume. In each case the same number of fission products, 0.0008×10^{24} atoms per cc, have been added to the core. This many atoms would accumulate if 3-1/3% of the combined Pu and U²³⁸ had fissioned in the metal reactor (or 33,000 Mw days per ton). The corresponding heavy atom burnup is 8.6% for the oxide, 5.8% for the carbide. As one sees, the effect on breeding of this considerable fission product accumulation is small, though not negligible.

TABLE VIII
EFFECT ON FISSION PRODUCTS (0.0008×10^{24} atoms per cc)
ON PERFORMANCE OF 1500 LITER REACTORS

Reactor Type	<u>Critical Mass</u>		<u>Breeding Ratio</u>	
	New Reactor	With Fission Products	New Reactor	With Fission Products
Metal	686	700	1.79	1.74
Oxide	562	592	1.47	1.39
Carbide	613	630	1.56	1.47

The performance of reactors lacking any internal breeding has not been examined herein. Enriched-oxide or metal dispersions within an inert matrix of stainless steel would fall into this category. In large sizes, reactors of this type will generally have much lower critical masses and very much lower breeding ratios.⁵ In very large reactors the capture of U²³⁸ causes it to lose reactivity, hence the reduction in critical mass for systems without it. But, there is a nearly complete loss of fast fission bonus (a small blanket contribution remains) and the parasitic capture in the plutonium rises appreciably. The result can be a drop of 0.6 or more from the breeding ratio of the usual metal-fueled system.

LONG TERM REACTIVITY EFFECTS

The long term reactivity losses due to net burnup of fuel and fission product accumulation in the core, and the reactivity gain due to buildup of plutonium in the blanket have been listed for three typical reactors in Table IX.⁴ Heavy atom burnups of 5% for the metal fuel and 10% for the oxide and carbide have been arbitrarily chosen for purposes of illustration. The major reactivity loss lies in net core burnup for all three reactors. The loss is huge for the oxide and carbide since the chosen compositions yield relatively low internal breeding ratios. The fission product buildup has only a moderate effect on reactivity and is approximately balanced by the gain associated with blanket plutonium buildup. Changes in plutonium isotopic composition affect the long term reactivity loss by changing the net core burnup of fuel. The loss is generally greater for pure Pu²³⁹ than for the equilibrium mixture obtained with recycle. The difference between the two is about the same order of magnitude as the loss due to fission product accumulation.⁴

TABLE IX

LONG TERM REACTIVITY EFFECTS

Reactor Type	Breeding Ratio	Internal Breeding Ratio	Atoms/cc of Pu + U ($\times 10^{24}$)	Burnup, % of Pu+U Fissioned	$\delta k/k$			
					Burnout in Core	Buildup in Blanket	Fission Product Buildup	Total
800 liter metal	1.82	0.73	0.012	5	-.054	+.026	-.017	-.045
1500 liter oxide	1.47	0.44	0.00467	10	-.169	+.025	-.037	-.181
1500 liter carbide	1.56	0.61	0.0069	10	-.147	+.031	-.028	-.144

Even a two or three percent reactivity loss can be a considerable amount to balance with control rods in a fast reactor, however. The small core dimensions and the absence of very high absorption cross sections, coupled with complicating considerations of reactor safety, all tend strongly toward reasonable limits on the "shim" reactivity requirements. Partial reloading helps in this respect but not without costs in shutdown time. (Fortunately, there is no xenon-override problem.) But a constant reactivity core seems highly desirable--and it is not beyond reach in large fast power reactors. Internal breeding ratios can be raised simply by increasing the concentration of fertile material in the core (by increasing either the density or volume fraction of fuel element). There is relatively little change in critical mass or overall breeding ratio as a consequence. The move would be an obvious one except for the adverse affect it has on a vital dynamic characteristic, namely, the sodium reactivity coefficient, which will be discussed next.

REACTIVITY COEFFICIENT OF SODIUM

In fast reactors like the Enrico Fermi Reactor the major contributions to the temperature coefficient of reactivity come from the changes in density of coolant and fuel in the core. Both are negative effects in this reactor, that is, the reduction in density upon heating allows more neutron leakage to occur and reactivity is lost.

Actually, the reactivity effect accompanying a change in sodium density is the net result of several phenomena.

1. The effect of neutron capture in the sodium: This effect is positive but is very small in the absence of thermal neutrons.
2. Leakage component: This effect is negative. The magnitude of the effect decreases as the core size increases, since leakage is then less likely. For a given core size it is smaller for those designs having higher densities of U^{238} or other materials with large cross sections for scattering or capture. As with larger cores, the change in leakage with loss of sodium is reduced.

3. Spectral shift: Sodium degrades the neutron spectrum at all energies, but is especially effective at low energies, where it has considerable moderating power and inelastic effects of other materials are gone. While the spectrum hardening effect which accompanies the loss of sodium depends on reactor design and may have positive or negative reactivity effects at various neutron energies, the overall effect in those large plutonium-fueled reactors examined thus far has been positive. The reason for this trend is rather well illustrated in Figure 8. Here, the neutrons emitted per absorption in all core materials combined is plotted as a function of energy for 1500 liter, plutonium-carbide reactors using steel or niobium structure. This parameter, which is closely related to the reactivity worth of a neutron in a reactor having low leakage, generally rises with neutron energy. This rise is due partly to the high energy fission contribution of U^{238} ; partly to the increase with energy of $\nu(E)$, the neutrons emitted per fission in Pu^{239} ; partly due to the rapid decrease with energy of the capture cross section of U^{238} and niobium; and considerably due to the rapid decrease with energy of fission-less capture (or alpha) in Pu^{239} .

FIGURE 8

η as a Function of Neutron Energy for 1500-Liter Plutonium Carbide-Fueled Reactors with Steel or Niobium Structure

There is also a contribution of the spectral shift to the Doppler effect, but this will be discussed later.

Since the negative leakage component falls off with increasing reactor size, while the spectral shift may well become more positive, one anticipates that the sign of the sodium reactivity coefficient for a specific reactor design will change from negative to positive as reactor size is increased. This change has been observed in preliminary analyses.² For metal-fuel reactors having a higher density of U^{238} present, it occurred at smaller core volumes (~1000 liters) than for carbide or oxide systems (2000-3000 liters). When niobium was taken as the structural material rather than steel, it occurred at much reduced sizes in each reactor type. When the concentration of U^{238} present was reduced, the coefficient became more negative for a given core volume.⁴

These effects are difficult to predict accurately by the usual theoretical techniques. The individual scattering resonances of the sodium, iron, carbon, etc. must be treated in detail, and new, highly elaborate computing methods are required.⁶ Recent, more accurate calculations⁷ have found the volume threshold for the onset of a positive sodium coefficient to be considerably smaller than had been previously estimated.²

The sodium coefficient will depend strongly on the specific geometry and composition of the reactor. It will vary considerably as a function of sodium location within the core, as well, and accurate techniques for the computation of this effect remain to be developed. Also much of the basic cross section information needed to complete an accurate calculation is non-existent or inadequate.

Hence, one is faced with a marked uncertainty in the sodium coefficient of any large, fast, plutonium-fueled reactor designed today. One expects the coefficient to become more positive if the internal breeding is raised in an effort to minimize long term reactivity losses. And one lacks a satisfactory guide as to the acceptability (or lack thereof) of a positive sodium coefficient from reactor safety considerations.

It is noted that sodium reactivity coefficients seem to be negative in U^{233} -Th fueled reactors in a wide range of types and sizes.² This is primarily due to the nearly constant alpha of U^{233} , so that the variation with energy of neutrons emitted per total absorption is much flatter than shown in Figure 8. Also, thorium has essentially no high energy fission; hence, a reactor using Pu-Th fuel in metal or ceramic form should have a more negative coefficient than the same reactor using the Pu- U^{238} combination.⁷

DOPPLER EFFECTS

When the temperature of the material of a nuclear reactor is changed, there is a change in the relative velocity distribution of neutrons and nuclei, since the velocity distribution of nuclei is changed by the changed thermal motion. With increasing temperatures, the change in relative velocity has the effect of widening a cross section resonance and lowering its peak value,

keeping the average cross section unchanged. However, since the neutron flux dips at each resonance, the widening of a resonance brings into play the higher flux outside the "dip" and increases the reaction rate; i. e., increase the effective cross section. This effect is called the Doppler effect. The broadening of the fission resonances tends to increase the production of neutrons; broadening of the absorption resonances tends to increase absorption of neutrons. The effect on the reactivity of a nuclear reactor of the changed reaction rates is called the Doppler temperature coefficient of reactivity.

As is known, the EBR-I reactor with the Mark I and Mark II loadings exhibited a prompt positive power coefficient of reactivity which was thought to possibly arise from a positive Doppler coefficient of reactivity associated with increased fuel temperature.

Although later experiments demonstrated that the coefficient seen arose from bowing of the fuel rods, attempts^{8, 9, 10} were made to calculate the Doppler coefficient of reactivity, and experiments were undertaken to measure the effect.¹¹ The results of both calculation and experiment indicated that the Doppler coefficient was so small as to be negligible in EBR-I.

As the interest in fast reactors developed, however, the interest in larger, more dilute cores, with attendant softer spectra, increased the interest in Doppler reactivity coefficients. This is particularly true when, as just discussed, calculation indicated the possibility of positive sodium coefficients for large systems.

Nicholson,¹² Greebler,¹³ et al., and Fossoul¹⁴ and Bhide,¹⁵ et al., have calculated these effects for a variety of cores. Nicholson has shown, with approximations, that the Doppler effect can be described by:

$$T^{3/2} \frac{\partial K_E}{\partial T} = \frac{A^{1/2}}{4 \nu \sqrt{2 \pi K}} \frac{\rho^2 \phi_E Q_E}{\langle \mu_t \rangle_E E^{1/2}}$$

where

- T = temperature °K
- A = atomic weight
- ν = number of neutrons emitted per fission
- K = Boltzman constant
- ρ = atomic density, atoms/cc
- $\langle \mu_t \rangle_E$ = total cross section
- E = neutron energy
- ϕ_E = neutron flux, normalized to give a fission rate of unity.

The quantity $Q(E)$, defined by Nicholson's equation 3.20, has been calculated for U^{235} and U^{238} in Reference 12 and for Pu^{239} in Reference 14. $Q(E)$ is shown in Figure 9. It can be seen that $Q(E)$ increases very rapidly at lower neutron energies. Thus, calculation of the correct energy distribution at the low energy end of the neutron spectrum is of great importance for calculation of the Doppler effect. Greebler, for a large oxide reactor with 33% fuel, 17% steel, and 50% sodium, with about 7 to 1 U^{238} to Pu^{239} , calculates about 80% of the fission power generated by neutrons above 9 Kev, but only 10% of the Doppler coefficient deriving from this range. About 75% of the Doppler coefficient is contributed by the energy region from 300 to 4000 ev. Plutonium contributed 40% to the coefficient, although it was only 11% of the fuel.

FIGURE 9

The Function $Q(E)$ for U^{235} , U^{238} , and Pu^{239}

Also, from the graph, comparing U^{235} and Pu^{239} , it appears that the $Q(E)$ function is about two times as large for plutonium as for uranium. However, in comparing U^{235} and Pu^{239} fueled reactors of essentially the same type, the factor $\rho^2/\nu < \mu_t >$ will be considerably less for plutonium.

Hence, it would appear that the positive contribution to the Doppler coefficient from the fuels U^{235} and Pu^{239} will not be much different.

It remains to quote some examples of Doppler coefficient calculations for more typical fast reactors. The coefficient will, of course, depend strongly on the spectrum. Some are shown in Table X. Details of the calculation may be found in References 12, 13, and 15.

TABLE X
CALCULATED DOPPLER COEFFICIENTS

Reactor	Fuel	U-238/fuel Ratio	Doppler Coefficient $\frac{dK}{dT} / ^\circ C \times 10^6$	Ref.
<u>Metal</u>				
EBR-II	U-235	~ 1	+0.4 at 300°K	13
Fermi (Core A)	U-235	~ 2.7	-1.7 at 561°K	12
<u>Large Oxide</u>				
50% Na, 17% SS, 33% Fuel	Pu	~ 7	-9.3 at 1170°K	13
52% Na, 16% SS, 32% Fuel	Pu	7	-17. at 300-750°K	15
52% Na, 16% SS, 32% Fuel	Pu	9	-24. at 300-750°K	15
<u>Large Carbide</u>				
52% Na, 18% SS, 30% Fuel	Pu	7	-12. at 300-750°K	15
52% Na, 18% SS, 30% Fuel	Pu	9	-18. at 300-750°K	15

It is obvious that the Doppler effect can be of extreme importance in the behavior of fast power reactors. There must be experimental verification of the magnitudes of the effects before fast power reactors can be designed with confidence.

PLUTONIUM RECYCLE EFFECTS

The equilibrium plutonium isotopic composition for continuous recycling of the core fuel will depend only on the reactor design and the feed material used to replenish core burnout. The initial composition will influence the time needed to get close to equilibrium but not the final composition.

The equilibrium composition upon recycle has been computed for most of the reactors discussed earlier in the section on reactor performance, assuming the feed plutonium to be the same as that initially present in the core.⁴ Representative results are given in Table XI. When the feed material is Pu-A (pure Pu²³⁹), very little Pu²⁴² has a chance to build up, and only about 5% of Pu²⁴¹ is present at equilibrium for all the cases considered. When the feed material has a large proportion of higher isotopes, the final constitution depends sharply on the internal breeding ratio. Large internal breeding adds primarily Pu²³⁹, and less of the feed material heavy in higher isotopes is needed. Thus, the metal-fuel core with larger internal breeding has less Pu²⁴¹ and Pu²⁴² at equilibrium than the oxide for the case of Pu-C feed.

TABLE XI

EQUILIBRIUM COMPOSITION OF FUEL WITH PLUTONIUM RECYCLE

<u>Reactor Type</u>	<u>Type of Feed Pu</u>	<u>Pu-239</u>	<u>Pu-240</u>	<u>Pu-241</u>	<u>Pu-242</u>	<u>Atoms Pu</u> <u>Atoms U-238</u>	<u>Atoms Pu</u> <u>Atoms U-238</u>
						<u>at Equil.</u>	<u>Initially</u>
800 Liter Metal	Pu-A	.653	.293	.041	.013	.172	.128
800 Liter Metal	Pu-C	.544	.281	.066	.109	.189	.163
800 Liter Oxide	Pu-A	.609	.324	.049	.018	.510	.336
800 Liter Oxide	Pu-C	.288	.266	.121	.325	.795	.439
1500 Liter Oxide	Pu-A	.582	.343	.054	.021	.391	.253
1500 Liter Oxide	Pu-B	.481	.372	.088	.059	.425	.269
1500 Liter Oxide	Pu-C	.329	.287	.109	.275	.542	.323

In all of the examples chosen, the fraction of total plutonium in the fuel was greater at equilibrium than initially.

The detailed stepwise approach to equilibrium plutonium composition has also been followed for a number of reactors.⁴ The results are summarized in Table XII for an 800 liter metal-fuel reactor using Pu-A, in Table XIII for a 1500 liter oxide reactor using Pu-A, and in Table XIV for a 1500 liter oxide reactor using Pu-C. The reactors are critical on the average during a cycle in this calculation and the heavy atom burnout is taken as 5% for the metal, 10% for the oxide. To attain 80% of the total change in Pu²³⁹ concentration requires 8-10 cycles. For each cycle the initial, final and average composition are given.

TABLE XII
STEPWISE APPROACH TO EQUILIBRIUM
800 LITER METAL Pu-A FEED

<u>Cycle No.</u>		<u>U-238</u>	<u>Pu-239</u>	<u>Pu-240</u>	<u>Pu-241</u>	<u>Pu-242</u>
1	Initial	.881	.119	0	0	0
	Final	.837	.105	.007	.000	.000
	Average	.859	.112	.004	.000	.000
2	Initial	.876	.117	.007	.000	.000
	Final	.832	.104	.013	.000	.000
	Average	.854	.110	.010	.000	.000
4	Initial	.868	.112	.018	.001	.000
	Final	.825	.100	.022	.002	.000
	Average	.846	.106	.020	.001	.000
7	Initial	.861	.107	.030	.003	.000
	Final	.818	.097	.031	.004	.000
	Average	.839	.101	.030	.003	.000
12	Initial	.855	.102	.037	.005	.001
	Final	.812	.094	.038	.005	.001
	Average	.832	.098	.038	.005	.001
20	Initial	.851	.100	.042	.006	.001
	Final	.809	.092	.042	.006	.002
	Average	.830	.096	.042	.006	.001
30	Initial	.851	.099	.042	.006	.002
	Final	.808	.091	.043	.006	.002
	Average	.829	.095	.043	.006	.002

TABLE XIII
STEPWISE APPROACH TO EQUILIBRIUM
1500 LITER OXIDE, Pu-A FEED

<u>Cycle No.</u>		<u>U-238</u>	<u>Pu-239</u>	<u>Pu-240</u>	<u>Pu-241</u>	<u>Pu-242</u>
1	Initial	.767	.233	0	0	0
	Final	.712	.167	.021	.001	.000
	Average	.739	.197	.012	.0004	.000
2	Initial	.752	.227	.021	.001	.000
	Final	.697	.163	.037	.003	.000
	Average	.724	.192	.030	.002	.000
4	Initial	.731	.214	.050	.005	.000
	Final	.678	.154	.060	.007	.001
	Average	.704	.182	.056	.006	.001
7	Initial	.713	.201	.074	.010	.002
	Final	.662	.146	.079	.011	.002
	Average	.687	.171	.077	.010	.002
12	Initial	.702	.192	.089	.014	.004
	Final	.651	.141	.090	.014	.004
	Average	.676	.164	.090	.014	.004
20	Initial	.696	.189	.094	.015	.005
	Final	.647	.139	.094	.015	.005
	Average	.672	.162	.095	.015	.005
30	Initial	.696	.188	.095	.015	.006
	Final	.646	.138	.095	.015	.006
	Average	.671	.161	.095	.015	.006

TABLE XIV
STEPWISE APPROACH TO EQUILIBRIUM
1500 LITER OXIDE, Pu-C FEED

<u>Cycle No.</u>		<u>U-238</u>	<u>Pu-239</u>	<u>Pu-240</u>	<u>Pu-241</u>	<u>Pu-242</u>
1	Initial	.716	.114	.028	.071	.071
	Final	.666	.098	.034	.039	.063
	Average	.691	.105	.032	.053	.067
2	Initial	.691	.128	.042	.058	.081
	Final	.644	.105	.046	.033	.071
	Average	.668	.116	.044	.044	.076
4	Initial	.661	.135	.063	.047	.093
	Final	.617	.109	.064	.030	.081
	Average	.638	.121	.064	.038	.087
7	Initial	.638	.132	.083	.045	.102
	Final	.596	.106	.080	.030	.088
	Average	.616	.118	.082	.037	.094
12	Initial	.624	.127	.096	.046	.106
	Final	.583	.102	.092	.031	.091
	Average	.603	.114	.094	.038	.099
20	Initial	.619	.125	.102	.047	.107
	Final	.578	.101	.096	.032	.093
	Average	.598	.112	.099	.039	.100
30	Initial	.618	.124	.103	.047	.108
	Final	.578	.101	.097	.032	.093
	Average	.598	.112	.100	.039	.100

It is assumed that no reprocessing loss of plutonium occurs between cycles in these calculations. However, a 2% loss of plutonium from the main stream every cycle has little effect on the equilibrium concentrations, as can be seen in Table XV.

TABLE XV
EQUILIBRIUM COMPOSITION WITH REPROCESSING LOSS

<u>Reactor Type</u>	<u>Feed Material</u>	<u>% Loss per Cycle</u>	<u>U-238</u>	<u>Pu-239</u>	<u>Pu-240</u>	<u>Pu-241</u>	<u>Pu-242</u>
1500 Liter Oxide	Pu-A	0	.708	.170	.100	.016	.006
1500 Liter Oxide	Pu-A	2	.714	.174	.093	.014	.005
1500 Liter Oxide	Pu-C	0	.631	.118	.105	.041	.105
1500 Liter Oxide	Pu-C	2	.638	.119	.099	.041	.103

A comparison of initial and equilibrium critical mass and breeding ratio is given in Table XVI for the 1500 liter, oxide-fuel reactor using Pu-A, B or C respectively as the initial fuel and the feed. ⁴ In all three cases the critical mass was lowered and the breeding ratio increased on the transition to an equilibrium composition. It is interesting to note that the breeding ratio for equilibrium composition with Pu-C feed is 0.49 above that for a reactor using pure Pu²³⁹, while the critical mass is only 3/4 as large. The large fast fission bonus in Pu²⁴⁰ and Pu²⁴² leads to this large difference in performance.

TABLE XVI
COMPARISON OF INITIAL AND EQUILIBRIUM CRITICAL MASS
AND BREEDING RATIO FOR 1500 LITER OXIDE REACTORS

<u>Plutonium Feed</u>	<u>Critical Mass*</u>		<u>Breeding Ratio**</u>	
	<u>Initial</u>	<u>Equil.</u>	<u>Initial</u>	<u>Equil.</u>
A	562	496	1.47	1.67
B	514	468	1.57	1.80
C	442	421	1.72	1.96

* Kg of (Pu-239 + Pu-241)

** (Pu-239 + Pu-241 produced) / (Pu-239 + Pu-241 consumed)

However, not all qualities of a reactor improve with the change in plutonium isotopic composition on recycle. The sodium reactivity coefficient has been followed during the transition to equilibrium composition for 1500 liter, oxide-fueled reactors using Pu-A or Pu-C feed, and in both cases the coefficient became appreciably less negative. The values for sodium coefficient listed in Table XVII are given in terms of the change from the critical fuel loading which would produce the same reactivity effect as removing 40% of the core sodium. It is interesting to observe that the Pu-C reactor starts more negative than the Pu-A reactor, but undergoes a considerable greater positive increment and ends up less negative. It is also noteworthy that the change during a cycle initially is great enough to produce measurable effects on kinetics experiments.

TABLE XVII
VARIATION IN SODIUM COEFFICIENT WITH RECYCLE
IN OXIDE REACTORS

<u>Feed Material*</u>	<u>Cycle No.</u>	<u>$\Delta M_c / M_c$</u>
Pu-A	1	-.0115
	3	-.0091
	6	-.0077
	9	-.0069
	12	-.0064
	Equil.	-.0061
Pu-C	1	-.0130
	3	-.0074
	6	-.0041
	8	-.0032
	10	-.0028
	Equil.	-.0021

*Initial Pu composition same as Feed
1500 liter reactors

Yet another parameter can incur changes sufficiently large during one burn-out cycle to produce measurable effects on kinetics experiments, namely the delayed neutron fraction. Calculations show that a change of between two and ten percent can occur with 10% heavy atom burnup, depending on the original composition.⁴ As shown in Table XVIII, the delayed neutron fraction gets larger as the plutonium isotope gets heavier. However, the delay fraction for U²³⁸ exceeds that for Pu²⁴²; hence, the change with burnout in any reactor will be peculiar to that design, and can be positive or negative.

TABLE XVIII
DELAYED NEUTRON FRACTIONS FOR FAST FISSION

<u>Fissile Nuclide</u>	<u>Delayed Neutron Fraction (β)</u>	<u>Reference</u>
Pu 239	0.0020	(a)
Pu 240	0.0027	(a)
Pu 241	0.0053	(b)
Pu 242	0.0080	(c)
U 235	0.0065	(a)
U 238	0.0147	(a)
U 233	0.0027	(a)
Th 232	0.0205	(a)

(a) Keepin, G. R. "Neutron Data for Reactor Kinetics" Nucleonics
Vol. 20, No. 8, August 1962.

(b) Reactor Physics Constants, ANL-6300, to be published

(c) Estimated

In any Pu-U²³⁸ reactor, however, one can expect an effective delayed neutron fraction greater than the 0.0020 value of pure Pu²³⁹. The estimates given in Table XIX for a variety of 1500 liter reactors using different fuels range from 0.0032 to 0.0059.⁴ These differences in delay fraction among the various reactor designs are found to be greater than the change on going from initial to equilibrium composition within any one reactor.

TABLE XIX

DELAYED NEUTRON FRACTION FOR CORE OF REACTORS
FUELED WITH Pu-A, B AND C

<u>Reactor Type</u>		<u>$\bar{\beta}_{\text{Core}}$</u>
Pu-A	Metal	.00454
	Oxide	.00326
	Carbide	.00372
Pu-B	Metal	.00501
	Oxide	.00384
	Carbide	.00427
Pu-C	Metal	.00596
	Oxide	.00491
	Carbide	.00530

It is noted that a reactor fueled with U²³³-Th will not benefit appreciably from augmentation of the delayed neutron fraction due to fission of the fertile material. As shown previously in Table III, there is very little fission in the thorium, and a delay fraction close to 0.0027 of pure U²³³ will result. The buildup of U²³⁵ on recycling will raise this fraction, but simultaneously the breeding ratio and sodium coefficient will change adversely.

SOME CONSIDERATIONS OF PRACTICAL DESIGN AND BLANKET MANAGEMENT

The performance figures presented earlier were all for idealized spherical reactors. Actual reactors of the same core volume will all have larger critical masses for a variety of reasons. Control rod channels, coolant flow transition regions, and, in particular, the transition from

spherical to cylindrical geometry may combine to raise the critical mass by 20% or more.

The overall breeding ratio should not be affected significantly by these considerations if the blanket is kept an efficient capturer of neutrons. However, various practical considerations enter herein, also. First, since the end blanket residence time is usually determined by fuel element burnup rather than optimum blanket economics, in some designs thick end blankets may prove to be uneconomical. Secondly, if the blanket fabrication and reprocessing costs are excessive or the blanket fluxes very low due to a low core power density, the outer portions of a radial blanket may prove uneconomical. Thirdly, in some designs, safety considerations relating to the obscure meltdown problem have led to a reduction of end blanket efficiency.

Design and management of the blanket in a specific reactor depend strongly on the existing economic factors, a subject left for other papers at this meeting. Some general studies¹⁶ have shown that if one could achieve blanket fabrication plus reprocessing costs of \$50/kg then, for a reactor with an average power density of one Mw/liter, all sections of an efficient blanket are economic if allowed the proper residence times. If the costs go up by a factor of three, or if the power density goes down by a factor of three, the outer portions of the blanket may become uneconomical.

In the case where flux levels are low or residence times limited, blanket thermalization with a consequent reduction in fertile material requirements may help appreciably. A blanket cycle involving inward radial motion of elements during their total residence time may sometimes improve the economics, but this depends strongly on the specific conditions present.

In an unmoderated blanket of a fast reactor, one can expect a buildup of two or three percent of Pu²⁴⁰ in the bred Pu²³⁹ for optimum cycling. Since the core physics performance is good for essentially any plutonium isotopic composition (disregarding sodium coefficient and possibly Doppler effect considerations), the reactor can be used to convert "dirty" plutonium to "clean" plutonium by feeding the core with the former from thermal reactors and extracting the latter from the blanket for use in thermal reactors, should suitable designs develop. Along these lines, interest has been expressed in the possibility of using irradiated fast reactor blanket rods directly in a thermal reactor without first reprocessing them. This procedure would introduce some reactor physics complication into the thermal reactor design, due to the non-uniform plutonium buildup in the fast reactor blanket. In addition to an axial, chopped-cosine effect, there may be significant transverse variations, particularly in blanket elements near the core.

Considerable interest has been shown in the possibility of designing large fast reactors to suffer no reactivity loss with high burnup, thus greatly simplifying control and reloading problems. Large fast reactors can be designed to give an internal breeding ratio of unity or greater, so

that the goal seems to be achievable. Unfortunately, as has been mentioned in the section on sodium reactivity coefficient, the same design features leading to a high internal breeding also lead to positive sodium coefficients. Thus, a major conflict of interest remains to be resolved.

UNCERTAINTIES IN THEORETICAL PREDICTION

While a major improvement in our knowledge of fast neutron cross sections has come about in the last several years, there remain large gaps in the available information. And, the plutonium isotopes lie among those which are less well measured. The crucial cross sections for plutonium are ν , σ_{fission} , and σ_{capture} . For Pu^{239} , all three have been measured but not sufficiently well. Neutrons per fission (ν) is known to 1-1/2 or 2% except at higher energies, while σ_{fission} is known to about 5% except at lower energies where a 10% or greater uncertainty is likely. Recent measurement of the capture cross section of this important isotope have helped narrow uncertainties therein to 20 to 25%, except in the low energy region where detailed resonance data are required. If these few uncertainties contributed cross section errors to the calculation all in the same direction, the error in reactivity would be 6 or 7% $\delta k/k$, representing a huge potential mistake in critical mass predictions from this source alone.

The higher plutonium isotopes are much more poorly known. ν and σ_{fission} have been measured over part of the energy range, but less accurately than for Pu^{239} . Neutron capture data are non-existent, forcing one to rely on estimates not good to better than a factor of two. The cross sections for elastic and inelastic scattering are also unknown, except poorly for Pu^{239} . This should not severely hamper the prediction of dilute fast reactors but does introduce errors into the analysis of integral experiments on these materials.

Most other fast reactor materials are also inadequately measured as to their fast neutron cross sections. If all these uncertainties were to add up in the same direction, the error in critical mass would be horrendous. Fortunately, if a careful, objective review of cross sections is made, the errors introduced should be partly random and tend to cancel to a considerable degree. However, considerable discrepancies can and do remain. In a recent study of this kind on the better measured U^{235} systems, in which no use of our knowledge of critical experiments was used to bias (or "bugger") the choice of cross sections,¹ errors of reactivity prediction as large as 4% $\delta k/k$ arose. While the existence of a considerable body of U^{235} -fueled critical experiments enables one to adjust calculations empirically, no similar body of information exists for plutonium.

Fortunately, the breeding ratio is fairly insensitive to the cross section uncertainties. Calculations have been made of the effect of "pessimistic" cross sections on breeding,^{4,5} with the following results:

1. Raising the inelastic cross sections led to a drop in breeding ratio of 0.05;

2. Raising the fission product capture led to a drop of 0.5 to 0.1;
3. Doubling the capture in all the higher plutonium isotopes led to a drop of 0.15-0.2.

Actually, the performance figures quoted in earlier sections are based on a 1960 cross section review.¹ The changes in plutonium cross section data since then tend to cancel in their effect on breeding. A lower alpha (or σ_{capture}) for Pu²³⁹ and a higher σ_{fission} for Pu²⁴¹ are partly balanced by lower values of ν for Pu²³⁹ and Pu²⁴⁰.

Our uncertain knowledge of cross sections will affect predictions of equilibrium composition on recycle,⁴ long term reactivity losses, Doppler effect, and sodium reactivity coefficient. The latter two dynamic reactivity coefficients are particularly sensitive, since the net effect in each is the algebraic sum of opposing contributions. Both suffer from a need of more powerful theoretical techniques. Both are strongly dependent on details of the low energy end of the spectrum, a region completely unmeasured in past fast reactor criticals. Indeed, past criticals have not been sufficiently dilute and large to make the measurement pertinent.

Estimates have been made of the sensitivity of the sodium coefficient to uncertainties in the plutonium cross sections.⁴ Errors of 50% are found to be plausible, even a change in sign for large power reactors near the threshold of a positive coefficient. Hence, physics specification of the reference design of a large, plutonium-fueled reactor is fraught with uncertainty today, particularly in regard to the crucial dynamic characteristics. Reactivity measurements in critical facilities on dilute plutonium systems should help tie down predictions of critical mass considerably, as will more accurate measurements of vital cross sections. But, Doppler and sodium coefficients are more difficult to calculate and to measure. A well planned, comprehensive theoretical and experimental program is required here. It should provide sufficient check points for an accurate theory so that detailed predictions can be made with accuracy and large, fast power reactor design can proceed with assurance.

KINETIC EFFECTS

Delayed neutron fractions β are given for the various fissile materials of interest in fast reactors in Table XVIII. As can be seen, the delayed neutron fraction, or dollar of reactivity, is small for plutonium. For breeder reactors, however, this must be modified by the contributions from the fertile material. Calculations for plutonium uranium metal fueled reactors with U/Pu ratios varying from ~ 16 to 0, with U²³⁸ blankets indicate effective β 's ranging between ~ 0.30 and 0.40.¹⁷ Th-U²³³ systems should have somewhat smaller β 's because of less fission in Th²³². Oxide and carbide fueled reactors will also have a somewhat lower effective delayed neutron fraction. In any case, the smaller effective β , coupled with the short lifetimes, varying between 10^{-6} and 10^{-8} seconds, makes it imperative that excess reactivities be kept low.

This means that reactivity additions must be made slowly and cautiously. However, since periods associated with reactivities measured in dollars are all approximately the same, control geared to cents per second should be no more difficult for plutonium-fueled reactors than for uranium-fueled reactors.

Feedback effects, i. e., expansion, density, burnup, rod motions, etc., however, will result in reactivity changes that are the same for the various fuels on an absolute scale. This means that feedbacks for plutonium-fueled reactors will appear to have coefficients 1.5 to 2 times the magnitude of comparable feedbacks in U^{235} -fueled reactors. Thus, reactor stability must be carefully analyzed. The instability threshold, if it exists, will be at lower powers. However, if the reactor is unconditionally stable, no adverse effect of the lower β will be noted.

SAFETY PROBLEMS WITH NEW FUEL ELEMENT TYPES

Most of the nuclear safety studies on fast reactors in the past have dealt with moderate sized reactors, using a one-piece, freely expanding uranium metal fuel element. The rapid shutoff coefficient with axial expansion, the absence of an appreciable positive Doppler coefficient and the presence of a negative sodium reactivity coefficient have all helped in efforts to demonstrate the safety of this reactor type. However, it is extremely unlikely that future, large, plutonium-fueled power reactors will use this type of fuel element. Undesirable swelling of the freely supported plutonium-uranium metal forces one to other alternatives, such as oxide, carbide, cermet, or constrained metal.

The substitution of plutonium, with its smaller delayed neutron fraction and possibly larger Doppler coefficient¹⁴ than U^{235} , of course, tends to make reactors less safe. Fortunately, preliminary studies of the course of typical startup and loading accidents in an EBR-II type reactor (moderate core size, freely expanding, one-piece-metal-fuel element) have shown little difference in consequences to the reactor if the usual delayed neutron fraction with U^{235} is reduced by a factor of two.¹⁸ However, one must expect that some classes of accidents will be more likely to produce serious difficulties with the lesser delay fraction. And the reactor will have less margin against resonant instabilities at high power levels.

The major new safety problems seem to be related to the increased reactor size and the changed fuel element characteristics, however. A prompt acting shutoff mechanism due to axial fuel expansion is certain with none of the new types. Surely the restrained metal fuel will lack this characteristic. Ceramic fuel may be multi-piece and subject to cracking, hence unlikely to provide rapid axial expansion in a super-prompt critical situation. The cermet remains to be analyzed and tested in this regard, but the chances do not seem good. Actually, one can postulate irradiation conditions with some ceramic fuels where a mass transport phenomenon relocates the fuel, producing a low density region near the middle region of the pin (in an axial sense). In this event, the expansion effect in a prompt burst could be autocatalytic, adding reactivity.

The problem of positive sodium reactivity coefficients associated with large core sizes has already been discussed. Design efforts for large oxide reactors have sometimes accepted the positive sodium coefficient as a necessary evil attending high internal breeding ratios, and attempted to override its effects with a large, negative Doppler coefficient.¹⁹ This approach is not without peril. One can postulate accidents, such as rapid boiling of sodium coolant from central regions of the core wherein reactivity will be gained not only from sodium loss but from the reduction of negative Doppler effect attending a hardening of the reactor spectrum.²⁰ Calculations by Hummel indicate a considerable change in Doppler effect upon loss of sodium, as much as a factor of two.¹⁵ Also, as has been pointed out, present day knowledge does not permit an accurate prediction of either Doppler or sodium coefficient.

There has been some attention given to cermet fuels consisting of highly enriched oxides of plutonium or U^{235} in an inert matrix, say steel. This fuel element type can be expected to have a positive Doppler coefficient, which may be the only prompt acting coefficient in a super-prompt critical burst. Such a condition could be fraught with danger, since a relatively small positive Doppler effect then changes any super-prompt-critical situation into an explosion.

The partly enriched cermet, on the other hand, may prove to be among the most desirable of the new fuel elements from a safety viewpoint. Enough U^{238} could presumably be present to keep the Doppler coefficient negative, but not large. This would reduce operating reactivity requirements, yet still give a prompt shut off mechanism. The reduced volume fraction of fertile U^{238} and the presence of fairly innocuous steel would help the acquisition of a negative sodium coefficient. Of course, the internal breeding would be reduced, and partial reloading to maintain reactivity would be increased. Its resonant stability characteristics remain to be analyzed, and no comment will be made thereon. However, all reactivity coefficients (neglecting bowing, a subject in itself) should be negative, including that of axial expansion, so that conditions should be favorable.

Of course, if axial expansion can be guaranteed in Pu-U-carbide plus metal-additive type fuels (or any other type, for that matter), if large amounts of strong absorbers like Nb or Mo are not needed, and if internal breeding is limited appropriately (thus helping to obtain a negative sodium coefficient) a similar happy combination of safety characteristics should be achievable.

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PLUTONIUM AS A FUEL FOR THE FAST REACTOR RAPSODIE

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I. INTRODUCTION

There seems to be no question that the ^{238}U - ^{239}Pu cycle is one of the most interesting, for the following reasons:

1. The abundance of ^{238}U in nature.
2. The availability of depleted uranium from natural uranium piles or isotope separation plants.
3. Very good value of plutonium as fuel, at least in the case of fast reactors.
4. The relative medium-term abundance of plutonium rich in isotope 240.

For these reasons we feel that on a medium and long term basis, plutonium will be the obvious choice of fuel for fast reactors. With this in mind we have proposed to use a plutonium fuel in the first of our fast power reactors. In this way we hope to obtain a certain amount of useful information on its fabrication and handling, its use in the reactors and its reprocessing, all of which will be indispensable for the future.

It should be noted that RAPSODIE is only a stage in the development of the fast neutron reactor scheme; as such it should first of all provide us with experience in the design, construction and operation of a sodium cooled fast power reactor, and secondly, serve as a testing ground in the development of different plutonium fuels liable to be used in later stages of the project.

RAPSODIE has three important characteristics as far as fuel is concerned:

1. It was designed to be capable of operation with a wide range of different fuels: metal alloys, oxides, carbides.
2. The chemical composition of this fuel must be very similar to that of the fuels for future reactors (large power plants).
3. The fast neutron flux and the cooling system correspond to specific powers which seem reasonable for future reactors.¹

It should be noted that in order to obtain the chemical composition similar to that of large power reactors we have had to use ^{235}U -enriched uranium in conjunction with the plutonium; this reduces the critical volume and therefore the total power of the pile, as well as the investment in fissile material and especially in plutonium.

The following paragraphs describe the possible fuels for RAPSODIE (oxide, metal, carbides); a few comments will be made on the respective merits of these various fuels, and the parameters characterising their neutronic and thermal properties will be given.

II. DESCRIPTION OF THE FUEL FOR RAPSODIE

A full description of the fuel is given in the paper by P. Bussy; nevertheless, for sake of convenience, a brief description will be given here.

The fuel subassemblies (Figure 1) contain the fuel pins in the centre and the fertile blanket at the upper and lower ends.

This set of fuel-pins is contained in a hexagonal stainless steel can; closed at the top by a cap which is used for handling and which contains holes to allow the passage of the sodium, and at the bottom by a foot which rests on the bed of the reactor.

The fuel pins, 37 per subassembly, are fixed at their lower end by grids (Figure 2) which are in turn fixed to an internal hexagonal sheath. The cylindrical canning is of stainless steel, wrapped around with a helical wire and closed by a head and a foot of stainless steel welded to the canning.

An expansion chamber for the sodium and the fission gases is installed above the fuel. Table I shows the dimensions of the various parts of the fuel subassembly.

TABLE I
FUEL PINS DIMENSIONS
(Inches at 20 C)

Fuel type	Alloy (61 - 1)	Oxide (62 - 1)
Rod length	15.75	13.39
Rod diameter	0.216	0.226
Nb clad O.D.	0.232	-
Nb clad thickness	0.008	-
SS clad O.D.	0.264	0.264
SS clad thickness	0.0157	0.019
Expansion space height	1.968	4.685

FIGURE 1
Fuel Subassembly

RAPSODIE

FIGURE 2
Support Grids

III. ADVANTAGES AND DISADVANTAGES OF THE VARIOUS POSSIBILITIES: CARBIDE, OXIDE, METAL (Present Situation)

Amongst the three possibilities originally open in the choice of fuel for RAPSODIE, i.e., carbides, oxides and alloys, carbides were avoided for the first loading in view of our lack of experience regarding their fabrication and their behaviour under irradiation.

In the other two cases the investigations have now reached the point where the fabrication of a set of fuel units can be considered.

Alloys

Amongst the possible solutions, i.e., the alloys of plutonium, particular attention has been paid to uranium based ternary alloys because of the possibility of breeding in the core. For this reason binary alloys such as Fe-Pu, for example, were discarded fairly early.

The ternary alloy, U-Pu-Mo was chosen originally because of the good resistance to irradiation of the binary U-Mo.

Metallurgically speaking these alloys show many similarities, but unfortunately their behaviour under irradiation is quite different.

We have made a fairly thorough study of the U-Pu-Mo ternary alloys - 36 samples in all were irradiated in thermal neutron fluxes at temperatures from 400 to 600 C, the burn-up varying between 4000 and 8000 MwD/T. The first results were published recently,² and Table II shows the swelling rates observed.

TABLE II. IRRADIATION TESTS ON U-Pu-Mo ALLOYS

Composition (weight %)			Surface Temp. (°C)	Burn up (Mwj/T)	$\frac{\Delta V}{V_0} \times 100$
U	Pu	Mo			
68	20	12	410	4,400	90.5
68	20	12	485	7,100	68.5
68	20	12	525	8,300	76.5
70	20	10	410	4,400	36
70	20	10	485	7,100	68
70	20	10	525	8,300	64
72	20	8	410	4,400	30
72	20	8	525	8,300	104.5

The extent of the swelling prohibits the use of these alloys as they stand, but this swelling can probably be reduced by the use of canning with a high mechanical resistance. Experiments are being carried out to find the maximum combustion rate admissible under these conditions.

The other disadvantage of these alloys is their poor compatibility with stainless steel. Attempts have been made to solve this problem with double sheathing rather than by the adoption of niobium canning, in view of the complications which the use of this latter would introduce in the sodium circuit.

The fuel pins will be prepared by injection casting of the alloy in niobium tubes. These tubes are narrower at each end, and will be capped with a resistance welded niobium capsule which will prevent contact with the stainless steel canning even in the case of swelling of the alloy.

Casting tests have shown that the metallurgical bonding between alloy and niobium is very good, and does not deteriorate in the course of thermal cyclings.

The niobium-sheathed rod is placed inside the stainless steel canning, the two being separated by a sodium joint which is checked for quality by means of an eddy-current detector.

Figure 3 shows the structure of the whole fuel-pin.

The double canning operation provides an additional complication which means eventually that the process of manufacture and canning is more complex for the alloys than in the case of oxides. On the other hand, if the sheathings prove to be capable of keeping the swelling down to a reasonable value, metal fuel seems very attractive because of its good heat conductivity, especially for high specific powers (RAPSODIE 20 MW).

Oxides

The method adopted for the manufacture of UO_2 - PuO_2 mixed oxide fuel is natural sintering of pellets 6 mm in diameter and 10 mm high. A workshop has been built at CADARACHE with the aim of producing the core of RAPSODIE within about four months.

- The oxides are mixed mechanically after being obtained separately:
- UO_2 by decomposition of the diuranate followed by a hydrogen reduction,
 - PuO_2 by decomposition of the oxalate.

It has been found that the sintering itself is sufficient to guarantee homogenisation; in other words, the solubilisation of PuO_2 in UO_2 . In particular, it is not necessary to resort to the coprecipitation of oxalates, which, although in fact gives excellent homogeneity at lower temperature, incurs a large preliminary chemical operation.

Sintering is an expensive method, but it seems that for a reactor where the core is relatively small and, therefore, the mass of fuel not excessive, this type of manufacture could be convenient and the cost of energy produced not prohibitive.

FIGURE 3
U-Pu-Mo Fuel Pin

The problem of heat transfer between sheathing and fuel is satisfactorily solved in the case of alloys by the use of a sodium bond, but this is not the case where oxides are concerned. Two solutions are being investigated for RAPSODIE; the first is to apply the canning to the pellets by passing through a die, the second is to use rectified pellets which are then passed into a calibrated sheath filled with helium. If the rectification does not pose any serious problems the second solution will probably be adopted, since for the first the canning must be sealed by seam welding, a method which gives unreliable welds by comparison with heliarc welding. On the other hand, the first solution provides a better thermal bonding between canning and oxide.

Various methods are under consideration to prevent the oxide pellets from rising into the expansion space, holding in place by a tube with or without gap, or holding with a spring, for example.

Uranium oxide fuel-pins of the RAPSODIE type were irradiated with thermal neutrons in the reactor EL-III at SACLAY, the heat transfer and specific power conditions being the same as those present in RAPSODIE. The behaviour of these pins was satisfactory. UO_2 - PuO_2 pins have been irradiated under the same conditions, and other irradiations are being carried out with higher burn-up and slightly different fabrication methods.

Choice of Fuel for the First Loading

At the present stage of the project the choice is still to be made, but it seems that the oxide will probably be adopted. It has the advantage of presenting fewer fabrication problems than the alloy, and in addition our experience of it is greater since we have already produced and irradiated whole fuel pins.

The alloy involves a more complex fabrication process, but its good thermal conductivity seems to us such an essential advantage that it is worth overcoming the final difficulties of manufacture and attempting to remedy the swelling.

For later sets the choice will be more difficult since they will be subjected to more rigorous conditions; in addition, a third choice will become available once carbides begin to be used.

Several types of fuel will probably be tested before the good and bad points of each are properly known. Tests on samples, generally carried out with thermal neutrons, are very inadequate and must only be considered as preliminary experiments; only tests in the reactor, which means under the true conditions of use, can permit definitive conclusions to be drawn.

IV. PHYSICAL CHARACTERISTICS OF FUEL FOR RAPSODIE

Before describing the neutronic and thermal characteristics of RAPSODIE, it is useful to note why we have chosen the chemical composition given below. Judging from research carried on abroad, and also from the discussions in the paper quoted above,¹ we feel that fast neutron plants should be large if they are to survive economically. The size of these plants

cannot be quoted accurately as yet, but we think generally in terms of constructing units of one to two thousand thermal megawatts. On the other hand, it seems that the specific power per litre of core shows an economic optimum value which varies from one fuel to another and from one plant to another, but of which the order of magnitude appears to fall below 500 kw/l. We have already mentioned these fast reactor characteristics.¹ Since then a report published by Russian authors² seems to confirm the existence of such an optimum but suggests a different reason from ours for its presence, involving the increase in cooling time of the fuel before reprocessing as a function of the specific power developed in the reactor.*

Although, we have not since had time to look into this in greater detail, we feel that the reasons produced in our report¹ and those of the Russian report should be taken together. Under these conditions we may assume that the specific power optimum will be still lower than that which we or the Russians had foreseen separately.

Let us take, as an example, a specific power of 400 kw/l and a thermal power built up in the core equal to 1600 Mw. This leads us to a core volume of 4000 litres. The curves shown (Figure 4) show the critical enrichment as a function of core volume of a typical fast power reactor, for oxide, carbide and metal fuels.

FIGURE 4

* This last comment could be untrue if a dry process could be developed (Pyrometallurgy), which would of course need to be economically worthwhile from the viewpoint both of reprocessing and refabrication.

If we examine these curves we notice that the critical enrichment expressed in ^{239}Pu atoms/(U + Pu) atom at approximately 4000 litres does not vary much, and is about:

- 8 % for the metal,
- 10 % for the carbides,
- 12 % for the oxides.

This enrichment, taking into account the isotopic composition of plutonium at equilibrium and the fact that the calculations from which they are obtained make use of simplifying hypotheses, leads to a chemical composition of the fuel, expressed in Pu atoms/(U + Pu) atom, of about:

- 23 % for the oxide,
- 15 % for the metal,
- 19 % for the carbide.

These, therefore, are the chemical compositions chosen for RAPSODIE. Table III sums up the main neutronic characteristics of the different fuel models considered:

- 61.1 - metal alloys,
- 61.2 A - oxides,
- 61.2 A' - oxides with thicker canning,
- 61.3 - carbides.

For purposes of comparison the characteristics of ^{235}U -enriched uranium fuel (without Pu) are also given: Models 61.1' and 61.2 A'.

In addition we have examined oxide fuels with more realistic isotopic compositions of plutonium than ^{239}Pu alone; Models 61.2 A bis and 61.2 A ter.

It may be useful to give some of the parameters characterising the large plutonium reactors discussed below - Tables IV, V, VI contain these characteristics.

Finally, Tables VII and VIII sum up the mechanical and thermal characteristics of the metal and oxide fuels for RAPSODIE.

To conclude this examination of the various properties of the fuel for RAPSODIE, we may note that for a relatively small reactor of this type the fact of using plutonium associated with ^{235}U or ^{235}U alone does not greatly change its neutronic characteristics.

TABLE III

RAPSDIE NEUTRONICS MODELS

MODELS		61-1	61-1'	61-2A	61-2A'	61-2ABIS	61-2ATBA	61-3	62-1
FUEL		U-Pu-Mo	U-Mo	PuO ₂ -UO ₂	UO ₂	(a) PuO ₂ -UO ₂	(b) PuO ₂ -UO ₂	PuC-UC	PuO ₂ -UO ₂
CORE COMPOSITION VOLUME %	FUEL	39.33	39.33	48.23	48.23	48.23	48.23	48.23	42.85
	STRUCTURES	22.83	22.83	18.66	18.66	18.66	18.66	18.66	23.9
	SODIUM	37.84	37.84	33.11	33.11	33.11	33.11	33.11	33.25
CORE VOLUME (LITERS)		40,714	40,714	40,682	40,682	40,682	40,682	40,682	40,682
RATIO U5 / U5+U8		0.275	0.51	0.40	0.753	0.44	0.462	0.163	0.542
MASS OF DIFFERENTS COMPOSANTS (kg)	U 235	55	124	50.5	127.5	55.5	58	29	61
	U 238	145	120	77	42	72	69	144	51
	Pu 239	40		44		39	35	57	39
	Pu 240					4.65	7.52		
	Pu 241					0.23	1.98		
	Mo	27	27						
	O			23	23	23	23		20
	C							12	
NEUTRON PROMPT LIFETIME (l)		$7.68 \cdot 10^{-8}$	$7.63 \cdot 10^{-8}$	$9.14 \cdot 10^{-8}$	$8.81 \cdot 10^{-8}$	$9.12 \cdot 10^{-8}$	$9.13 \cdot 10^{-8}$	$8.88 \cdot 10^{-8}$	$9.42 \cdot 10^{-8}$
EFFECTIVE FRACTION OF THE DELAYED NEUTRON (β_{EFF})		0.00613	0.00726	0.00609	0.00739	0.00610	0.00603	0.00556	0.00621

(a) Pu 239 89% Pu 240 10.5% Pu 241 0.5%

(b) " " 78.5% " " 17% " " 4.5%

TABLE IV
METAL

$\alpha = \text{U ATOMS} / \text{Pu ATOMS}$	0.5	4	6	9	41	12
CORE VOLUME (LITERS)	14	155	374	1070	3140	5400
Pu CRITICAL MASS (kg)	42	142	245	580	1200	1900
Pu 239 FISSIONS	0.322	0.291	0.282	0.271	0.265	0.262
Pu 239 CAPTURES	0.0233	0.0343	0.0371	0.0403	0.0419	0.0426
DILUENT FISSIONS	0.0110	0.0480	0.0595	0.0722	0.0794	0.0825
DILUENT CAPTURES	0.0168	0.133	0.206	0.319	0.397	0.436
Na CAPTURES	0.000154	0.000965	0.00177	0.00339	0.00471	0.00543
STRUCTURAL CAPTURES	0.00560	0.00781	0.0112	0.0162	0.0195	0.0212
INTERNAL BREEDING RATIO	0.0377	0.360	0.573	0.914	1.153	1.275
EXTERNAL BREEDING RATIO	1.961	1.558	1.307	0.911	0.639	0.499
TOTAL BREEDING RATIO	1.999	1.918	1.879	1.825	1.792	1.774
Pu 239 CAPTURES / FISSIONS	0.072	0.118	0.132	0.149	0.158	0.162
LEAKAGE FROM BLANKET	0.0454	0.0207	0.0141	0.00755	0.00448	0.00322

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TABLE V

OXIDES

$\alpha =$ U ATOMS / Pu ATOMS	0.3	4	5	6	7	8
CORE VOLUME (LITERS)	27	505	870	1500	2700	5250
Pu CRITICAL MASS (kg)	56	268	385	570	893	1545
Pu 239 FISSIONS	0.328	0.298	0.293	0.289	0.286	0.282
Pu 239 CAPTURES	0.321	0.0521	0.0557	0.0587	0.0613	0.0635
DILUENT FISSIONS	0.00628	0.0428	0.0485	0.0534	0.0580	0.0619
DILUENT CAPTURES	0.00966	0.157	0.205	0.254	0.304	0.356
Na CAPTURES	0.000391	0.0072	0.0103	0.0138	0.0175	0.0216
STRUCTURAL CAPTURES	0.00346	0.0160	0.0194	0.0231	0.0267	0.0302
INTERNAL BREEDING RATIO	0.024	0.439	0.575	0.716	0.863	1.013
EXTERNAL BREEDING RATIO	1.891	1.267	1.089	0.908	0.727	0.543
TOTAL BREEDING RATIO	1.915	1.707	1.664	1.625	1.589	1.555
Pu 239 CAPTURES / FISSIONS	1.098	0.175	0.190	0.203	0.215	0.225
LEAKAGE FROM BLANKET	0.0401	0.0134	0.0102	0.0076	0.0054	0.0036

TABLE VI
CARBIDES

$\alpha = \text{U ATOMS} / \text{Pu ATOMS}$	0.4	4	6	8	9	10
CORE VOLUME (LITERS)	19	265	686	1760	3000	5460
Pu CRITICAL MASS (kg)	49	187	347	694	1060	1760
Pu 239 FISSIONS	0.325	0.093	0.084	0.076	0.072	0.069
Pu 239 CAPTURES	0.0300	0.0444	0.0496	0.0534	0.0550	0.0563
DILUENT FISSIONS	0.00869	0.0468	0.0582	0.0675	0.0717	0.0753
DILUENT CAPTURES	0.0113	0.139	0.221	0.307	0.351	0.395
Na CAPTURES	0.000279	0.00382	0.00745	0.0118	0.0142	0.0167
STRUCTURAL CAPTURES	0.00277	0.0111	0.0160	0.0208	0.0233	0.0257
INTERNAL BREEDING RATIO	0.031	0.405	0.655	0.920	1.059	1.198
EXTERNAL BREEDING RATIO	1.926	1.435	1.132	0.822	0.664	0.506
TOTAL BREEDING RATIO	1.958	1.840	1.787	1.742	1.723	1.705
Pu 239 CAPTURES / FISSIONS	0.092	0.152	0.175	0.194	0.202	0.209
LEAKAGE FROM BLANKET	0.0431	0.0174	0.0111	0.00664	0.00488	0.00338

TABLE VII
CORE DESIGN

Fuel Type	Alloy - (61.1)	Oxide (62.1)
Core power	16.6 Mw	16.6 Mw
Core volume	40 l.	40 l.
Heat transfer surface	14m ² (151 ft ²)	14m ² (151 ft ²)
Core diameter	35.8 cm (14.1 in)	38.8 cm (15.3 in)
Coolant flow area	470 cm ² (72.8 in ²)	470 cm ² (72.8 in ²)
Core composition (vol %)	{ U Pu Mo - 39.3 SS - 16.9 Na - 38 Nb - 5.8	{ UO ₂ - PuO ₂ - 42.85 SS - 23 Na - 33.25 He - 0.9
Fuel composition	{ 75 % U (27.5 % enriched in 235) 15 % Pu 10 % Mo (weight %)	{ 75 % UO ₂ (40 % enriched in 235) 25 % PuO ₂ (vol. %)

TABLE VIII
CORE THERMAL PERFORMANCE

(These values are related to a 20 Mw reactor power and a 590° C (1004° F)
maximum sodium outlet temperature)

Fuel type	Alloy	Oxide
Core inlet temperature	450° C (842° F)	
Average coolant temperature rise	90° C (162° F)	
Core outlet temperature	540° C (1004° F)	
Core sodium flow rate	4.5 x 10 ⁵ Kg/hr (10 ⁶ lb / hr)	
Core sodium velocity	{ Zone I - 1.95 m/sec (6.4 ft / sec) Zone II - 1.7 m/sec (5.5 ft / sec) Zone III - 1.4 m/sec (4.6 ft / sec)	
Maximum heat flux	200 W/cm ² (632,000 BTU/hr ft ²)	225 W/cm ² (720,000 BTU/hr ft ²)
Maximum to average heat flux	1.61	1.56
Power density	414 Kw / l	406 Kw / l
Maximum nominal coolant temperature	610° C (1130° F)	610° C (1130° F)
Maximum nominal outer SS clad temp.	620° C (1148° F)	625° C (1157° F)
Maximum nominal outer Nb clad temp.	650° C (1202° F)	-
Maximum nominal outer fuel temp.	770° C (1418° F)	920° C (1688° F)

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PLUTONIUM AS A FUEL FOR THE EXPERIMENTAL BREEDER REACTOR II (EBR-II)

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INTRODUCTION

The EBR-II is an experimental sodium-cooled fast reactor consisting of a complete reactor and power system including a conventional 20,000 Kw turbine generator and a facility for recycle of the reactor fuel. Figure 1 is a drawing of the complete facility. Construction of the plant is nearing completion at the National Reactor Testing Station in Idaho, and initial operation is contemplated late this year. Figure 2 is an aerial photograph of the facility as of May, 1962.

One of the major objectives of the EBR-II is to establish the feasibility of a plutonium-fueled fast power reactor under operating conditions comparable to those required in large scale power plants, including the fuel cycle and operation with recycled fuel. Although the EBR-II is an experimental facility, it is planned to operate the plant (for limited periods) in the manner of a central power station with an integrated fuel cycle.

Fast power reactors pose unique problems because of the high enrichment required. This necessitates operation at high power density in the core with a resultant high rate of fuel burnup. (In the EBR-II the burnup rate is approximately 1 percent every sixty days at full power operation.)

These requirements have a significant effect upon the design of the fuel, the reactor system, the fuel handling system, and the fuel cycle. Paramount among these are: (1) the requirement for finely divided fuel (small geometrical cross section and large surface to volume ratio, to permit high thermal performance); (2) the use of high performance, non-moderating coolant (such as sodium) with attendant compatibility requirements; (3) high levels of fission product decay heating with resultant cooling requirements in the reactor, during fuel handling and in the fuel cycle; (4) high specific activity in the fuel after irradiation with resulting limitations on fission product separation processes; and (5) the high value of the fuel with the resultant incentive to minimize total fuel inventory.

In consideration of these requirements, the following basic philosophy for the EBR-II fuel cycle evolved. Pyrometallurgical processing of the fuel would be employed because of the many advantages it appeared to offer for this system as compared to conventional aqueous processing. Fuel could be processed soon after removal from the reactor, thus avoiding the long cooling periods required for aqueous processing and with a consequent reduction in external inventory. Larger batch sizes could be employed because of the absence of moderator in the process. Because the fuel is

FIGURE 1
EBR-II Plant Arrangement

FIGURE 2
Aerial View of EBR-II Facility

handled in a very concentrated form, small equipment and a small overall plant is required, thus making possible considering the location of a process plant adjacent to the reactor and avoidance of fuel shipping costs, with an additional reduction in the external fuel inventory. In addition, the fission products are extracted in a highly concentrated form which should reduce fission product storage costs, and perhaps enhance the utilization of fission products. The major disadvantage of this separations process is the incomplete decontamination which is effected. The residual fission products which remain in the purified alloy require the fabrication of radioactive fuel by remote controlled procedures. This was not considered a total penalty since upon recycle of the fuel, higher isotopes of plutonium and uranium would build up, precluding direct fabrication. The heavier shielding required because of the fission products was not believed to be an unreasonable penalty.

The selected process dictated certain features of the EBR-II design. The composition of the metallic fuel alloy was established by the process with respect to the alloying constituents, i. e., the residual fission products. Many unique ancillary problems result such as the need for fission product decay heat removal during return of fuel to the reactor.

In the design and development of EBR-II, all of these requirements were considered. The design of the fuel elements and assemblies reflects the requirements of the total fuel cycle and includes some compromise to accommodate these requirements. (For example, the EBR-II fuel element does not represent the optimum design for maximum thermal performance - but is relatively simple and amenable to remote controlled fabrication.)

REACTOR ARRANGEMENT

The complete EBR-II reactor and power system has been described previously;^{1, 2} only those features pertinent to the reactor and fuel cycle are briefly reviewed here.

The reactor arrangement is shown in Figure 3 and consists of a close-packed geometry of hexagonal subassemblies. A single geometry is employed for both the core and blanket. Because of the wide variation in power density across the core and blanket, a split flow coolant system is employed. (The power density variation is approximately a factor of 100 from the center of the core to the outer periphery of the blanket.) A high pressure sodium flow system is provided for the core and inner blanket and a low pressure system for the outer blanket. The composition and geometry of the inner blanket and outer blanket are identical; "inner" and "outer" define the regions of high pressure and low pressure coolant supply respectively.

Shutdown cooling of the reactor to remove fission product decay heat is provided by natural circulation of the sodium coolant through the reactor. Natural circulation through the subassemblies is also provided during fuel handling operations. The reactor and fuel handling system are submerged below a pool of sodium which provides the sodium environment necessary to insure natural convection through the reactor and through the subassemblies during fuel handling which involves transporting them above the reactor.

FIGURE 3
Reactor Arrangement

A "submerged concept" has been employed in the basic arrangement of the EBR-II reactor and primary system as shown in Figure 4. The primary pumps, piping, and heat exchanger, as well as the complete fuel handling system are contained in the large "primary tank" and operate submerged in sodium. Coolant is circulated from the bulk sodium in the tank through the reactor and heat exchanger, returning to the tank. The major components of the primary circulation system can be seen in Figure 5.

The fuel handling system consists of a series of electromechanical devices which remove the subassembly from the reactor and transfer it to a storage rack within the primary tank for fission product decay. The subassemblies are submerged in sodium at all times during these operations. The units comprising this system as well as the fuel unloading machine for subsequent removal of subassemblies from the primary tank are shown in Figure 6. The EBR-II Fuel Cycle is based on processing the fuel after fifteen days cooling in the storage rack. After this cooling period, the subassemblies are removed from the tank and transferred in appropriate shielded containers to the adjacent Fuel Cycle Facility. Fission product decay cooling is provided by natural circulation of sodium for the first fifteen days and by forced circulation of argon gas upon removal from the primary tank. Forced convection gas cooling is required until the subassemblies are disassembled and the fuel elements separated from the cluster.

The fuel loading procedures are the same but performed in reverse sequence. Fuel transport between the storage rack in the primary tank and the Fuel Cycle Facility can be effected with the reactor in operation. Transfer between the reactor and the storage rack can only be accomplished with the reactor shut down.

EBR-II FUEL CYCLE

The EBR-II Fuel Cycle is shown in simplified form in Figure 7. The fuel is a metallic alloy containing residual fission products from the melt refining operation. The equilibrium composition of fission products is maintained by virtue of the side stream from the melt refining operation to the skull reclamation process. The reclaimed material from this process is completely decontaminated from fission products.

The initial loadings for the EBR-II contain synthetic fission products in the approximate proportions of the estimated equilibrium composition. This is done to avoid a constantly changing composition as would occur if these elements were permitted to "grow in" during each cycle. The first loading for the EBR-II consists of an enriched uranium alloy to be employed as a "stand-in" for the uranium-plutonium alloy. The initial development of the processes, fabrication techniques, etc., were developed with uranium alloys to avoid the additional complexity presented by plutonium development. Also the performance of the uranium alloy is superior and provides a preferable fuel for initial operation of the plant. The composition of these two alloys is given in Table I. The initial loading of each alloy (prepared with cold synthetic fission product elements) is indicated as Core I and Core II, respectively, while the composition for recycled fuel with each alloy is given for a

FIGURE 4
Primary System

16.8

HW-75007

FIGURE 5
Reactor Inlet and Outlet Piping System

FIGURE 6

Principal Components of Fuel Handling
and Unloading Systems

range in total content of fission products. This range will be determined by the percentage of material withdrawn in the process side stream. It should be noted that only the composition of the alloy given under "Core I" is fixed (the actual composition of the first loading as fabricated); the remaining compositions are estimates of particular systems and should be considered representative of possible alloy compositions.

TABLE I
EBR-II REFERENCE FUEL ALLOYS

Element	Uranium Fissium		Plutonium Fissium	
	Core I	Recycle	Core II	Recycle
	wt %	5.0-10.0 wt % Fissium	wt %	7.0-12.0 wt % Fissium
Molybdenum	2.46	1.9 - 3.9	2.92	1.7 - 2.9
Ruthenium	1.96	1.5 - 3.0	4.20	3.5 - 4.3
Technetium	----	0.6 - 1.1	----	0.6 - 1.1
Rhodium	0.28	0.26 - 0.53	0.65	0.43 - 0.49
Palladium	0.19	0.17 - 0.34	2.17	1.4 - 2.5
Zirconium	0.10	0.5 - 1.0	0.07	0.5 - 1.0
Niobium	0.01	----- - 0.02	-----	----- - -----
Uranium	94.90(a)	94.8 - 89.0	Balance	Balance
U ²³⁵	45.64	45.0 - 46.0	15.0 - 18.0	15.0 - 18.0
Plutonium	----	----- - -----	18.8 - 15.0	18.8 - 15.0

(a) 48.13 isotopic percent U²³⁵.

It should be noted that the plutonium-uranium alloy contains enriched uranium. This is a requirement imposed by EBR-II because of its small size. The reactor cannot be made critical with the plutonium enrichment selected, which is, however, an appropriate enrichment for large fast reactors. Also the plutonium-uranium alloy system has very poor properties above about 20 percent plutonium. It should be noted that the plutonium fission product yield is relatively higher in most of those elements which are retained in the alloy. It should be noted also that the fission product elements given in Table I are for the uranium and plutonium ratios given and would be different if all of the fissionable isotopes were plutonium.

The EBR-II core subassembly is shown in Figure 8. It consists of a closely-packed geometry of small diameter fuel elements contained in a hexagonal subassembly. Above and below the fuel section are included the

FIGURE 8
EBR-II Core Subassembly

depleted uranium sections which, when loaded in the reactor, comprise the upper and lower blanket. Special attention was given to the design of the fuel elements and subassembly to simplify the remote controlled operations to the maximum extent practicable. Every effort was made to maximize the number of preparatory operations which could be performed on "cold" material prior to fuel fabrication and final assembly.

In the EBR-II, "cold" blanket and structural parts will be used throughout. The fuel alloy will be radioactive requiring remote fabrication of the fuel element and final assembly of the subassembly. Briefly, the EBR-II fuel cycle consists of the following basic operations (these apply to both the uranium-fissium and uranium-plutonium-fissium alloys). An irradiated subassembly is removed from the primary tank after fifteen days cooling and transferred to the Fuel Cycle Facility. It is disassembled and the individual 91 fuel elements are mechanically stripped of their cladding to make available the fuel pins. The fuel pins are broken and charged into the melt refining furnace where the alloy is melted under vacuum. Fission products are removed by volatilization and by oxidation (oxygen removed from the crucible). The purified alloy is poured into a mold to provide an ingot for fabrication. The oxidized fission products, principally the rare earths, remain in the oxide slag. The fabrication sequence for fuel element fabrication begins with the purified ingot which is remelted in the casting furnace and pins are fabricated by precision casting (by pressurization) directly into vikor (glass) precision tube molds. After casting, the molds are broken away and the pins which have been cast directly to finished diameter are sheared to length. They are inspected for diameter, length, and weight, as well as visual observation for surface flaws, and eddy current for subsurface flaws. They are then assembled into the fuel element tubes. The fuel tubes are a "preassembly" including the tube, lower adapter and spiral wound spacing wire (see Figure 9). They have also been filled with a predetermined amount of sodium (extruded as a solid) to provide the sodium bond between the fuel and fuel tube. The pins are inserted, the sodium melted down, and the top cap inserted. The cap is then welded to the fuel tube. The completed fuel elements are inspected for leak tightness of the closure weld and given a "bonding" treatment and bond test to insure a continuous, void free, sodium bond between the fuel pin and tube. The procedures involved in fuel element fabrication are described in detail in a series of papers published in Nuclear Science and Engineering.^{3, 4, 5, 6, 7}

The completed fuel elements are then assembled into the subassembly. This is accomplished by "hooking" the lower adapter of the fuel element to the "T-bar" strips which are attached to the upper end of the lower blanket section. This lower section of the subassembly is preassembled "cold" including the lower adapter of the subassembly plus the lower blanket section. After the 91 fuel elements are assembled the outer hex tube assembly including the upper blanket and upper subassembly adapter are then lowered over the fuel element cluster and spot-welded to the lower adapter. During the assembly of the fuel element cluster and during all subsequent operations, forced convection gas cooling around the fuel elements is required. The subassembly is then returned to the Reactor Plant and inserted into the storage rack for subsequent loading into the reactor.

FIGURE 9
Core Subassembly Elements

Figure 10 is a drawing of the EBR-II Fuel Cycle Facility which contains two large shielded cells. The rectangular cell contains an air atmosphere and is relatively conventional in construction. It is served by a crane, two electromechanical manipulators, and six Model 8 master slave manipulators. The circular cell is a gas-tight enclosure and contains an argon atmosphere. It is served by two cranes and six electromechanical manipulators. All operations involving exposed fuel alloy are performed in the argon cell. All operations involving clad fuel (protected from the atmosphere) are performed in the air cell. The most critical operations, including decladding of the fuel elements, fission product separations, fuel pin fabrication, and fuel element assembly are performed in the argon cell. Many unique problems were encountered in the development of the argon cell and the process equipment resulting from the high radiation levels and the pure, dry, inert gas environment. These have been described in detail previously.³

The argon cell is constructed on a modular basis with identical provisions for services to each module of the working area. This will permit experimental flexibility in modification or replacement of process equipment. The circular design simplified the power and control wiring for the cranes and manipulators and also eliminates the need for trailing cables, etc., which present a difficult maintenance problem. In addition, a defective unit can be "pushed" by an operative unit and since the cell is circular does not result in an inaccessible work area. All equipment and services in the argon cell are remotely removable and replaceable, including lights, manipulator and crane components, process equipment, etc. The 5-foot thick windows are designed to permit replacement of the inner two glass slabs and in addition are provided with heavy steel "shutters" to protect the glass from irradiation when not in use. Figure 11 is a photograph of the exterior working face of the two cells (the air cell to the left - argon cell in the center). Figure 12 is a photograph of the interior of the argon cell. The floor penetrations for services to several work area modules can be seen. Figure 13 is a photograph of the interior of the air cell. The inner slab of one of the windows is shown swung on its hinges for removal. Figure 14 is a photograph of the melt refining furnace with the bell housing removed. The service connections to the floor penetrations can be seen. In the foreground is the purified ingot removal machine with the rectangular mold pivoted at the top.

The development of the fuel cycle for EBR-II has been accomplished primarily with the uranium-fissium alloy. The first loading for the reactor was fabricated utilizing prototype equipment and remote control procedures to the maximum extent possible. Initial work performed on the uranium-plutonium alloy has indicated that the same basic processes and the same basic equipment will be adequate. The performance of the uranium-plutonium alloy is inferior, however, and this may impose new fuel cycle requirements which in turn may require modification of the fuel cycle.

Irradiation data indicate that the uranium-plutonium-fissium alloy is far less irradiation damage resistant than is the uranium-fissium alloy.⁸ The swelling temperature is lower and the irradiation damage occurs at a much lower operating temperature. Efforts to improve the alloy by changing

16.16

FIGURE 10
EBR-II Fuel Cycle Facility

HW-75007

FIGURE 11
Fuel Cycle Facility Operating Annulus

FIGURE 12

Fuel Cycle Facility Argon Cell

FIGURE 13
Fuel Cycle Facility Air Cell

FIGURE 14
Melt Refining Furnace

the alloy composition have been essentially unsuccessful (except by reduction of plutonium). The most promising solution appears to be in the direction of providing "cladding restraint". Niobium, vanadium, and molybdenum base alloys appear to have the desirable properties of high temperature strength and are chemically inert with the fuel alloy. Their major disadvantage is difficulty of fabrication. Work is continuing on the development of suitable cladding alloys for this application including fabrication and welding development. This approach results in the fuel expanding tightly inside the cladding. The uranium alloy fuel elements will be decled by mechanically slitting the fuel tube and "peeling" it from the fuel pin. This is shown in Figure 15. This method may not be successful when cladding restraint is employed and alternate methods are under investigation, including chemical removal as well as chemical attack (to weaken or embrittle the material for removal).

FIGURE 15
Spiral Decanning Operation

The potential advantages and disadvantages of plutonium-uranium metallic alloys can be identified. The advantages are: high density which directly and indirectly enhances breeding; predictable thermal expansion which can be made to contribute to reactor safety and stability (by providing negative power coefficient); reasonable thermal conductivity (but this must be considered in conjunction with maximum permissible operating temperature); and simple fabrication which is conducive to remote operation. The disadvantages are: poor irradiation damage resistance; and relatively low melting point which presents a limitation to high temperature operation (if melting or near melting is to be avoided). Operational experience and additional developmental data are required before these factors can be properly assessed.

The basic premise underlying the EBR-II concept and program is that plutonium fuel systems will be developed and demonstrated on the basis of a total system (including fuel cycle) to the maximum extent possible. Experimental irradiations will be conducted on promising fuels, of course, without having in hand an applicable fuel cycle, but complete full loadings will emphasize the total operation. It is believed that the uranium-plutonium-fissium metallic alloy represents such a system even though certain operational limitations have been identified. Other systems are being investigated on a longer range basis for possible application in the EBR-II. One such system is the uranium-plutonium-carbide system. Preliminary investigations have indicated that fission product separation can be effected in liquid metal solvent systems and that the purified fuel can be precipitated from these solutions as the carbide. Irradiation data indicate that the carbide is quite stable and that it is capable of high temperature operation.

In summary, a primary objective of the EBR-II is to demonstrate the feasibility of operation of an experimental fast power reactor with an integrated fuel cycle operating on short cooled fuel and requiring a small inventory. It accepts incomplete decontamination with the resultant requirement of remote fuel fabrication. The immediate emphasis is on metallic fuel alloys, but other fuel systems can be investigated. The uranium-plutonium-fissium fuel alloy is inadequate to meet the EBR-II operational objectives but probably can be made to satisfy them if cladding restraint is employed. The initial loading of uranium-fissium alloy appears to be adequate for the reactor, and will provide a suitable substitute fuel for initial operation of the complete plant. The transition to uranium-plutonium fuel appears relatively straight forward with the exception of new requirements which may be imposed by the employment of cladding restraint.

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PLUTONIUM AS A FUEL FOR THE FAST CERAMIC REACTOR

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Design, testing and evaluation of mixed plutonium-uranium oxide fuel for use in large, sodium-cooled fast reactors are being carried out as a major part of the Fast Ceramic Reactor program.¹ The high performance potential of this fast reactor, coupled with the good neutron economy of the fast neutron spectrum and the high thermal efficiency of sodium-cooled systems, leads to an estimated fuel cycle cost of approximately 1 mill/ekwh.² The ceramic fuel also provides a key feature of importance to safe and stable reactor operation, viz. a large negative Doppler coefficient.³

DESIGN

Design effort on the FCR concept has focused on power reactors in the 500 to 1,500 MWe range. Detailed physics and core analysis has been performed on a 520 MWe reference design.^{3,4} Prime emphasis in this design has been to meet rigid safety and mechanical design standards while optimizing fuel cycle costs.

Some of the important parameters of the reference FCR design are summarized in Table I.

TABLE I

REFERENCE FCR DESIGN

Reactor Power, Net Electric	520 MW
Gross Thermal	1395 MW
Core Composition	
PuO ₂ -UO ₂ /Na/SS	33/50/17 v/o
Fuel, PuO ₂ Conc., Fiss./Total	12.8/19.1 w/o
Exposure, Avg.	100,000 MWD/T
Linear Rod Power, Max.	18.5 Kw/ft
Specific Power, Avg.	1,000 Kw/kg Fiss. Pu
Δk , Doppler Effect	-\$1.8
Excess Operating	+\$1.8
100% Na Loss	\$0
Coolant, Temp., In/Out	785/1050°F
Velocity, Avg./Max	20/34 ft/sec
Pressure Loss	70 psi

The power output of the reference design is achieved with a cylindrical core 3.25 feet in height by 6.5 feet in diameter. The core height/diameter ratio of 0.5 reduces the reactivity effect due to accidental fuel movement such as slumping or assembly bowing and also improves core hydraulics. The chosen sodium content (50 v/o) is calculated to give a zero reactivity effect in the event of accidental sodium loss.

The core fuel consists of cylindrical rods of mixed plutonium-uranium dioxide of tabulated Pu Concentration, clad in stainless steel. The rods are grouped in triangular array in hexagonal assemblies, with 127 fuel rods per assembly, thus limiting the maximum reactivity worth of a new element to less than \$1.

Individual fuel rod design is based on an average discharge fuel exposure of 100,000 MWD/T, a maximum rod power of 18.5 kw/foot and an average specific power of 1,000 kw/kg of fissile Pu.

Resulting dimensions for the rod are given in Table II. The rod diameter is derived directly from assumed rod linear power and specific power. Considering both pressure and thermal stress, the optimum ratio for cladding thickness to fuel diameter corresponds to a cladding thickness of 0.015 inch for the 0.25 inch OD rod.⁵ At this optimum a maximum fission gas pressure can be contained, amounting to 1,800 psi.

To achieve long fuel life with the observed high fission gas release, fuel rod design must provide for gas pressure relief. In the reference design, this is accomplished by venting internally to an end gas plenum. With the conservative assumption of 100% fission gas release, a reservoir approaching the core fuel length is calculated to be required in accordance with the gas law.

TABLE II
REFERENCE FUEL ROD DESIGN

<u>Rod Diameter</u>	0.22 inch meat diameter 0.25 inch clad OD
Linear rod power, max.	18.5 kw/ft
Specific power, ave.	1,000 kw/kg fiss. Pu
Power distribution, max./ave.	2.0
<u>Clad Thickness</u>	0.015 inch Type 347 S.S.
$(\sigma_{Pr} + \sigma_{Th}) \leq \text{Elastic limit}$	
$(\frac{t_c}{D_f}) \text{ opt.} = 1/15$	
<u>Fission Gas Reservoir</u>	2.9 feet long (for 3.25 feet core)
Fuel exposure	100,000 MWD/T
Fission gas release	100 %
Gas T/P	1050 F/1800 psi

Thus, as shown in Figure 1, the reference core fuel design includes within a single 0.25-inch O.D. rod, a 39-inch mixed oxide length, bracketed by two 15-inch lengths of uranium oxide as top and bottom axial blanket, plus a 35-inch length of fission gas reservoir at the top end.

In an alternate design, shown on the right in Figure 1, gas venting to the coolant is under investigation. In what is considered to be the most promising vent-to-coolant design, an inverted trap is used to permit escape of gas while preventing sodium back-flow. The trap, which is intended to serve as a check valve without having moving parts, may be located either at the top or the bottom end of the fuel rod. Its use would allow reduction in the total length of the rod and in the cladding thickness, at the expense of releasing some fission products to the coolant system.

TESTING

In a preliminary test series completed nearly a year ago, capsule irradiations of 40 mixed oxide fuel specimens were conducted at burnups up to 100,000 MWD/T at maximum rod powers up to 22 kw/ft ($\int kd\theta = 60$ watts/cm), and at stainless steel clad temperatures approximating 1100°F.⁶

The oxide powder preparation and fuel fabrication flow sheet for these specimens is shown on Figure 2. The mixed oxide was prepared by ammoniacal co-precipitation, drying and reduction. The spike of 40% U²³⁵ was added to maintain the U/Pu ratio nearly constant even after high burnup in a thermal flux. While the 75% theoretical density for the swaged specimens was accepted for this preliminary study, densities of 90% or slightly higher are expected to be attainable by swaging or vibratory compaction techniques being developed for UO₂.

A sketch of typical pelleted and swaged test specimens is given in Figure 3. The active fuel zone was insulated at either end by natural UO₂ pellets, and a fission gas plenum was incorporated to accommodate released fission gas.

The fuel specimens were irradiated, four to a capsule, in the thermal flux of the General Electric Test Reactor. Burnups ranged from 5000 to 100,000 MWD/T and maximum rod powers from 7 to 22 kw/ft. Despite these high performance conditions, no cladding failure, either rupture or even significant change in cladding dimensions, was observed with any of the 40 specimens. However, as might be expected, radial redistribution of the fuel occurred leading to central void formation at all but the lowest rod powers. As shown in Figure 4, internal fission gas release ranged from 10 to 70%, there being no obvious correlation with either heat flux, i. e., rod power, or burnup as given in thousands of MWD/T by the numbers adjoining the release data points. Release values were slightly higher and more scattered for the low density swaged fuel than for the pelleted fuel.

NOTE: NOT TO SCALE

FIGURE 1
Fuel Pin Designs

FIGURE 2
FCR Fuel Preparation and Fabrication

FIGURE 4

Fission Gas Release Data, Percent Release Versus Heat Flux

Representative transverse cross-sections of the fuel are shown in Figure 5. Major points worthy of note include closure of the fuel-clad gap in the pelleted fuel case from an as-fabricated diametral value of 0.006 to 0.008 inch. The fuel is apparently refabricated in-pile to give good fuel-clad contact in the case of pellets and high density in the case of swaged fuel.

Another significant point: No major discontinuity in fuel appearance or performance occurred as a function of increasing burnup or power density in these irradiations. Accordingly, extension of these thermal flux irradiations to higher exposures and rod powers is planned to establish exposure limits, as well as fast flux irradiations to achieve more representative conditions as soon as appropriate facilities are available.

Additional post-irradiation examination of the mixed oxide fuel specimens has included determination of possible (a) separation of plutonium and uranium as a result of the large radial thermal gradients during irradiation and (b) axial migration of fission products from the active fuel zone to the cooler UO_2 insulator pellet which separated the fuel from the end gas plenum.

The plutonium migration is of interest because it could result in separation of the positive and negative contributions to the fuel's Doppler coefficient due to Pu^{239} and U^{238} ; the F.P. migration, because of the threat of a plug of distilled fission products interfering with escape of released fission gas to the pressure-relief plenum or vent.

In preliminary investigation of plutonium migration, radial variation of uranium and plutonium isotopic composition was determined on the highest burnup swaged and pelleted specimens.⁷ Observed variations in composition were in general agreement with values calculated from cross section and thermal flux depression data for this highly enriched fuel, i. e., within the 20% limits of uncertainty estimated associated with the isotopic analyses and physics calculations. Thus, there was no evidence of gross thermally-induced migration of plutonium relative to uranium. However, in view of the importance of this phenomenon to the very important Doppler effect, further investigation to define limits of plutonium migration is planned.

To investigate possible fission product migration, radiochemical analyses for long-lived fission products were performed on samples taken along the length of a pelleted mixed oxide specimen, with results plotted on Figure 6.⁸ Here fission product concentration is plotted semi-logarithmically against longitudinal location, proceeding from the natural UO_2 pellet region to the adjoining active mixed oxide fuel zone. The dotted line, actually that for Ce^{144} , essentially defines the variation in fission intensity: from a high, nearly constant value in the mixed oxide core fuel, through a slight peak near its end due to power peaking dropping quite sharply to a value attributable to U^{235} fissioning in the natural UO_2 material. Thus, for the relatively refractory Ce^{144} and the Zr^{95} which parallels it, there is negligible migration indicated from hot core to cool blanket. With

Specimen IV-4-S;
9,000 MWD/T

Specimen V-1-S;
99,000 MWD/T

Specimen IV-3-P;
10,900 MWD/T

Specimen V-2-P;
77,400 MWD/T

FIGURE 5

Transverse Cross-sections of Irradiated $\text{PuO}_2\text{-UO}_2$ Fuel

FIGURE 6

Axial Fission Product Distribution in Specimen 1X-1-P

Cs^{137} , however, appreciable axial migration occurs in line with its higher vapor pressure. This mobility of cesium has, of course, been noted before by numerous observers and has come to discourage the use of Cs^{137} as a localized burnup indicator. The even greater apparent migration of Cs^{134} may well be due to mobility of its relatively long-lived gaseous precursor, 5.3d Xe^{133} , as compared to the 3.8m Xe^{137} . Evidence for slight Sr^{90} migration was also noted.

While no blockage of gas flow to the end plenum has been observed in the short specimens tested to date, the possibility of this occurring in long prototypical fuel pins appears sufficient to warrant further investigation.

"Failure" testing of oxide fuels has also been initiated with preliminary tests using UO_2 as a stand-in for mixed fuel in investigations of (a) defected ceramic fuel performance in sodium coolant, i. e., sodium-logging, and (b) response of fuel and cladding transient overpower irradiation in TREAT.

A series of short-term irradiations of cladding-defected UO_2 fuel specimens is being conducted to investigate the effect of accidental admission of sodium into such fuel operating at high power density. With very pessimistic assumptions as to the consequences of the entry of sodium into a central void during reactor shutdown followed by its flash evaporation at startup, one can calculate a sodium vapor pressure sufficient to distort or even burst the fuel cladding, which could in turn perhaps lead to propagating failure within an assembly.

The first four specimens (an undefected control and three specimens containing 0.005-inch diameter holes in their cladding) have been subjected to 5-8 rapid thermal cycles from low to high (20-23 kw/ft) rod power.⁹ No swelling, bowing or other dimensional changes occurred with any of the specimens. The control specimen (no defect and, therefore, no sodium entry) had developed a small central void within the 2 hours total it had been at high power. Sodium had penetrated the hole in each of the three defected specimens. However, although they had operated at approximately the same rod power as the control, no central void had developed in any of them, apparently due to the increased heat transfer provided by the entering sodium.

Additional more severe defect tests have been proposed. These include exposure of a fuel specimen prefabricated with a central void containing sodium and having no defect hole in the cladding for sodium escape. Results of the first of these tests are indicated in Figure 7. Again, no distortion of the cladding had occurred. Postirradiation examination, in particular a determination of the relocation of the sodium initially inserted in the central void, is now in progress in an attempt to explain this somewhat surprising but very encouraging result.

FIGURE 7

Specimen B-1-F

Defected fuel testing will also be extended to a determination of the effects of prior burnup and more severe overpower excursions on actual $\text{PuO}_2\text{-UO}_2$ mixed oxide fuel.

In another test series, transient overpower testing of fuel has been performed, initially on simulated EFCR fuel specimens (1 inch d UO_2 fuel encapsulated in a NaK medium). Irradiations have been carried out in the Transient Reactor Test facility in Idaho with the cooperation of Argonne National Laboratory personnel.

Single transient irradiations of four specimens have established the approximate gross damage threshold at a calculated maximum fuel temperature of about 7000°F .⁹ This is, of course, well above the melting point of UO_2 . In fact, flash boiling of the fuel had apparently occurred, as shown in Figure 8, a view of the "volcanic" action at the top end of the fuel. While the extreme overpower condition (7 kw/cc over a 3 second period) produced incipient fuel failure, this is well beyond that required for useful transient operation of EFCR. Further tests are planned to investigate both single and repeated transient limitations for smaller diameter rods of actual mixed oxide fuel, the effects of prior irradiation and the efficacy of increasing clad thickness and using higher strength materials to increase resistance to transient overpower.

EVALUATION

Advantages associated with ceramic fueling of fast reactors have been evaluated in the two important areas of reactor dynamics and fuel cycle costs.

In a large Fast Ceramic Reactor a sizeable and rapid-acting excursion limiting capability is provided by the large negative Doppler effect and short neutron lifetime. The reference FCR, for example, has a calculated prompt negative Doppler effect of about \$1.8 if the fuel temperature were to increase from its normal operating value to the melting point at the position of maximum rod power.

In small fast reactors the prompt negative coefficient is associated with uniform thermal expansion of the core. In large reactors this expansion coefficient becomes small and furthermore unreliable, as the possible modes of mechanical distortion increase with size. In the case of the ceramic-fueled reactor, the latter point is further complicated by the fact that the fuel cracks under operating conditions.

Since the Doppler effect represents the only reliable prompt negative power coefficient in a Fast Ceramic Reactor, its importance for safety and operating stability can hardly be overemphasized. It has, therefore, been proposed that a small, i. e., 10-50 MWt, test reactor be built to demonstrate the Doppler effect in operation.¹⁰ This reactor, the Experimental Fast Ceramic Reactor, would fulfill a role in the fast reactor program analogous to that of the Borax and Spert reactors in the water reactor program. Its successful operation would confirm the calculated inherent excursion limiting characteristics and thus provide a major step towards development of a commercially acceptable Fast Ceramic Reactor.

FIGURE 8

Upper Section APED TREAT Sample #10

A fast oxide fuel cycle cost projected for 1975 is given in Table III. The total cost, including first core capitalization, is estimated to be 0.84 mill/ekwh.⁴ Assumed unit costs and key economic ground rules giving this total are tabulated under the Reference column. Values of the various parameters which, if changed individually, would lead to a 0.1 mill increase in total cost, i. e., to 0.94, are tabulated under the Reference +0.1 column. The fabrication and reprocessing reference unit costs are those estimated for a 3,000 MWe processing capacity assuming a sintered pellet and aqueous reprocessing flow sheet, respectively. The detailed basis for these processing methods and costs will be subject of a paper to be presented at the 1962 Winter Meeting of the ANS (November, 1962).¹² The fissile plutonium price in 1975 is based on a thermal recycle market value. In the fast reactor, fertile plutonium is taken to have approximately 15% of the fissile plutonium value. The plutonium inventory charge of 10% per year reflects assumed private ownership of fuel material by 1975.

TABLE IIIESTIMATED 1975 FUEL CYCLE COST

Fabrication	0.32	
Reprocessing	0.11	
Pu Credit	-0.07	
Pu Inventory	0.36	
First Core Capitalization	<u>0.12</u>	
TOTAL	0.84 mills/ekwh	
	<u>Ref.</u>	<u>Ref. + 0.1</u>
Fabrication, \$/kgU + Pu		
Core & Axial Blanket	135	182
Radial Blanket	52	87
Re processing, \$/kgU + Pu		
Core & Axial Blanket	33	99
Radial Blanket	30	80
Pu Price, \$/g		
{ Fissile Isotopes	6.50	8.70\
{ Nonfissile Isotopes	1.10	1.50\}
Pu Inventory %/yr.	10	13
First Core Capit., %/yr.	14	26
Load Factor, %	80	66

A comparative study of the application of mixed plutonium-uranium carbide and mixed plutonium-uranium oxide fuels in a large FCR indicates a potential reduction in fuel cycle cost of about 0.3 mill/ekwh with carbide due to its higher thermal conductivity and density.¹¹ However, this analysis assumed as yet undemonstrated equal fuel exposure and fabrication costs and successful resolution of formidable cladding and heat transfer problems for the carbide fuel. The FCR program will for the present continue to place major emphasis on mixed oxide fuel development.

TO SUMMARIZE:

Experimental and analytical investigations completed to date indicate that large, plutonium-uranium oxide fueled fast reactors can be designed to yield stable and safe reactor dynamics characteristics and attractively low fuel cycle costs. To confirm this potential, an accelerating program of fuel and reactor development is planned.

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FUEL ELEMENTS FOR FAST REACTORS

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INTRODUCTION

Fast reactors offer the attractions of competitive nuclear power and the breeding of new fuel from fertile material included in the core and blanket. The need to use highly-enriched fuel, however, demands the use of compact highly-rated cores cooled by liquid metals; and hence more arduous service conditions are imposed on the fuel elements than in any other type of reactor. In addition high burn-up levels are possible owing to the small reactivity changes which occur with burn-up; and burn-up levels of 5-10% of the heavy atoms are likely to be necessary to produce sufficiently cheap power. It is the basic function of the Fuel Element Group at Dounreay to develop fast reactor fuel elements capable of withstanding the proposed service conditions and high burn-up levels.

The fast reactor programme in the United Kingdom is geared to civil fast reactor stations being built by the mid 1970's. Consequently it is possible to consider for development only those fuel materials the feasibility of which has already been established, and thus attention at the moment is confined to plutonium-bearing cermet, oxides, carbides and metals.

NUCLEAR CONSIDERATIONS

A reactor with an output of around 500 MW(E) is considered as the size most likely to be chosen for the first fast reactor power station in the United Kingdom and nuclear physics calculations have been carried out for this size of reactor. The breeding ratios obtained by these calculations have an important bearing on the choice of fuel, both from the economic aspects and also because a high breeding ratio will permit acceleration of the rate of installation of fast reactors in later years.

The density of heavy atoms in the fuel will govern the U/Pu ratio which can be used and in consequence will affect the breeding ratio, while another important factor is the amount and nature of non-fertile, non-fissile atoms present. Typical initial breeding ratios for the four fuels considered are:

Oxide	1.6
Cermet	1.1
Carbide	1.7
Voided Metal	1.5

Although these will be affected to some extent by the detailed design of the reactor they do, however, form a basis of comparison for the various fuels. The voided metal fuel suffers slightly with respect to oxide largely because of the thicker cans which are thought to be necessary, while cermet fuel gives a low breeding ratio owing to the low density of heavy atoms and the presence of a large amount of steel.

OXIDE FUELS

General

Oxide nuclear fuels have the advantage of good stability under irradiation to burn-up levels up to 2% and, in their densest form, some retention of fission product gases up to temperatures in the region of 1600 C; but as the temperature is increased above this limit a larger proportion of the fission product gases is released. Owing to the poor thermal conductivity of oxide fuels, a highly-rated fuel element designed to work with a central temperature limit of, say, 1600 C is of very small diameter and the alternative of a design to operate at higher temperature and to accommodate the released gas has a great attraction. This accommodation for gas may take the form of a central or end void; or low density fuel. In this connection, it is advantageous to use high-strength canning materials to provide the maximum degree of restraint possible.

In the case of uranium dioxide fuel it has been established that the stoichiometric material retains fission product gases more successfully than nonstoichiometric material. It is feasible, therefore, that a similar solid solution of UO_2 and PuO_2 will behave in an identical manner, and hence oxide fuel with an oxygen/metal ratio of 2.00 is currently being considered for fast reactors in the United Kingdom. It has yet to be established any such limitation is necessary. Similar considerations also apply to the ceramic phase of cermet fuels to be discussed later in the paper, although this is likely to be of minor importance in the cermet case owing to the low temperatures at which the oxide component is expected to operate.

In the event of a can becoming perforated sodium would enter during a thermal cycle and it has been suggested that the sodium could become vapourised with sufficient rapidity to cause sudden rupture of the can. Similar fears were also expressed in the early days of water reactors and have now been proved groundless. A theoretical treatment of the fast reactor case has shown that sodium entry into oxide fuel elements is unlikely to cause damage.

Fabrication

$(\text{UPu})\text{O}_2$ pellets are fabricated from material produced by the controlled coprecipitation of an intimate mixture of plutonium hydroxide and ammonium di-uranate from mixed nitrate solutions. The precipitate is then calcined and reduced to yield a single-phase solid solution of the oxide, and the resultant agglomerates are broken down to fine powder by ball milling. This material is then mixed with a binder, and granulated to give a free-flowing feed to the

pelleting press. Pellets are pressed, debonded in a slightly oxidising atmosphere and sintered in dried hydrogen. The oxygen/metal ratio is adjusted by heating in an atmosphere of gas-bottle hydrogen; giving as a final product stoichiometric (UPu)O₂ pellets of 95-97% theoretical density.

Irradiation Testing

A small number of irradiation tests have been carried out with uranium oxide specimens consisting of 55% enriched UO₂ pellets of 0.1-inch diameter canned in "Nimonic" 80 of 0.0115-inch wall thickness, with 37% free space within the can, and were irradiated in DMTR to a maximum burn-up of 5.4% of heavy atoms at a rating of 560 w/g with predicted temperatures of 340 C at the surface and approximately 1300 C at the centre.

Postirradiation examination revealed extensive cracking and porosity of the fuel; and the absence of a central cavity. Three zones were apparent; namely, a surface zone containing porosity (barely resolved at a magnification of x 500) interspersed with small patches of almost featureless material (Figure 1); a transition zone, containing a relatively small number of irregular shaped pores and areas of extremely fine porosity (Figure 2); and a central region with irregular shaped voids apparently outlining grain boundaries (Figure 3). It was noticeable that sudden changes in the type of porosity tended to occur at circumferential cracks, implying a temperature effect. At the end of the specimen, extrusion of the UO₂ into the free space had occurred, but the appearance of the original pellet junctions implies that this degree of plasticity was purely a local effect. Twelve percent of the calculated amount of fission produce gases was released from the fuel.

In further tests, UO₂ specimens 0.125-inch diameter irradiated to 4% burn-up with centre temperatures of approximately 2000 C invariably showed a central void, the size of which appeared to increase as burn-up proceeded. Gas releases occurred of up to 50% of the gases produced.

An oxide fuel element with the maximum centre temperature limited to below 1600 C is of small diameter. The narrow coolant passages which result would be liable to blockage. If the centre temperature is increased to avoid these narrow coolant passages it is necessary to incorporate free space within the fuel pin to accommodate the release of fission product gases. This may be located either in the end of the fuel pin, in which case the problem arises of ensuring that the gas can reach it without generating unacceptable pressures at other parts; or it may be distributed throughout the length of the pin which may give rise to undesirable movements of fuel. A further point is regarded as important with oxide fuels is the effect of fuel to can clearance on fuel performance. It has been postulated that moderately large gaps will not adversely affect performance (remembering that the elements may be designed to accommodate the release of all fission product gases formed) and that any very high temperatures produced will be self-correcting. Experiments are in hand to examine the phenomena associated with such large fuel/can clearances.

FIGURE 1

Appearance of Porosity
Near Surface of Irrad-
iated UO_2 Pin x 500

FIGURE 2

Appearance of Porosity
in Transition Zone
or Irradiated UO_2 Pin
x 8000

FIGURE 3

Appearance of Porosity
Near Centre of Irrad-
iated UO_2 Pin x 1000

CERMET FUELS

General

By incorporating a metal component in an oxide fuel or by dispersing the oxide in a metal matrix it is possible to increase the thermal conductivity and consequently to increase the permissible diameter of the fuel rod. This can be achieved by using a small concentration of a metal of a high thermal conductivity, or by employing a larger concentration of a metal of lower thermal conductivity but of higher strength. If the latter approach is selected, then the ceramic phase can be present as discrete granules embedded in a continuous matrix of metal to give a dispersion fuel, and this approach to cermet type fuels is being investigated in the United Kingdom, using a 316L stainless steel as the matrix material because of its high creep strength and ductility.

Because of the economic incentive to maintain as high a concentration of fissile and fertile material in the fuel as possible the main line of development has been concentrated on a cermet containing 50% by volume of the ceramic phase. This has raised a number of problems not previously encountered with cermets of lower ceramic concentrations. Cermets containing a high concentration of ceramic phase must be expected to have a relatively low macroscopic ductility, and it is considered that if any cracks appear under thermal stresses, either directly or as a result of repeated cycling, these will result in poorer performance, in part because the fission product gases will no longer be contained by the individual "pressure vessel" around each ceramic granule, and in part because the cracks may act as stress raisers to the bonded supporting can. Swelling or blistering will also be a problem at some burn-up level, and failure from this cause is most likely to be initiated at imperfections. Consequently considerable development effort has been expended in an attempt to achieve a uniform granule distribution. Moreover, in order to maintain a web of matrix round each granule undamaged by fission fragments it is necessary to keep the average granule size large; although an upper limit will be set by the need to maintain a uniform distribution in a narrow cross section and by the increased tendency to fracture of the larger granules during subsequent cermet fabrication.

Fabrication Development

While a considerable amount of experience has been accumulated, particularly in the USA regarding the fabrication of cermets with a low volume concentration of ceramic phase, the fabrication of the cermet fuel with 50 vol % ceramic phase presented a number of new and challenging problems. In addition the (UPu)O₂ ceramic phase was likely to show some differences in strength and physical properties compared with uranium dioxide so that even data which at first sight appeared to be directly relevant needed to be confirmed; and indeed important differences have been found. This work is still proceeding, and will be reported later. The fabrication practice is aimed throughout at developing production methods which are potentially economic on a large scale, and are thus capable of being employed in the manufacture of fuel elements for a civil fast reactor programme.

The first problem was the manufacture of uniformly-sized spherical granules, and the development of methods suitable for routine production. A number of methods are now available for this, differing mainly in the mechanical details. All involve essentially the addition of a small quantity of binder to assist the controlled agglomeration of fine particle sized coprecipitated $(UPu)O_2$ feed material followed by a mechanical spinning treatment to spheroidise and densify the green agglomerates. The amount and nature of the binder used is important affecting both the quality of the product and the yield in various size fractions and varies from batch to batch. The green granules are carefully debonded and sintered to give spherical particles of stoichiometric $(UPu)O_2$ of about 95% theoretical density. The granules are then sieved to remove coarse agglomerates and fines, and to select the required range. Consideration is now being given to methods of eliminating nonspherical granules. A photograph of typical granules is given in Figure 4.

FIGURE 4

Typical $(UPu)O_2$ Granules x 30

The granules are then coated with stainless steel powder, pressed into pellets, sintered in hydrogen to reduce superficial oxide films present on particles of the steel which retard matrix self-bonding and partial densification, and are densified further by mechanical working. Two methods of fabrication have been shown to be satisfactory for densification with minimum damage to or redistribution of the ceramic phase; namely, hot swaging, and repeated cold-coining with intermediate annealing. In swaging, the critical factors are work-piece temperature, swaging rates, reduction in each pass, and total reduction. It has been found that using too large a total reduction (overswaging) leads to lower densities associated with the presence of cracks within the fuel. The coining operation consists of pressing the

sintered pellets in a range of slightly large-sized dies, to accommodate elastic recovery and stress-relaxation on annealing; and the critical factors here are the sizes and clearances of the dies, the coining pressures used and the number of such operations governed by the work-hardening capacity of the matrix material. In this work the object has been to achieve the maximum density possible in the matrix to avoid permeability of the fuel to gaseous fission products. As a result of this fabrication development programme, it is now possible to make 50 vol % cermet specimens having room temperature tensile strengths of 14,600 psi with elongations to fracture in excess of 5%.

Irradiation Testing

An irradiation programme was undertaken to determine, in the first instance, the liability of the 50% cermet fuel to cracking and to swelling. Consequently, the first tests were of short duration to determine whether cracking occurred due to a few applications of the thermal stress only; and these were followed by tests of longer duration, mainly to determine whether swelling or blistering occurred before an economical burn-up was reached.

As the irradiation programme and the development of fabrication processes proceeded concurrently, the first specimens departed markedly from the ideal and contained defects such as cracks, poor distribution of granules, nonspherical granules, or low density matrices. In the first phase of the work, to low burnup only, no cracking which could be attributed to thermal stresses were detected after irradiation; although in the majority of cases some release of fission product gases from the specimens occurred. At higher burn-ups, one specimen irradiated to 5% burn-up of heavy atoms did not suffer any change of external dimensions, while in other specimens irradiated to considerably higher burn-up cracking of the cladding occurred accompanied by varied swelling of the cermet core. In all cases, large numbers of gas bubbles were formed in the oxide phase, which has swelled to fill the available space. In the unfailed specimen the bonding between the granules and matrix, and the matrix and can, has become closer (Figure 5). It is possible that the failures can be attributed primarily to the poor quality of the specimen, and in particular the low degree of integrity of the matrix. The specimen which did not fail appeared to be of marginally better quality but in particular the thickness of cladding was considerably greater and this latter fact is probably the reason for the different behaviour, rather than the quality or the burn-up level.

FIGURE 5

Structure of Centre of Irradiated Cermet Sample x 200

Out-of-Pile Testing

Tests have been carried out to simulate the thermal stresses which occur during service by passing a heavy electric current through the specimen while it is water cooled through an alumina thermal barrier to keep the surface temperature at about 600 C. In one such test, a power of 2KW was dissipated in a 2-inch specimen and the specimen cycled until failure occurred. A thermocouple recorded a centre temperature of 810 C at the start of the test, but this rose to over 900 C before failure occurred after 75 cycles. Metallographic examination of the specimen showed only localised damage and suggested that oxidation may have played some part in the failure mechanism; while large volumes of the cermet including the point at which the temperature was measured, showed no sign of damage and had the same electrical resistivity as before testing. Thus, the test indicates that good quality 50 vol % cermets are not susceptible to cracking. A series of tests of this nature is being carried out to establish fully the behaviour of the fuel under thermal stress fatigue conditions.

CARBIDE FUELS

Uranium monocarbide has been shown to have excellent irradiation stability and gas retention to moderate burn-ups at low temperatures, and diffusion experiments at higher temperatures lead to the belief that this will be maintained at high temperatures. This factor coupled with the high thermal conductivity of carbides, and the higher density of heavy atoms

(compared with oxides) makes a solid solution of plutonium and uranium monocarbides an attractive fuel for fast reactors. The irradiation stability of plutonium bearing carbide fuels to high burn-ups under fast reactor conditions still awaits convincing demonstration.

The thermal bond for carbide fuels requires careful consideration and solid, liquid and gaseous bonds have been suggested. A solid bond must be able to withstand thermal stresses produced by temperature gradients and differing coefficients of thermal expansion and while there is some hope of solving these problems, success for the conditions pertaining to fast reactor fuels cannot be guaranteed at present. A gaseous bond, on the other hand, presents a few real problems; but it suffers the inherent disadvantages of imposing a large thermal barrier between fuel and can, where otherwise thermal conductivities would be high. Moreover, the conductivity of a gas bond will decrease as irradiation proceeds due to dilution by escaping fission product gases. An alternative is to use a sodium bond, and, while this may mean limiting the centre temperature of the fuel, the thermal conductivity of carbide is sufficiently high to give attractive sizes for the design of fuel pins.

The thermal conductivity of a solid solution of uranium and plutonium carbides has been measured by a thermal comparator method. The specimens were buttons, produced by arc-melting a series of binary U-Pu alloys with carbon chips; although stoichiometric composition was aimed at, the specimens were mainly carbon deficient, although single-phased. Results are depicted graphically in Figure 6. It will be observed that the attractions of high-thermal conductivity, attributed to carbide fuels, are less significant in the case of PuC-UC alloys of compositions of interest for fast reactor fuels, which have about half the thermal conductivity of plain UC.

FIGURE 6
Thermal Conductivity
of (UPu)C at 20 C

A further problem with carbide fuels is the compatibility with the various canning materials as this is largely unknown for plutonium-bearing carbides. This problem might be aggravated when a sodium bond is used, as carbon might be transferred through the sodium (acting as a mass transfer agent) to the can.

Metal fuels have the advantage of the highest densities, which would lead to a good breeding ratio, and consequently an advantageous effect on the economics of the reactor system providing the fuel will operate with sufficiently high can temperatures to permit economic power generation. The limitation on burn-up with metal fuels can be overcome by enclosing it in a sufficiently strong can, leaving about 30% voidage within the can to take up a certain amount of swelling. Further swelling is then restrained by the can, although it may be necessary to design the fuel element to undergo a limited creep strain during its lifetime.

While the feasibility of the use of metal fuels in a strong can has been demonstrated, more work is required in the optimisation of the design. It is known that swelling can be restrained up to 5% burn-up but the minimum amount of canning material and/or voidage to achieve this needs to be resolved. Manufacture of the fuel may present problems as close tolerances are likely to be required; because, although hot spots caused by areas of poor heat-transfer are likely to be self-healing, there can be no guarantee that distortion or other damage may not be caused at adjacent regions of good heat transfer by high fuel-can interface temperature, before correction occurs.

The main problem in the design of such a fuel element is in the choice of a suitable can. Certain "Nimonic" alloys (such as "Nimonic" 80) have sufficient strength, but suffer the disadvantage of poor compatibility, particularly with U-Pu alloys. Moreover, the mechanical properties which are largely dependent upon the microstructural stability of these alloys have yet to be proved during irradiation under fast flux conditions, and it is possible that rapid over-ageing and consequent loss of strength may occur. Alternatively, molybdenum appears to offer a sufficiently high creep-strength (but not as high as unirradiated "Nimonic" 80) and good compatibility with sodium at 650 C. The technology of molybdenum tube fabrication is, however, not well advanced at present. Yet another possibility is the use of duplex tubes whereby a high creep-strength tube is lined with a compatible refractory-metal tube. The protective layer may be deposited or formed by pressurising one tube within another, possibly with an intermediate bonding alloy. If the former course is chosen, care must be taken to ensure that an impermeable coating is formed, and in either case the behaviour of the fuel element under thermal cycling needs to be fully investigated, as a serious mismatch in thermal expansion characteristics appears to be inevitable.

Irradiation Testing

Enriched uranium specimens 0.1-inch diameter containing a longitudinal slot giving a total void space amounting to 30% of the can volume and clad in "Nimonic" 80 have been irradiated in DMTR at design temperatures of 490 C at the surface and 630 C at the centre. The ratings were approximately 500 watts/gm and the maximum burn-ups achieved were slightly in excess of 4%.

On piercing the can after irradiation approximately 5% of the total fission product gases were released. Metallographic examination showed that the slot had almost completely disappeared (Figure 7) and axial extrusion of the fuel had occurred, to fill the domed end caps (Figure 8). The interfaces between the original uranium pellets have also been entirely eliminated. Porosity was evident throughout the specimens, but its form varied according to the position in the pins, three zones being evident, namely: a narrow band at the surface where the bubbles are small and widely spaced; an underlying zone, 0.015-inches deep, densely populated with bubbles; and a central zone where large irregular pores become increasingly prevalent (Figure 9). Contact between fuel and can was everywhere excellent, and incipient reaction could be detected in some cross sections while adhesion of the fuel and the can is so good at other points that rupture in the uranium has occurred on cooling.

On heating an irradiated specimen for one hour at 630 C no significant distortion or dimensional changes occurred. The specimen was subsequently pierced at its mid-point with a 1/16-inch drill and heated for 48 hours at 510 C to determine whether extrusion of the fuel was a likely consequence of can rupture during irradiation. No extrusion occurred and the diameter of the can was unchanged. No in-pile experiments with defective cans have been performed, but these are necessary since it is believed that the plastic behaviour of the fuel will be very different under irradiation, and that extrusion through a defective can is likely. No changes due to irradiation could be detected in the microstructure of the "Nimonic" can and end plugs, although higher hardness values (510 VPN near the fuel) were obtained than would be expected. It should be remembered, however, that these irradiations took place in a thermal flux and results are not necessarily indicative of behaviour under fast flux conditions.

CONCLUSIONS

Four fuel types are at present considered feasible for use in fast reactors, namely

- (a) Oxides
- (b) Cermets
- (c) Carbides
- (d) Metals

All these present some considerable technical problems of fabrication, containment, or irradiation stability; but the first two appear to be further advanced technologically than the latter two. Carbides and metals on the other hand appear to have a higher potential for the ultimate production of cheap nuclear power from fast reactors.

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FIGURE 7

Cross Section of
Irradiated Uranium
Pin Showing Residue
of Original Slot
x 136

FIGURE 8

Longitudinal Section
of Irradiated Uranium
in x 75

FIGURE 9

Distribution of
Porosity in Irradiated
Uranium Pin x 56

PLUTONIUM AS A FUEL FOR THE LOS ALAMOS MOLTEN PLUTONIUM REACTOR EXPERIMENT

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LAM-1
C 10 31 12

The Los Alamos Scientific Laboratory has been engaged in the development of plutonium fuels for fast breeder power reactor applications. This program is directed toward nonfabricated fuel forms, principally metallic alloys in a molten form with the objective of eliminating costly fuel charges arising from repeated fabrication and distortions from irradiation damage. With the use of a fluid fuel, it is hoped that fuel handling and reprocessing can be carried out with simplified procedures and so realize significant economies in overall fuel costs.

The major first step in this program has been the construction and operation of LAMPRE I, a 1 Mw reactor to be used for environmental evaluation of fuels. This reactor has been described before the ANS by E. O. Swickard in 1959¹ and the experiences during the initial startup were reported by G. L. Ragan² and B. M. Carmichael³ in 1960. The present report is intended to give a brief recapitulation and a status report of the experience to the present. Some extrapolation to future plans is permissible, but it must be clearly stated that nothing in the present report should be construed as implying that the problems of handling molten plutonium fuels have been solved. Significant and, I believe, promising progress toward the solution of these problems has been made, but much work remains to be done.

LAMPRE I is a molten plutonium fueled reactor experiment built at Los Alamos and is currently operating at its design power of 1 Mw. The fuel is 24 kg of plutonium as an alloy, $\text{Pu}_{0.9}\text{Fe}_{0.1}$, contained in 140 separate tantalum capsules cooled by sodium. Figure 1 shows the schematic arrangement of the components. The fuel capsule is shown in Figure 2. The capsule is filled to a height of 6 inches with 175 g of alloy, leaving a 2-inch space for accumulation of fission product gases. Since the melting point of the alloy is about 412°C , the entire system must be constantly maintained above this temperature to avoid cyclic melting and freezing of the fuel. Figure 3 is a graph of early data taken of the density of the fuel as a function of temperature. As can be readily appreciated from the graph, the fuel alloy expands upon freezing, becoming less dense and may float to the surface. Just as in the case of water freezing, this process can form a plug which is capable of causing ruptures in confining spaces. The density changes in the solid phases can be controlled by the use of delta stabilizing additions such as aluminum, as has been reported by K. A. Gschneidner, et al.⁴ in the Grenoble Conference of 1960. This can be appreciated from

FIGURE 1

Schematic Arrangement of the Components

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FIGURE 2

Typical LAMPRE Fuel Capsule
(The wall is 0.028-0.030 inch thick)

FIGURE 3

Density of Pu-Fe Fuel as a Function of Temperature

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an examination of the phase diagram of the binary alloy which shows the solid state of the reactor fuel to be a mixture of the compound Pu_6Fe and plutonium metal with its multiple phase transitions occurring with temperature changes.

The problem of containing the molten fuel at temperatures of interest to reactor applications has been a most vexatious one. Only limited satisfaction can be derived from the apparent success in limiting the attack on the tantalum containers by the use of high purity fuel and container materials. The note of caution stems from the realization that the operating fuel, of necessity, will be highly contaminated with many chemical species. In static out-of-pile tests, there are indications that calcium and magnesium may be detrimental to containment, although the detailed mechanism is not known at this time. Whether or not the fission product members of the same chemical group, strontium and barium, have the same effect remains to be seen. This work is under way now that LAMPRE I is operating as a test reactor.

The fluid nature of the fuel has introduced some operating problems which are unique to the specific design of LAMPRE I. Because the fuel has no strength, the accumulation of some fission products nucleate within the reacting region and produce entrained gas bubbles. These bubbles, for the most part, rise to the surface by buoyancy and by convection currents in the fuel established by temperature gradients. In the first fuel loading, the fuel was complicated by the presence of solid additives of carbon or PuC to inhibit corrosive attack of the tantalum by the fuel at high temperatures. These additives interfered with the release of accumulated gas. Figure 4 shows radiographs taken of capsules which had been operated in the reactor at various powers. The separation of large portions of the fuel regions resulted in reactivity changes with time as shown in Figure 5. This fuel loading, referred to as the Mark I core loading, accumulated approximately \$10 of "lost" reactivity before it was replaced by an improved fuel without the presence of additives. At no time, however, was there any indication of instability in this froth or low density portion of the fuel. A collimated gamma ray scan of a fuel element removed from the reactor using the technique described by D. M. Holm⁵, shows a strong peak of lanthanum activity located at the surface of the fuel. Figure 6 is a typical scan taken of such a capsule. The activity level as a function of capsule length is convincing evidence of the circulation within the fuel and of the separation of some fission products, notably barium and lanthanum.

In April of this year the reactor was reloaded with fuel capsules characterized by improved purity and absence of solid anticorrosive additives. This new loading, termed Mark II, has demonstrated the same qualitative behavior of reactivity changes with irradiation as the first core. The disengagement of evolved gases is cleaner, as shown in Figure 7 and results in a reactivity change with time as given in Figure 8 where it is compared with the first core loading and, in Figure 9 where a longer

FIGURE 4

Radiograph of Exposed Fuel Containing Anticorrosion Additives
(Mark I Fuel)

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Changes in Reactivity of LAMPRE I with Power History

FIGURE 6

Collimated Gamma-Ray Scan of Irradiated LAMPRE Capsule

Negative Number 620203

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FIGURE 7

Radiograph of Exposed Purified Fuel

Negative Number 616524

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FIGURE 8

Reactivity Loss During Reactor Operation

FIGURE 9

Reactivity Loss Versus Integrated Power

exposure period is covered. In this last run an apparent temporary equilibrium had been established between the rate of formation and the rate of release of the gas. At the present time, more than 0.2% burnup, averaged over the core, has been realized in the fuel.

The reactor has been designed to remain serviceable in the event of complete fuel element failures. This provision includes an alpha-tight enclosure on top of the reactor for fuel element withdrawal and a completely contained fuel transfer system. Within the reactor, gross leakage of a fuel capsule resulting from any type of fuel failures will be contained in a catchpot below the core in a low temperature region. Samples taken to measure the activity of the cover gas system have revealed the presence of characteristic fission product gases. The origin of these fission products can not be determined with certainty, but may arise from fuel penetrating some defective capsules, or from minor leaks in the plutonium capsule welds. In any event, the problem is a minor one and full within the designed functions of the testing reactor.

In this description of LAMPRE I and its operation, it should be pointed out that the reactor has been designed to provide a testing function and should not be regarded as a prototype design. The particular fuel arrangement of separated capsules does eliminate the problem of fuel distortions with irradiation, but gas build-up within the capsule is just as severe a problem. Indeed, the design chosen to provide the maximum flexibility for fuel testing combines most of the disadvantages of solid fuels and of liquid fuels. With this capsule design, one is limited in burnup, is faced with fuel manipulation problems, and at the same time must have an absolute leak proof container for liquids. The objective of the program is to evolve a design which can take full advantage of the fuel mobility.

Laboratory experiments directed toward the circulation of plutonium have been started and will continue in the future, culminating in a reactor test if the results continue to be encouraging. One of the ways by which the fuel has been circulated is the use of a sodium injection lift pump. Several small scale experiments, and one 3 kg fuel circulation experiment, have been tried with varying degrees of success. The most recent experiment, called the plutonium test assembly (PTA), shown in Figure 10, was conducted by a group working under J. E. Deverall at the Los Alamos Scientific Laboratory. The sodium, carefully purified before the experiment, was used in an annular lift pump to circulate the Pu-Fe fuel in a closely coupled circuit constructed to represent a possible reactor geometry. Although the circulation flow rate of the fuel was modest (estimated at a few gallons per minute), it would be sufficient to provide for forcible disengagement of contained gas. If circulation for heat removal is attempted, the required flow rate is much greater and the problems much increased. In the experiment just described, the plutonium flow was observed directly with the aid of a transmission gamma-ray image converter in conjunction with a closed circuit television viewer. The experiment was operated for several days and conclusively established the feasibility of moving the Pu-Fe fuel both by pressure and by the lift pump action.

FIGURE 10

Function Control Panel of PTA

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The fuel which has been described so far is the concentrated alloy, $\text{Pu}_{0.9}\text{Fe}_{0.1}$ which has a density of about 16 g/cm^3 . This concentrated nature of the fuel presents severe restrictions in heat removal when designs are pushed toward high performance as must be done to achieve economical performance. If the fuel is contained in cylindrical tubes, and if the limitation is set by thermal stresses in the tube walls produced by the heat flux, it can be easily shown that the allowable capsule diameter must be inversely proportional to the plutonium density of the fuel for a given performance level. With the Pu-Fe fuel alloy, this fact of life forces the designer to small fuel ligaments which make fuel circulation difficult, if not impossible. In order to alleviate this restriction, a search has been made for molten plutonium alloys having a reduced plutonium concentration. Two possible choices are now receiving full attention in the determination of their physical properties. These are ternary alloys of Pu-Co-Ce and Pu-Cu-Ce.⁶ In both of these alloys, the plutonium can be varied from 2 to 4 g/cc at temperatures which are not excessive and so permit the conceptual design of high performance reactors. Test capsules containing the Pu-Co-Ce alloy have been introduced into the core of LAMPRE I for initial evaluation.

The preliminary results of the container testing that has been done with the dilute fuels show a much reduced attack of the intergranular penetration type but, in the case of the Pu-Co-Ce alloy, a possible temperature dependent attack mechanism attributed to the cobalt and an associated mass transfer problem may be present. On the other hand, so far as is known, copper is completely inert with respect to tantalum and so the Pu-Cu-Ce alloy is not expected to have the same problem as the Pu-Co-Ce alloy. This exploratory work is just beginning and it is too early to quote results.

The extension of the fuel testing and testing of conceptual core designs will be difficult, if not impossible, with the present equipment limitations. It is for this reason that the Los Alamos Scientific Laboratory has embarked on an ambitious program of constructing a versatile core test facility to be used in the plutonium fuel program. The Fast Reactor Core Test Facility, now undergoing its required safety analysis, is intended to provide a sodium heat removal system that can be coupled to a succession of core designs. In this way the cost and effort of duplicating expensive, but noncontributing, heat removal facilities will be avoided. The capacity of the Fast Reactor Core Test Facility has been arbitrarily set at a total power dissipation of 20 Mw. This figure, which is difficult to justify on any absolute scale is a compromise between small systems of 1 Mw and practical prototypes which must be sized at greater than 100 Mw. It is fully realized that it will be impossible to solve all of the problems of reactor design at this power level, but many important factors of fuel performance can be evaluated in a scaled test where the fuel is operated at a specific power, either watts per gram or watts per cubic centimeter, typical of large power plant designs.

The first tests which are conducted in this Fast Reactor Core Test Facility may not be the most adventurous that can be imagined for molten plutonium fuels, but it has been decided that this method of development will afford the most progress over a reasonable number of evolutionary core designs.

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THE VALUE OF PLUTONIUM IN FAST REACTORS

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INTRODUCTION

This study deals with the value of plutonium in fast reactors. This plutonium value is set by relating to U-235 which is assumed to be a material with an already established value. This study does not present information on the influence of plutonium on fuel cycle costs for fast reactors.

The basic data used for this study of plutonium values in fast reactor systems were developed by R. Avery, G. Fischer, and B. Toppel of Argonne National Laboratory. The preliminary reporting of the codes and the initial cost studies was presented at the June, 1962 meeting of the American Nuclear Society in Boston, Massachusetts.^{1,2} This work is being developed into detailed reports to be issued by these people as Argonne National Laboratory reports.

The code used for the physics calculation is called CYCLE. This is an IBM-704 code written in FORTRAN. The code was developed for the study of fuel cycles in fast reactors. It is particularly oriented to the study of fuel recycles in fast breeder reactors operating on either the Pu-U-238 or the U-233-Th chain under a wide variety of fuel management procedures. The code alternately solves the long-term, one-group isotopic change equations and the one-dimensional, multigroup, multiregion neutron diffusion theory equations (using the Argonne Re-122 code) to determine the properties of the equilibrium fuel cycle. The equilibrium cycle is the asymptotic repeating cycle for the selected mode of fuel management, choice of feed materials, and reprocessing procedure. Criticality is achieved for the system having that composition corresponding to the average composition of an equilibrium cycle.

Twelve isotopes are considered in the isotopic change equations. The first five correspond to the chain U-238, Pu-239 through Pu-242, and the next five to the chain Th-232, U-233 through U-236. Isotopes eleven and twelve correspond respectively to fission products and a burnable poison. Radioactive decay is not considered; therefore, there is no dependence upon reactor power level, but only on integrated flux time.

The code allows a maximum of 32 materials (including the required 12 in the isotopic change calculations), 30 energy groups, and 15 regions.

The output includes the regional initial, average, and final volume fractions for all materials, the average neutron inventory, and the material input-output inventory.

The CYCLE data used for this study have been made in spherical geometry and with 16-energy groups. The cross sections are basically those

of Reference 3 except that ELMOE⁴ derived cross sections have been used to more accurately account for resonance structure effects on the transport and elastic transfer properties of the light elements.

PROCEDURE

The plutonium value calculations are made by equating the variable costs in a given reactor fueled with U-235 with the same category of costs in the same reactor operated in an identical manner, except that it is fueled with plutonium.

The following assumptions are made:

1. Fuels of comparable reactors have the same fuel cycle costs. That is, all fabricating, processing, storage, and handling charges are identical for fuels with either U-235 or plutonium as the prime fissile material. While no fuel cycle need be specified for such an assumption, the EBR-II pyrometallurgical cycle for metallic fuels is an illustration of one that might be used.
2. The price of U-235 is the established base for setting the value of plutonium. The price for U-235 is taken as unity.
3. Since the blanket material in all reference reactor systems is removed at the same concentration of plutonium buildup, the blanket cost contribution to the fuel cycle can be ignored. Whatever the blanket cycle costs are in comparable reactors fueled with U-235 or plutonium the costs per unit of blanket plutonium recovered will be identical.
4. The only parameters of significance are the annual inventory charge on the fuel and the quantity of fuel in inventory. Two combined extremes were taken. The low one has the annual inventory charge of 4.75%; the quantity of fuel in inventory, both in and out of the reactor, is 1.5 times the core loading. The high extreme has the annual charge of 12%; the quantity of fuel in inventory is 10.0 times the core loading.
5. The plant factor is 80%. This point has no bearing on the value of plutonium. It merely affects the annual quantity of surplus plutonium and makeup fuel shown in the tables.
6. In a reactor fueled with U-235, all plutonium recovered at the end of a cycle, both in core and blanket, is sold. All fissile makeup is purchased U-235. In a reactor fueled with Pu-239, the burnup in the core at the end of a cycle is made up with Pu-239 recovered from the blanket. Thus, there is no burnup charge. The factors to be considered are the inventory charge and the income from the sale of surplus plutonium. In a reactor fueled with low-quality plutonium, two methods were used initially. In the first method, it is assumed that all blanket plutonium is sold and the core burnup is supplied from an outside source. In this instance, the value of the blanket plutonium is assumed to be the same as that calculated for the comparable core

where the feed plutonium was pure Pu-239. The value of the low-quality plutonium is then the single unknown. In the second method it is assumed that the value of all plutonium isotopes is equal. Thus, the low-quality plutonium fed to the core to compensate for burnup can, for value calculations, have an equivalent quantity subtracted from the blanket production. This latter method is used for all of the values presented except where noted otherwise.

7. U-235 and all plutonium isotopes are taken as fissile material.

With the foregoing assumptions, the only factors significantly influencing costs in the fuel cycle are the fissile content of the core, the quantity of fissile material from outside the reactor system purchased to compensate for core burnup at the end of a cycle, and the surplus plutonium from the system. The value of plutonium can be calculated by equating the applicable portion of these three cost factors in two comparable systems, one of which is U-235 fueled.

For example, the income from the sale of plutonium from a reactor having a metallic fuel containing U-235 minus the inventory and the burnup costs are equated to the income from the sale of the surplus plutonium from a reactor having a metallic fuel containing plutonium minus the inventory charge. (There is no burnup charge since the core burnup is supplied by the blanket-formed plutonium.) In these two metallic-fueled reactors, the factors of power level, the core volume fractions of structure, fuel and coolant, the core volume, the blanket volume and composition, the core burnup fraction, and blanket plutonium buildup are identical for each reactor case.

The reference systems had as their background the development of cores having a thermal capability in the core alone of 175, 350, and 700 electrical megawatts. Since electrical production has no purpose in this study, only the indications of the thermal output are shown. In all cases compared to calculate plutonium value, the core power levels are equal. However, there may be as much as 5% variation in overall reactor power level due to variations in the power generated in the blanket.

RESULTS

The characteristics of the reference cases are given in Tables I and II. The comparable electrical power output resulting from core power alone results from assuming that the carbide and oxide cores can produce coolant temperatures to yield a 40% cycle efficiency where the metallic core can operate at a temperature to produce 35% cycle efficiency. This statement is an explanation of the variations in the table, but has no bearing on the calculations for this study. The variation in overall reactor power can be seen in Table I. A plutonium-fueled reactor in the small sizes studied generated about 4% more overall power than did its U-235 counterpart due to the more rapid rate of plutonium production in the blanket.

TABLE I
CHARACTERISTICS OF REFERENCE FAST POWER BREEDER REACTORS

Fuel Type (Core and Blanket):		Carbide			Oxide			Metal		
Average Core Power Density, Mw/1		1.0			0.5			1.0		
C	Volume, liter	438.0	875	1750	875	1750	3500	500	1000	2000
	Power Fraction-Range	0.68	0.73	0.78	0.76	0.80	0.85	0.74	0.78	0.82
		0.73	0.76	0.80	0.79	0.83	0.87	0.78	0.81	0.85
O	Fuel Volume Fraction	0.26			0.32			0.23		
R	Na Volume Fraction	0.61			0.52			0.66		
E	SST Volume Fraction	0.13			0.16			0.11		
	Burnup Spec.	0.05			0.05			0.02		
B	Radius, cm	45			50			40		
L	Fuel Volume Fraction	0.65			0.65			0.65		
A	Na Volume Fraction	0.20			0.20			0.20		
N	SST Volume Fraction	0.15			0.15			0.15		
K	F. P. Buildup Spec.	0.0125			0.0125			0.005		
E										
T										

TABLE II
CHARACTERISTICS OF REFERENCE
FAST POWER BREEDER REACTOR

Core Feed:	U-238 + U-235 U-238 + Pu-239 U-238 + 40%-239, 25%-240, 15%-241, 20%-242
Blanket Feed:	99.8 U-238, 0.2% U-235
Structural Material in Core and Blanket:	Stainless Steel
Coolant in Core and Blanket:	Sodium
Core Burnup per Cycle:	5% Carbide and Oxide, 2% Metal
Fuel Lost in Process and Fabrication per Cycle:	2%
Blanket Buildup per Cycle:	1.25% Fission Products, Carbide, and Oxide 0.5% Fission Products, Metal
Fission Products Extracted per Cycle:	95%

Of the total data generated using the CYCLE code, only those cases using a cyclic mode of fuel managements are used. The fuel management mode involves five core and five blanket zones moving in an out-in direction.

Tables III, IV, and V present the comparable quantities used for the carbide, oxide, and metal cores, respectively. The breeding ratios are shown only to give something of the relative magnitude of the plutonium formed. The plutonium production and core burnup are calculated from the material balances given in the problems using the CYCLE code. The surplus plutonium from the plutonium-fueled cores represents only that amount not needed to sustain the reactor. The surplus plutonium from the U-235 fueled cores represents all the plutonium available at the end of core and blanket cycles. Since the U-235 fueled cores buy the quantity to compensate for burnup from outside the reactor system, these are the only reactors listing a core makeup.

TABLE III
CARBIDE FUEL CORES

<u>Core Volume, Liters</u>	<u>Fuel Feed</u>	<u>Breeding Ratio</u>	<u>Core Load Fissile, kg</u>	<u>Surplus Pu, kg/yr</u>	<u>Core Makeup, kg/yr</u>
438	Pu-239	1.83	241	152	146
	U-235	1.30	371	210	
	L. Q. Pu	2.01	399 (194)	147	
875	Pu-239	1.81	392	280	283
	U-235	1.27	580	397	
	L. Q. Pu	1.91	606 (340)	273	
1750	Pu-239	1.79	660	510	543
	U-235	1.25	947	745	

NOTE: Quantities in parentheses are Pu-239 and 241 content only.

TABLE IV
OXIDE FUEL CORES

<u>Core Volume, Liters</u>	<u>Fuel Feed</u>	<u>Breeding Ratio</u>	<u>Core Load Fissile, kg</u>	<u>Surplus Pu, kg/yr</u>	<u>Core Makeup, kg/yr</u>
875	Pu-239	1.67	378	117	151
	U-235	1.16	553	184	
	L. Q. Pu	1.78	622 (323)	111	
1750	Pu-239	1.65	639	213	284
	U-235	1.14	899	346	
	L. Q. Pu	1.69	967 (582)	207	
3500	Pu-239	1.62	1126	389	545
	U-235	1.12	1536	688	

NOTE: Quantities in parentheses are Pu-239 and 241 content only.

TABLE V
METAL FUEL CORES

<u>Core Volume, Liters</u>	<u>Fuel Feed</u>	<u>Breeding Ratio</u>	<u>Core Load Fissile, kg</u>	<u>Surplus Pu, kg/yr</u>	<u>Core Makeup, kg/yr</u>
500	Pu-239	2.07	281	171	159
	U-235	1.41	453	240	
	L.Q. Pu	2.27	416 (234)	166	
1000	Pu-239	2.07	456	329	310
	U-235	1.40	711	464	
	L.Q. Pu	2.15	612 (408)	325	
2000	Pu-239	2.05	770	630	600
	U-235	1.39	1162	893	

NOTE: Quantities in parentheses are Pu-239 and 241 content only.

The inventory charges used are representative at the lower end, 4.75%, of the current AEC rental charge for fuel and at the higher end, 12%, of the possible charge with private ownership of fuel. The lower range of fuel inventory, 1.5 times the quantity in the core, is representative of the quantity which can exist in a fuel cycle having a process such as a pyrometallurgical one and where fabrication and shipping times are short. The higher range of fuel inventory, ten times the quantity in the core, is representative of processes, such as aqueous ones, requiring appreciable cooling, processing, fabrication, and shipping times. However, for all cases presented here, a factor 10 is too high to be realistic. Of the cases used in this study, none are likely to have inventories in excess of five core loadings. To reach any inventory quantities as high as ten times that in the core, the core burnup would probably be less than 1% when the power density is high, probably of the order of 1 Mw per liter. So the low end is a good illustration of potential conditions; the high end illustrates an extreme, that is, a factor of two to five higher for the range of power densities and burnups presented. The product of the annual inventory charge and the inventory quantity is called the inventory cost factor.

As stated previously, the calculations of the plutonium value can be made by assuming all isotopes of either equal or unequal value. Table VI shows the contrasting difference in value of the low quality plutonium fuel used in this study. When the equal value method is used, low-quality plutonium feed to the core has the same unit value as the high quality plutonium produced in the blanket. Thus, an equation can be established directly between a low-quality plutonium feed case and its U-235 counterpart. When the unequal value method is used, it is necessary to first solve for the value of Pu-239

from a case having Pu-239 feed. Then, in turn, assigning that value to the blanket plutonium for the reactor version having low-quality plutonium feed. The difference between the two methods results in 15-20% lower value for the particular low-quality plutonium used here when the unequal isotope value assumption is used and the low inventory cost factor is used. When the high inventory cost factor is used, the difference is only about 5%.

TABLE VI
RELATIVE PLUTONIUM VALUES

	Pu-239		Low Quality Pu			
			All Pu Isotopes Equal Value		Pu Isotopes Unequal Value	
	Low Charge	High Charge	Low Charge	High Charge	Low Charge	High Charge
Carbide - Medium	2.23	1.67	1.94	1.15	1.63	1.10
Oxide - Medium	1.94	1.51	1.67	1.05	1.34	1.01
Metal - Medium	2.14	1.70	1.96	1.33	1.66	1.28

Table VII gives the relative plutonium values of all the reference cores. In all cases where the feed is Pu-239, the value shows the influence of change in breeding gain with size. The metal cores have little or no change in plutonium value over the range of core sizes considered; the carbides have a small decrease in value as the core sizes increase; and the oxide cores have a significant decrease in plutonium value as the core sizes decrease.

In the size range considered for the metal reactors, the neutron spectrum changes little. As a consequence, the breeding rate is fairly constant. The spectrum softening which occurs with carbide cores and to a slightly greater degree with oxides affect the ultimate yield of surplus plutonium to a greater degree than is the core loading affected by larger size. Thus, the inventory cost becomes a more dominant factor.

Where the feed to the core is the very low-quality plutonium used in this study, the reverse direction takes place. As the core sizes increase, the plutonium value increases. This condition results when all four plutonium isotopes are considered in the calculation as fissile material. In this instance, the rate of decrease of the core loading is greater than rate of decrease of the surplus fissile material. The inventory influence decreases at an appreciable rate; so as the core size increases, the plutonium value will increase. If only the Pu-239 and 241 isotopes were considered directly in the calculations, the reverse condition of plutonium value would not occur.

TABLE VII
RELATIVE PLUTONIUM VALUES

	<u>Low Inventory Cost Factor</u>		<u>High Inventory Cost Factor</u>	
	<u>Low Inventory Quantity^a</u>		<u>High Inventory Quantity^b</u>	
	<u>Low Inventory Charges^c</u>		<u>High Inventory Charges^d</u>	
	<u>Pu-239</u>	<u>L. Q. Pu</u>	<u>Pu-239</u>	<u>L. Q. Pu</u>
Carbide - Small	2.29	1.88 (3.4)	1.70	1.09 (2.0)
- Medium	2.23	1.94 (3.5)	1.67	1.15 (2.1)
- Large	2.16		1.63	
Oxide - Small	2.32	1.62 (3.0)	1.57	1.00 (1.8)
- Medium	1.94	1.67 (3.0)	1.51	1.05 (1.9)
- Large	1.73		1.45	
Metal - Small	2.15	1.85 (3.4)	1.73	1.23 (2.2)
- Medium	2.14	1.96 (3.6)	1.70	1.33 (2.4)
- Large	2.15		1.68	

^a1.5 x quantity in core

^b10.0 x quantity in core

^c4.75%

^d12.0%

NOTE: Numbers in parentheses in L. Q. Pu columns represent plutonium value based on Pu-239 and 241 content only.

Figure 1 is a plot of the change in plutonium value with increasing inventory influence due to increasing the quantity of fuel in inventory and/or increasing the annual charge applied to inventory cost. Figure 1 covers the value of Pu-239 feed and the low-quality plutonium feed for the medium-sized carbide core considered in this study. The lower two curves are for the low-quality plutonium calculated by the equal and unequal isotopes values approach.

FIGURE 1

Relative Plutonium Value versus Inventory Cost Factor
Carbide Core - Medium

Figure 2 illustrates something of the magnitude of the additional allowable fuel cycle costs for a plutonium over a U-235 cycle. The illustration is the medium metal core. It represents the difference in value between U-235 and Pu-239 translated into allowable fuel cycle cost. The magnitude of the possible cost differential shown in Figure 2 is somewhat larger than would actually exist since the background physics calculations

are ideal and the resulting breeding gains high. However, the potential of a significantly improved economic condition exists with plutonium even though in the long run, the plutonium fuel has a higher unit fabrication cost than U-235 fuel.

FIGURE 2

Difference in Value Between U-235 and
Plutonium Applied to Fuel Cycle Cost

Tables VIII, IX, and X have the data of Pu-239-fueled cores in which some characteristics have been significantly changed from that used in the comparable reference core. The changes are noted in the tables.

TABLE VIIIVARIATION FROM REFERENCE CARBIDE CORES

Fuel Feed - Pu-239

<u>Core Volume, Liters</u>	<u>Breeding Ratio</u>	<u>Core Load Fissile, kg</u>	<u>Surplus Pu, kg/yr</u>	<u>Variation from Reference</u>
438	1.83	241	152	Reference
438	1.82	245	154	Double Core Burnup
583	1.85	319	140	Lower Power Density (25%)
875	1.81	392	280	Reference
	1.77	404	216	Increase Cycle Loss (5x)
	1.76	397	264	Decrease f. p. Extraction (10x)
	1.79	398	276	Double Core Burnup
	1.60	460	199	Substitute Nb for SS
	1.72	385	243	Decrease Blanket Fuel Density (23%)
1750	1.79	660	510	Reference
2333	1.77	984	487	Lower Power Density (25%)

TABLE IXVARIATION FROM REFERENCE OXIDE CORES

Fuel Feed - Pu-239

<u>Core Volume, Liters</u>	<u>Breeding Ratio</u>	<u>Core Load Fissile, kg</u>	<u>Surplus Pu, kg/yr</u>	<u>Variation from Reference</u>
875	1.67	378	117	Reference
1095	1.63	427	113	Lower Power, Fuel Density (20-32%)
1750	1.66	762	106	Lower Power Density (50%)
1750	1.6	639	213	Reference
	1.61	661	151	Increase Cycle Loss (5x)
	1.59	650	194	Decrease f. p. Extraction (10x)
	1.62	649	209	Double Core Burnup
	1.63	619	234	Double Blanket Buildup
	1.33	825	100	Substitute Nb for SS
	1.58	643	187	Decrease Blanket Fuel Density (23%)
3500	1.62	1126	389	Reference
4375	1.57	1254	389	Lower Power, Fuel Density (20-32%)
7000	1.60	2589	360	Lower Power Density (50%)

TABLE X
VARIATION FROM REFERENCE METAL CORES

Fuel Feed - Pu-239

Core Volume, Liters	Breeding Ratio	Core Load Fissile, kg	Surplus Pu, kg/yr	Variation from Reference
500	2.07	281	171	Reference
667	2.11	371	166	Lower Power Density (25%)
1000	2.07	456	329	Reference
	2.02	470	193	Increase Cycle Loss (5x)
	2.04	459	311	Decrease f. p. extraction (10x)
	2.06	455	344	Double Core Burnup
	2.05	440	362	Double Blanket Buildup
	1.89	500	269	Substitute Nb for SS
	1.97	459	293	Decrease Blanket Fuel Density (23%)
2000	2.05	770	630	Reference
2667	2.06	1135	626	Lower Power Density (25%)

Table XI indicates the percentage change of plutonium value from the corresponding reference cases given in Tables VIII, IX, and X. Actually, this is not a change in the value from a comparable U-235 fuel case. This is a deviation from some selected standard. Three of the changes shown are actually not suitable for the type comparisons used. These are the cases where the core burnup is doubled and where the blanket is left in to a level in which the fission product concentration is doubled. In both variations from the reference, the rate of core or blanket material handled in the fuel cycle has decreased markedly. Thus, the unit cost of fabricating and processing is not a factor which can be neglected in a reasonably fair comparison. However, no such compensation was made in the calculations. As a result, the percentage changes listed for these cases are less positive than they should be.

In cases where the core power density is lowered, the core loading goes up, and as a consequence, the inventory cost has greater importance, and the value relative to the reference case goes down. In the oxide cases, where the power density is decreased by a factor of two, the percentage decrease is quite marked.

The cases having an increase in fuel loss per cycle have an obvious loss in plutonium value. The value change is greater when the inventory cost factor is low because the decrease in surplus plutonium has a greater influence in the calculations than when the inventory cost factor is high. In cases where the rate of fission product removal is less or where niobium is substituted for stainless steel as core structural material, the value

TABLE XI
PERCENT CHANGE OF PLUTONIUM VALUE
FROM REFERENCE PLUTONIUM CORES

	<u>Carbide</u>		<u>Oxide</u>		<u>Metal</u>	
	<u>Low Cgs</u>	<u>High Cgs</u>	<u>Low Cgs</u>	<u>High Cgs</u>	<u>Low Cgs</u>	<u>High Cgs</u>
*Double Burnup (small)	+ 6	- 1	---	---	---	---
*Double Burnup (medium)	- 2	- 1	- 3	- 2	+10	+ 3
Lower Power Density	-19	-23	-29	-46	-12	-22
Lower Power Density (small) (low fuel ρ)	---	---	- 8	-11	---	---
Lower Power Density (large)	-14	-19	-26	-52	-23	-31
Lower Power Density (large) (low fuel ρ)	---	---	- 2	- 8	---	---
Increase Fuel Loss/Cycle (medium)	-30	-12	- 8	- 4	-45	-18
Decrease f. p. Extraction (medium)	-10	- 3	-10	- 3	-10	- 3
*Increase Fissile Buildup in Blanket (medium)	+40	+13	+14	+15	+25	+ 8
Nb Structure Instead of SS (medium)	-37	-22	-31	-27	-27	-14
Decrease Blanket Fuel Density (medium)	-20	- 5	-13	- 3	-17	- 5

*These cases are not fair comparisons with their reference cores since there are appreciable changes in fuel process rates. The fuel cycle cost which is not significant when process rates are equal becomes a significant factor when comparisons are of cases where the rates are not equal.

decrease occurs for a similar reason. In these two types of changes, the plutonium production is decreased due to the added parasitic neutron absorption in the fission products or niobium. For those cases where the fuel density is decreased in the blanket, the plutonium value change is again in the same direction for the reason of decreased plutonium production.

CONCLUSIONS

The value of plutonium in fast reactors can be, even under some unfavorable conditions, at least equivalent in value to U-235, and under favorable conditions, twice the value of U-235. This condition applies to qualities of plutonium as low as that used in this study which contained 55% of the 239 and 241 isotopes. But an exact or narrow range of values for plutonium is not reasonable to state unless some conditions such as neutron spectrum, inventory quantity, and inventory charge rate are established. Without such conditions being set, the value of plutonium will vary. For example, it will decrease as neutron spectrum softens and inventory quantity and charge rate increase.

As the price of plutonium is set above its value, the U-235 fueled reactors are favored strictly from the standpoint that the income is improved by selling the high price fissile material and burning low priced. When the plutonium price is below the value, it becomes better to burn the plutonium priced below its value and receive any income possible from the surplus, rather than to burn high cost U-235 and have an income from the relatively low-priced plutonium.

The value calculations based on idealized physics calculations should be valid for fast reactors. The same relative position should exist in comparable reactor systems whether the comparison be made between the one-dimension spherical geometry approach or a two-dimension cylindrical geometry approach. Also, the value calculations are as good as is warranted by the present status of fast reactor data. The refinements which should be made are those involving extensive heat analysis to a degree that fair comparisons might be made between systems having different fuel compositions (e.g., metals compared to carbides). In addition, comparisons should eventually be made incorporating possible different fabrication and handling costs between U-235 and plutonium. The first study involving the cross comparison of fuels requires good data on fuels irradiation. Maximum operating temperatures and burnup limits are needed for metal and ceramic fuels both with and without significant quantities of plutonium. For both studies, there is a need for cost information such as fabrication costs with varying sizes and configurations of fuel. With this information, it is then possible not only to calculate plutonium's values under a wider set of conditions, but also, and more important, to make good calculations of the fuel cycle influence on power production costs.

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AN ASSESSMENT OF PLUTONIUM USE

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In several studies^{1,2,3} it has been shown to be practicable to design heavy-water natural-uranium reactors to operate with an average burn-up of 10,000 megawatt-days per metric ton of uranium (MWd/tonne U). The first full-scale 200-eMW reactor of this type is under construction and scheduled to be in operation by the end of 1964 and capable of $9000 \pm 10\%$ MWd/tonne U burn-up. The fraction of this power derived from the fission of plutonium would be about 43% at 8,000 MWd/tonne U and 49% at 10,000 MWd/tonne U. (See Appendix 1.)

The large program of gas-cooled graphite reactors undertaken in the U.K. is expected to derive about 3,000 MWd/tonne from natural uranium and about 23% of this energy will come from plutonium. From the total of about 20,000 thermal MW capacity scheduled for operation in 1966, the plutonium fission at 0.23 g Pu fissioned/MWd and 275 days/year would amount to 1,300 kg Pu fissioned per year, certainly the largest use expected in the near future. At the same time the annual production of unused plutonium would be at the rate of about 0.78 g/MWd or 4,300 kg Pu/year, and would become available for extraction two or three years later when the fuel is discharged. It would, however, be at a low concentration of about 2.3 kg Pu/tonne U.

Significant amounts of plutonium are also produced and fissioned in the operation of the Dresden type of reactor of which about 2,500 thermal MW capacity are committed for construction.

Except in the heavy-water type of reactor, more plutonium remains in the spent fuel than is fissioned in the types mentioned. It is, however, possible to extend the contribution from plutonium considerably in several types of reactor without introducing chemical extraction and recycling steps. For example, the seed-and-blanket system used in the Shippingport PWR in which the Core I blanket is associated with three successive seeds, has already fissioned more plutonium than remains in the natural uranium blanket.

In comparison with the above uses, which promise to expand very rapidly, other uses of plutonium as yet in prospect appear small.

To bring into focus the costs that would control the economic use of extracted plutonium, the effect of the USAEC base cost of \$17,000/day for extraction plant operation may be considered. Assigning such a cost to 3 kg Pu imposes a high unit price (\$5.67/g Pu) but spread over 6 kg Pu it might be acceptable (\$2.83/g Pu). On consideration of both economic balance and technical prospects the total amount to be extracted seems likely to be of the same order as, and certainly not many times more than, the rate of fissioning the extracted plutonium. It would follow that the use of extracted

plutonium would prove economic only when it amounts to about 6 kg Pu/day supporting a thermal power of 5,000 MW, or, at 30% efficiency, 1.5 million kilowatts of electric power. Since this represents quite large-scale use that will not develop very suddenly, it is expected that the use of extracted plutonium for power production will develop first with by-product plutonium. It is suggested that for several decades it will not prove economic to process spent fuel in which the concentration of plutonium is less than 4 or 5 kg/tonne U. It seems, however, likely that several types of spent fuel will be processed for other reasons and the by-product plutonium would be sufficient to meet the demand.

The high cost of extracting plutonium from spent fuel is attributable to the high levels of radiation from the fission products present with it. These high fields determine the nature of the plant, which can very properly be considered as a plant for the treatment and disposal of fission products. The complexity of the chemistry and the radioactivity of all side streams, washes and extracts, makes the plant costly to operate on a small scale. In pilot plant operations at Chalk River separating a few kilograms of plutonium, we never succeeded in reducing extraction costs to \$50/g Pu. On the other hand, when operations are taken to a large scale, a new complication arises from the hazards of criticality, not only in the main stream but also in cumulative segregations from the "losses".

The residues of spent fuel from reactors and the secondary products from processing plants, may very well contain large amounts of plutonium that it is not economic to process. The situation is economically comparable with that existing in mining and refining operations where ores and tailings of low grade are abandoned. Additionally in the case of plutonium the "ore" may be abandoned because of the cost of disposal of radioactive fission products. The "ore" may sometimes be more cheaply stored, particularly if it is a corrosion-resistant material such as UO_2 in a corrosion-resistant cladding such as Zircaloy or stainless steel.

To utilize any large fraction of the plutonium supply it is necessary to look for large-scale uses where plutonium would have sufficient value to compete with other separated fissile material and in particular with U-235 now priced at \$12.01/g. In most, if not all, such uses, however, plutonium has a lower unit value than U-235. It might still be used if it can be extracted at low cost and its low value is recognized so that inventory charges are not high.

Three special uses for separated fissile material will be reviewed, (1) enrichment of fuel for thermal reactors, (2) for reactor fuel of very long life; and (3) in fast neutron breeder reactors.

(1) Recycle in Thermal Reactors

The extensive analysis presented at the first United Nations Conference on the Peaceful Uses of Atomic Energy at Geneva in 1955⁴ will not be repeated here. It was shown that by designing for neutron economy and removing fission products from natural or recycled uranium after an energy

yield of 4,000 MWd/tonne U, and by reirradiating the plutonium in separate fuel elements in the same reactor, an ultimate total burn-up of 28,000 MWd/tonne of natural U could be reached. Moreover on a large scale of 1 to 2 million kilowatts, the whole process could be economical. Reasons for the choice of cycle and the cost allowances for reprocessing and refabricating fuel were given. No significant advance or improvement towards the objective of maximum burn-up by recycle in thermal reactors has been published. Towards the objective of lower fuel costs involving partial recycle, studies⁵ have shown that the highest value assignable to recycled plutonium arises when it is used to prolong the irradiation of natural or very slightly enriched uranium fuel. The value is higher, the higher the fabrication cost of the main fuel. In a competitive world, however, reactors with costly fabricated fuel would be driven from the large-scale uses. The use of plutonium for such purposes will therefore be restricted.

In a typical continuously-refuelled and neutron-economical reactor the addition of 1 g extra fissile material per kg of natural uranium would extend the attainable burn-up from 10 MWd/kg U to 13 MWd/kg U, so that an extra 3 MWd yield is assignable to the 1 g of extra fissile material. (See Appendix 2.) If the natural uranium fuel costs \$60/kg U or \$6/MWd, the extra 3 MWd is worth \$18. Against this \$18/g for the extra fissile material must be set the extra cost involved. For example, if the fuel is distributed as uniform enrichment, there may be costs attributable to (1) control of composition, (2) extra cost of wastage, and consequent recovery and recycle in fabrication, and (3) the toxicity if it is plutonium. If the fabricated fuel cost increased from \$60/kg U to \$70/kg U, the value of the fissile addition would be reduced from \$18 to \$8/g, and if the final cost of fabricated fuel were \$78/kg U or more, all value from the addition would have been cancelled unless some subsidiary advantage such as reduced operating costs, e.g., fuel changing, can be adduced for the enriched fuel. In general, lower costs may be achieved by recycling the plutonium as spikes or separate fuel elements as proposed for example in Reference 4. Whether plutonium would be chosen for this use would depend on the relative cost of using some other separated fissile material such as U-235 or U-233. These have their own disadvantages such as the toxicity of U-234 associated with U-235 and the penetrating radiations of radioactive daughters of U-232 associated with U-233.

Although \$60/kg U may now be considered a low price for fabricated natural uranium fuel, it may be bettered as the scale of fuel fabrication expands. At present natural uranium from the mine may cost \$6/lb U₃O₈, which is equivalent to \$15.60/kg U. At 10 MWd/kg U the cost component of the raw uranium is then \$1.56/thermal MWd (or at 35% efficiency 0.186 mill/ekWh). An extra 3 MWd/g of fissile material on this basis would justify only a value of $\$1.56 \times 3 = \$4.68/g$.

The low cost of natural uranium and the high burn-up attainable explains why other recycle uses of plutonium would be unlikely to be extensive.

The above discussion has been approximate, but exact evaluation requires detailed specific information. It appears impossible on general

grounds to demonstrate any specific value within the wide range from zero to \$18/g without specific information on the costs of fabricating and processing fuel.

(2) Reactor Fuel of Long Life

Both thorium-232 and uranium-238 are fertile materials that yield energy when irradiated by neutrons in a two-stage process; first a fissile material, U-233 or Pu-239, is formed and then energy released when these fission. After a very long irradiation the accumulation of fission products reduces the yield, but not before about 2.5 fissions per neutron supplied have been obtained from U-238 and more than 8 fissions per neutron supplied from Th-232.^{6,7} (See Appendix 3.)

Such long irradiations may be achieved by supplying fissile material as required to maintain the fission chain reaction. The eventual fissioning of even 10% or more of the thorium may be practicable in ThO₂ fuel diluted if necessary by ZrO₂.

So much cost might be saved by leaving the fertile fuel in the reactor without reprocessing or refabricating, that several studies of such systems have been made. The simple seed-and-blanket system already mentioned lies at one extreme and the fast breeder reactor at another. A third extreme is the fixed fuel reactor⁸ in which all the separated fissile material is inserted at the outset and no change is made to the fuel for 10 or 15 years.

Plutonium has special merit in the fast breeder reactor because of its high yield of neutrons per fission and this case will be evaluated in a separate section following. Plutonium was not found to have any net advantage for the fixed fuel reactor. Although it is necessary in such a design that the fissile material should initially be self-shielding so that some remains at the end of the irradiation, and this is easily achieved with plutonium because of its high thermal and resonance cross-sections, yet these high cross-sections lead ultimately to destruction that is too rapid.

Separated plutonium would have a value not much less than U-235 if used as spiking or seeding fuel in a less extreme design of reactor. If extracted plutonium is available more cheaply than U-235, its fabrication at high enrichment need not cost so much that the advantage is lost. Moreover the presence of Pu-240 is somewhat advantageous in this use because it slows up the rate of destruction to one that is comparable with U-235 as discussed in Reference 4.

(3) Fast Breeder Reactor

The difficulty of design of the fast reactor is to achieve a fuel cycle cost low enough to compete with other nuclear reactors. At one time it was hoped by designers that breeding could be so rapid that revenue from the sale of extra plutonium would offset the inventory charges. However, the need to reprocess the fuel, the cost and the time involved in such operations, seems to put such a prospect beyond reach. The following discussion shows the difficulty.

The fuelling cost for breeder reactors may be expressed as

$$c_{fs} = \frac{R - GBV/E}{24 Be} + \frac{IVa}{W 876.6 u} \text{ mill/kWh}$$

where

- R = recycle cost in \$/g Pu per cycle of Burn-up B,
- B = burn-up in MWd/g Pu supplied,
- e = thermal-to-electrical conversion efficiency,
- $24 Be$ = burn-up in eMWh/g Pu supplied,
- G = breeding grain = increase of Pu in g per g Pu destroyed,
- V = value of Pu in \$/g Pu,
- E = fission energy in MWd/g Pu destroyed,
- W = electrical power in kW,
- I = total Pu inventory in cycle in g Pu,
- a = % annual charge rate on inventory,
- u = utilization factor.

It follows by differentiation that

$$\frac{dc_{fs}}{dV} \text{ is positive unless } a < G \cdot \frac{W}{I} \cdot \frac{876.6 u}{24 eE} .$$

Supplying the following typical values

$$\begin{aligned} u &= 0.8, \\ e &= 0.35, \\ E &= 0.8, \\ G &= 0.2, \end{aligned}$$

this reduces to

$$a < \frac{W}{I} \cdot \frac{0.2 \times 700}{8.4 \times 0.8} = \frac{W}{I} \cdot 20.8 .$$

Now I/W is not likely to be less than 5 g Pu/eKW, so a must be less than about 4.

Conversely if $a > 4$ and $I/W > 5$ g Pu/eKW, the inventory charge will exceed the credit from the gain by breeding and will contribute to the fuelling cost a component that increases with the value assigned to plutonium. Since such circumstances appear probable, the value assigned to plutonium should be kept low.

Even assuming that advances in technique have brought this inventory component low, there remains the component from the cost of recycle, namely $R/24 Be$ mill/kWh. For $e = 0.35$ this component is $R/8.4 B$. If, for example, $B = 0.1$, i. e., 10% Pu burn-up per cycle, the recycle cost would have to be less than \$1.2/g Pu for this component to be less than 1.4 mill/kWh that might be considered the maximum allowable to be competitive.

It has been suggested⁹ that 20% PuO₂ + 80% U-238-O₂ could be taken to a burn-up of 10% heavy atoms fissioned, and with added burn-up from blanket fuel of UO₂, B could be as high as 1.0. Experimental justification for this appears to be still lacking. Irradiation of oxide fuel to 10% heavy atoms fissioned has been shown^{10, 11} to require mechanical constraint to limit elongation or swelling of fuel pins. Moreover, since fast neutron fission cross-sections are small, side reactions such as O-16(n, α)C-13 can introduce changes in fuel composition for which high temperature irradiation experience is lacking. Relatively small amounts of carbon or nitrogen diffusing into steel cladding at high temperature can lead to mechanical failure under stress. In view of the importance to fast reactors of high burn-up at high heat ratings the technology deserves intensive study and development.

For the present the economic prospect of the fast breeder therefore appears very dubious, and there is no promise yet that it would provide a large-scale use for extracted plutonium.

Conclusion

It is to be noted that comparisons have been made that depend on the relative costs of natural uranium and of processing and fuel fabrication with enriched fuel. It is to be expected that in the course of time when the richer ores have been exhausted the cost of natural uranium will rise. However the very low contribution to power cost of about 0.2 mill/kWh assignable to the raw uranium indicates that a considerable rise could take place before the conclusions would change. For example, if the cost of uranium were to increase to \$40/kg, that is, by a factor of $2\frac{1}{2}$, the contribution from raw uranium could still be less than 0.5 mill/kWh.

The comparison is not, however, sufficient for making an economic choice that must include other factors such as differences in the structural costs of reactors. These differences now are quite large, but in the long term are expected to become so much smaller (~\$20/ekW) that the relative costs of fuel cycles will largely determine the choice.

Acknowledgment

We would like to thank Mr. A. G. Ward for helpful discussion of these problems.

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APPENDIX 1ESTIMATING PLUTONIUM PRODUCTION AND FISSION

Approximate estimates for plutonium production and fission may be made from published core physics data for power reactors. (See, e.g., Directory of Nuclear Reactors; Vol. 1, Power Reactors, International Atomic Energy Agency, 1959.)

Estimates are most readily made from values of conversion ratio (γ), burn-up (B) in MWd/kg U, fast fission factor (ϵ) and U-235 depletion (D_5) in g/kg U or burn-up versus irradiation (nvt or n/kg).

Pu produced/kg U = $\gamma B' / 0.8$
 where 0.8 = MWd/g U-235 destroyed
 B' = thermal fission burn-up in MWd/kg U
 approximately $B' = B / [1 + (\epsilon - 1) \gamma_5 / (\gamma_8 - 1 - \alpha)]$.

For $\gamma_5 = 2.43$; $\gamma_8 = 2.84$; $\alpha = 0.348$ = (fast capture/fast fission)

$$B' = B / [1 + 1.629 (\epsilon - 1)]$$

Also, Pu fissioned/kg U = $(B' - 0.8 D_5) / 0.964$
 $= (B' / 0.964) (1 - 0.8 D_5 / B')$

where 0.964 = MWd/g Pu fissioned.

$$\begin{aligned} \text{Hence Pu remaining/kg U} &= (\gamma / 0.8 - 1 / 0.964) B' + 0.8 D_5 / 0.964 \\ &= (B' / 0.964) (1.205 \gamma - 1 + 0.8 D_5 / B') \end{aligned} \quad (1.1)$$

$$\text{Pu fissioned/Pu remaining} = 1 / \{ [1.205 \gamma / (1 - 0.8 D_5 / B')] - 1 \} \quad (1.2)$$

$$\begin{aligned} \text{Pu fission power/total burn-up} &= (1 - 0.8 D_5 / B') B' / B \\ &= \frac{1 - 0.8 D_5 / B'}{1 + 1.629 (\epsilon - 1)} \end{aligned} \quad (1.3)$$

For the examples used in the text

1. CANDU-type D_2O -moderated reactor

Irradiation n/kb	B MWd/kg U	B' MWd/kg U	D_5 g U-235/kg U	$1 - 0.8 D_5 / B'$	Pu Power Burn-up
2.0	8.104	7.722	5.218	0.459	0.438
2.5	9.990	9.518	5.752	0.5165	0.492

$$\sigma_5 = 0.6611 \text{ kb}; \quad \epsilon = 1.0289;$$

$$\text{Fast fission/thermal fission} = 0.0495; \quad B' / B = 1 / 1.0495.$$

2. Hinkley Point - UK CO₂-graphite-natural U

$$\epsilon = 1.029; B = 3.00 \text{ MWd/kg U}; B'/B = 1/1.0495 \text{ (as above)}$$

$$D_5 = 2.724; B' = 2.858 \text{ MWd/kg U}; 1 - 0.8 D_5/B' = 0.2377.$$

$$\text{Pu Power/Burn-up} = 0.2377/1.0495 = 0.2265.$$

$$\gamma = 0.85;$$

$$\text{Pu remaining/kg U} = (2.858/0.964)(1.205 \times 0.85 - 0.2377)$$

$$= 2.33 \text{ g Pu/kg U.}$$

$$20,000 \text{ thermal MW} \times 275 \text{ days} = 5.5 \times 10^6 \text{ MWd/yr}$$

$$\text{at } 3,000 \text{ MWd/tonne U} = 1833.3 \text{ tonnes U.}$$

$$1833.3 \text{ tonnes U at } 2.33 \text{ kg Pu/tonne U} = 4275 \text{ kg Pu.}$$

$$\text{Pu fissioned/Pu remaining} = 1/\{ [1.0242/0.2377] - 1 \} = 1/3.31.$$

$$\text{Pu fissioned/yr} = 4275/3.31 = 1290 \text{ kg Pu.}$$

3. PWR Shippingport.

From analysis of blanket UO₂ rods, P. S. Lacy and J. H. Leonard,
Trans. Am. Nuclear Soc. 3, 401, December 1960

$$\text{and } \epsilon = 1.073.$$

Rod No.	γ	B	B'	D ₅	$1 - 0.8 D_5/B'$	Pu Fissioned	Pu Remaining	
		MWd/ kg U	MWd/ kg U	g U-235/ kg U		Pu Remaining	Calc'd	Obs'd
1	1.06	6.35	5.675	4.056	0.428	0.504	5.00	4.56
2	1.24	3.80	3.396	2.718	0.360	0.317	4.00	3.792

after 5800 full power hours. Irradiation now exceeds 15,000 full power hours.

APPENDIX 2

EXTENSION OF BURN-UP IN CONTINUOUSLY-FUELLED NEUTRON-ECONOMICAL REACTORS BY ENRICHED FUEL

Attainable burn-up is discussed in DM-52 (AECL-651, June 1958) from which typical cases are illustrated in Figure 2.1. The figure shows the variation of neutron excess with burn-up for cases in which the neutron leakage is defined by a constant value, 0.0392 (see page 4, DM-52), for $B_g^2 M^2$. Three curves relate to the scale on the left where the neutron excess is expressed in barns per U-238 atom. The other three curves relate to the scale on the right expressed in barns per initial U-235 atom.

FIGURE 2.1

For illustration, two reactors, A and B, are selected with neutron wastage that limits the burn-up to 10,000 MWd/tonne. Reactor A is supplied with fuel enriched to 1.15 C_0 and Reactor B with natural uranium.

The effect of incremental enrichment is shown by the horizontal lines to meet the curves for higher or lower enrichment. The lines through A and B correspond to a pessimistic extreme where the neutron wastage in the reactor is kept at the same magnitude in bifa (barns per initial fissile atom) when the enrichment is increased.

The optimistic extreme is indicated by the lines through A' and B'. The same leakage is assumed but the wastage is kept at a constant magnitude in barns per U-238 atom.

Any practical case is likely to lie between these extremes because increased enrichment leads to increased flux depression in the fuel and therefore relatively greater wastage in structural materials when expressed in barns per U-238 atom. It would, however, imply excessive self-shielding if extra fissile material increased the total wastage in proportion to the total amount of fissile material in the fuel. Such a proportional increase is implied by setting the wastage constant in bifa and that is accordingly the pessimistic limit.

It will be seen from the figure that for Reactor B the burn-up is increased by the incremental enrichment of 0.15 C_0 to 13 MWd/kg U in the pessimistic case and to 13.7 MWd/kg U in the optimistic case. The burn-up of the other (AA') is correspondingly increased to 12.4 MWd/kg U in the pessimistic case and to 13.3 MWd/kg U in the optimistic case when the enrichment of the fuel supplied is raised by 0.15 C_0 to 1.30 C_0 .

The relative increase in burn-up is therefore greater for BB' (i.e., for the more neutron-economical reactor) than for AA', but the value of the enrichment is likely to be greater for AA' because its fuel is more costly.

On the other hand it may also be noted from the figure that in the reactor AA' its initial enrichment of 0.15 C_0 was of considerably greater value than the next addition of 0.15 C_0 . In the case of A' the initial 0.15 C_0 raised the burn-up from 5.8 MWd/kg U to 10 MWd/kg U.

It is of interest to calculate the value that can be assigned to the incremental enrichment per gram of U-235. First it is to be noted that the increase of 0.15 C_0 means that $0.15 \times 7.115 = 1.066$ g U-235 is added per kg U. The value of the extension ΔB of the burn-up B is $P\Delta B/B$ \$/kg U where P is the total cost of the fabricated fuel in \$/kg U. Expressed in \$/g U-235, the value is $P\Delta B/1.066$ B and is therefore

for	A	$0.24 P_A / 1.066$	\$/g U-235
	A'	$0.33 P_A / 1.066$	
	B	$0.30 P_B / 1.066$	
	B'	$0.37 P_B / 1.066$	

Now P_B is likely to be about \$60/kg U and P_A will be higher by at least \$2/kg U and more likely by \$7/kg U, due to the higher cost of fabricating enriched fuel. Setting $P_B = 60$, $P_A = 67$, the values become

for	A	\$15.1/g U-235
	A'	\$20.7/g U-235
	B	\$16.9/g U-235
	B'	\$20.8/g U-235

These values apply to the added enrichment only when in the fuel. Any extra fabrication cost ΔF \$/g U-235 must be deducted to obtain the acquisition value of the extra U-235.

Moreover, the above values apply to fissile material added as an improvement to once-through fuel that is not reprocessed. If, on the other hand, the plutonium is extracted, and before extraction is valued at $\$V_r/\text{g Pu}$, the increase in residual plutonium of about 0.15 to 0.2 g/kg U in the cases considered, adds 0.15 to 0.2 $V_r/1.066$ \$/g U-235. Because of the low concentration of Pu, about 3 to 3.5 g Pu/kg U, the value before extraction, V_r , is likely to be low and no more than \$2 to \$4/g Pu, so the extra plutonium yield does not add much to the value of the enrichment.

APPENDIX 3

REACTOR FUEL OF LONG LIFE

Reproduced below are figures illustrating the long irradiation of fuels containing U-238 and Th-232 as fertile materials. Figure 3.1 is Figure 2 from DL-25. The hatched areas indicate the excess of neutron yield over neutron absorption for natural uranium and for a mixture of 1 atom U-235 with about 70 atoms of thorium. The calculations involved poorly known cross-sections, etc., and are less reliable than those of Figure 3.2 which is Figure 6 from DM-47. Figures 3.3 and 3.4 are Figures 4 and 5 from DL-42. It is possible from Figure 3.4 to follow the energy yield with irradiation. If the U-235 curve is added to that for 138 atoms of U-238 the rate of fissioning, as is well known, remains very constant with irradiation. The thorium curve is drawn for 50 atoms of thorium because that has much the same neutron absorption as 138 atoms of U-238 in the neutron spectrum envisaged. The energy yield is higher at first and then falls off much as occurs with enriched uranium.

FIGURE 3.1

FIGURE 3.2

FIGURE 3.3

FIGURE 3.4

There is a neutron flux limit for thorium set by the capture cross-section and radioactive half-life of Pa-233. In the neutron spectrum envisaged the flux should be limited to 3×10^{13} n/cm²/sec or 1 n/kb per year. To reach 10 n/kb the thorium would therefore remain in the reactor for 10 years.

DESCRIPTION OF THE BELGONUCLEAIRE - C. E. N.
PLUTONIUM PROJECT*

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I. HISTORICAL BACKGROUND, GENERAL ORGANIZATION
OF THE PROJECT AND DESCRIPTION
OF THE BELGONUCLEAIRE - C. E. N. PLUTONIUM FACILITIES

I. 1 HISTORICAL BACKGROUND

In 1956, BelgoNucléaire established a tentative nuclear program for Belgium, recommending that our country concentrates on fast reactors and on the plutonium cycle.

As a consequence of that suggestion, BelgoNucléaire sought from its mother company, Union Minière du Haut-Katanga, a credit of \$100,000 which enabled our group to start studies on plutonium, as far back as 1957.

BelgoNucléaire also asked for the collaboration of C. E. N. at Mol to co-operate in its plutonium research.

Later on, when Euratom issued an invitation to submit proposals for research contract, BelgoNucléaire decided to present a tentative program on plutonium and offered the C. E. N. to be its associate in that project, which was accepted.

As a result of this, Euratom granted the BelgoNucléaire - C. E. N. Association an important contract on plutonium technology within the scope of the Euratom - U. S. A. Joint Program.

The work carried out under that contract is the topic of the present paper.

I. 2 GENERAL ORGANIZATION

The technical team in charge of the Plutonium Project is broken up into 8 groups as indicated in Table I. This table also shows the number of people actively engaged in this program.

*See page 22. 36 for contributors.

TABLE I
TOTAL MANPOWER IN CHARGE OF THE BELGONUCLÉAIRE -
C. E. N. PLUTONIUM PROJECT

	<u>Staff</u>		<u>Technicians</u>		<u>Clerks</u>
	<u>Scientific</u>	<u>Technical</u>	<u>Technicians</u>	<u>Workmen</u>	
Management and Administrative Support	1.5				10
Ceramic Preparation Group	4	2	11	--	
Ceramic Fuel Fabrication Group	2	4	10	--	
Nonceramic Fuel Fabrication Group	1	--	2	--	
Nondestructive Testing and Irradiation Group	5	3	4	--	
Properties of Materials Group	1.5	1	2	--	
Chemistry Group	2.5	3	6	--	
Miscellaneous Technical Services	1	3	9	8	
Pu Laboratory Installation and Maintenance Group	2.5	1	7	7	
Total	21	17	51	15	10

I. 3 DESCRIPTION OF THE PLUTONIUM LABORATORIES

Figure 1 shows the general lay-out of the plutonium facilities located in the hot wing of the C. E. N. chemistry building.

The four laboratories on the left side of the wing are completely installed and in operation. The laboratory was first operated in early 1960. The last two laboratories on the right side of the wing are still under construction.

22.3

FIGURE 1

General Layout of the Belgonucleaire -C. E. N. Plutonium Facilities at Mol with the Disposition of the Glove Boxes

HW-75007

Figure 1 also shows the location of the various glove boxes in the laboratories.

Figures 2 and 3 show the two lines of glove boxes of the ceramic preparation laboratory and Figure 4 represents the fuel fabrication laboratory.

Figure 5 is a view of the properties of materials laboratory and Figure 6 shows part of the chemistry laboratory. The argon purification system located on a gallery in the fuel fabrication laboratory is shown on Figure 7. It is presently loaded with a 50/50 Ti-Zr getter.

FIGURE 2

Ceramic Preparation Laboratory
Glove Box Line No. 1 and 2

FIGURE 3

Ceramic Preparation Laboratory. Glove Box Line No. 2 and 3

FIGURE 4

Fuel Preparation Laboratory

FIGURE 5
Properties of Material Laboratory

FIGURE 6
Part of the Chemical Laboratory

FIGURE 7

Argon Purification System
in the Fuel Fabrication Laboratory

The facility also includes an X-ray room and a laboratory used for nondestructive testing (Figure 8).

FIGURE 8

Views of the Nondestructive Testing Laboratory

I. 3.1 Ceramic Preparation Laboratory

This laboratory contains 4 lines:

- The powder processing line (box 1 to 6). This line permits the calcination of the uranium-plutonium salts, grinding and mixing of the powders, sieving, particle size determination, determination of the oxygen or carbon content of the powders by oxygenation and reduction and low temperature sintering (up to 1500 C).
- The pellet fabrication and small metallographic line (box 7 to 11). This line includes a 50 ton hydraulic press for cold pressing, a high temperature (1900 C) sintering furnace designed for use with oxidizing as well as reducing atmospheres, an analytical balance and all the metallographic equipment for cutting, grinding, polishing, etching, and examining the metallographic samples. A Nachet metallographic microscope is located outside the glove-box and the specimens are viewed through a glass (maximum magnification: 600 X).
- The powder characterization line (box 12 to 14). This line contains the instrumentation for density measurements (water, mercury and helium), particle size determination (Fisher Sub-Sieve-Sizer) and specific surface measurements (modified BET method).
- The physical properties line (box 15 and 16). This line includes an apparatus for thermal expansion experiments, thermal simulation experiments and grain growth determination. These two last instruments permit simulation of the behavior of packed powder at temperatures up to 2800 C.

I. 3.2 Two Chemical Laboratories

Our chemical laboratories serve for:

- the recovery and purification of plutonium from processed specimens and wastes,
- the preparation of plutonium salts and compounds prior to calcination,
- the routine chemical analysis.

The first laboratory contains ten boxes equipped for dissolution, concentration of plutonium solutions, precipitations from aqueous solutions, purification of aqueous Pu solutions by ion exchange method, analysis of plutonium and uranium solutions (pH meter, centrifuges, electric furnaces, microbalance, etc...).

Beside the same apparatus as above, but to a larger scale, the new laboratory contains a thermo-balance, an apparatus for differential thermal analysis, a furnace to analyse the oxygen content, a special box for various carbide analyses, a box for analyses on fluorides and a spectrometer.

I. 3.3 Two Fuel Fabrication Laboratories

The first laboratory is devoted to the first step of the fabrication. It consists of three units:

- The vibratory compaction line (box 1 to 3). This line consists of three interconnected boxes:
 - the powder box, equipped for crushing, grinding, sieving, mixing, weighing and tap density measurements of the powders,
 - the vibration box with a 2 kW electro-magnetic vibrator located underneath the box,
 - the arc welding box containing a helium leak detection group, a welding torch and an arc furnace: tubes up to 17 mm diameter and 500 mm length can be welded in this box with either DC or AC power.
- The work shop box (box 4). This box is used as a repair shop for plutonium contaminated equipment and as a fabrication step for machining and polishing the fuel elements. It is equipped with a lathe, a drilling machine, a dual-purpose cutting machine and small hand-tools.
- The swaging line (box 5 and 10). This line contains equipment for blending oxygen--and nitrogen--sensitive powders (for instance zirconium) with the plutonium bearing powder (PuO_2 , PuC , etc...) an annealing furnace, a pre-heating furnace, a bubble test device and a Farmer-Norton swaging machine (up to 27 mm diameter).

The new laboratory will contain additional work shop equipment, a straightener, a welding machine, a decontamination line and additional all purpose equipment.

I. 3.4 Properties of Materials Laboratory

This laboratory is primarily devoted to carbide preparation and physical metallurgy tests. It contains 2 lines and 2 individual boxes:

- The carbide preparation line (box 11 to 13). This line contains a standard 2 ton hydraulic pelletizing press, a blender, a balance and a sintering furnace reaching 1700 C.
- The metallography line (box 7, 8, 9, and 14). It is a full size metallographic line including automatic grinding and polishing equipments, etching cells, a large Reichert metallographic microscope (fully equipped for reflection and transparency microscopy) and a microdur-ometer.
- The electron Beam Welder (box 15). This machine can handle specimens up to 30 mm diameter and 170 cm long.
- The diffusion box (box 6). This box contains a high-temperature bell-type furnace (up to 2000 C), a quenching furnace (1000 C), three low-temperature furnaces (1000 C), and two dual-temperature treatment treatment furnaces (up to 1500 C).

I. 3. 5 X-Ray Laboratory

This laboratory especially designed for X-ray work on plutonium contaminated specimens contains a fully equipped Siemens X-ray device, including diffractometry and fluorescent analysis.

I. 3. 6 Nondestructive Testing Laboratory

This laboratory contains equipment for testing the tubes and the elements including high sensitivity equipment for helium-leak testing (10^{-9} cm³ atm/sec), a gamma density apparatus, an X-ray machine for radiography, a dye penetrant room, eddy-current testing equipment, an ultrasonic thickness gauge and dimensional tolerance checking equipment.

II. DESCRIPTION OF THE PROGRAM

The present program includes the study of three types of fuels:

- mixed uranium and plutonium oxides,
- mixed uranium and plutonium carbides,
- dispersion of plutonium oxides and carbides in metallic matrices.

II. 1 MIXED URANIUM AND PLUTONIUM OXIDES

II. 1. 1 Fabrication of Oxide Fuel Elements

a) General Consideration

The process used for the fabrication and densification of the fuel elements is based on vibratory compaction and swaging.

The fabrication cost of enriched fuels varies less with enrichment in the case of plutonium fuels than in the case of U-235 enriched fuels, principally because the same facilities and safety rules have to be used whatever the Pu enrichment may be. This is in contrast with the fabrication cost of uranium-235 enriched fuels which is lower at low enrichments, but increases significantly with enrichment. Consequently, the difference in cost between plutonium enriched and uranium enriched fuels decreases with increasing enrichment.

Furthermore, the charge for fissile elements contained in a fuel does not show the same dependency upon enrichment for plutonium enriched and U-235 enriched material. Figure 9 represents the charge for U-235 versus enrichment and the combined charge for the fissile material (Pu and the 0.7% of U-235 contained in natural uranium) assuming a plutonium cost of either 10 or 12 \$/gram. It can be seen that the cost of U-235 increases faster than the cost of fissile Pu + 0.7% U-235.

FIGURE 9

Variation of the Charges of U-235 and Pu + 0.7% U-235
in Function of Enrichment

On the basis of these two simple considerations it could be anticipated that plutonium is economically more attractive in relatively highly enriched reactors.

Under the assumption of the validity of the above conclusion, it becomes obvious that those fabrication methods have to be developed that are best suited to fuels of relatively high enrichment. As such fuels are mostly in the form of small diameter rods (less than 10 mm or 0.4 inch) the emphasis has to be put on manufacturing techniques adapted to those sizes.

A comparison of the diameter dependence of the fabrication cost of fuels rods made by pelletizing, vibratory compaction and swaging, is shown on Figure 10 for one meter long stainless steel tubes filled with three percent

plutonium enriched fuels. The densities are:

90% TD for vibratory compacted fuel,

94% TD for swaged fuel,

94% TD for pelletized fuel.

FIGURE 10

Variation of Fuel Fabrication Cost with the Decrease in Diameter of Fuels Fabricated by Pelleting, Swaging, Vibratory Compaction

These curves show that, for this type of element, vibratory compaction is less expensive than pelletizing for diameters below 10 or 11 mm. Swaging is less expensive than pelletizing when the diameter decreases below about 8 or 9 mm.

The very sharp cost increase of the pelletizing technique for small diameter rods is mostly due to the large number of pellets to be fabricated, weighed and inspected. This number varies as the inverse of the cube of the diameter.

Contrary to this, the fabrication cost by vibratory compaction is less diameter dependent because with a proper powder feed system to the tubes, a given weight of fuel can be prepared in one batch regardless of the tube size. However, difficulties are encountered when high densities have to be achieved in rods of internal diameters of less than 7 mm (0.3 inch). These arise from the difficulty of classifying subsieve powders and handling these very fine powders in glove boxes without external contamination.

Swaging is made expensive by the numerous nondestructive tests needed. However, in the case of small diameter fuel pins, this method remains less expensive than pelletizing.

Hence, it appears from such economic considerations, why vibratory compaction and swaging seem better suited to plutonium fuels than pelletizing.

b) Uranium-Plutonium Oxide Fabrication

The fabrication flow sheet that has been developed is shown in Figure 11. It is based on physical mixing of the fissile and the fertile materials. This mixing is normally performed during the vibration process by means of adequate feed equipment.

Vibratory Compaction¹- For plutonium concentrations exceeding five percent, the plutonium oxide is separately added during filling as the fine fraction of the mixture. For concentrations below five percent, it is first prepared in the form of a solid solution with the uranium oxide by pelletizing, sintering and grinding adequate amounts of PuO_2 and ceramic grade UO_2 .² An alternative consists in co-precipitating the plutonium and the uranium from a nitrate solution. This method was found to be more expensive because of the higher amount of material to be handled from the beginning of the preparation line and also because of the criticality restrictions for handling the plutonium salts in the presence of water.

It is well known that the vibratory processing characteristics required to reach a given density vary with the size of the material with which the tube is to be filled. To avoid costly handling of plutonium, the development work for each kind of fuel is performed on uranium oxide. The optimum granulometry and the required frequency are then determined. The vibration characteristics are recorded on tape which is then directly fed to the vibrator performing the densification of the uranium and plutonium mixture in the glove box. This procedure allows time saving and increases reproducibility.

The apparatus for cold development work includes a glove box mock-up and is shown on Figure 12. Figure 13 represents the equipment for work on plutonium.

FIGURE 12

Vibratory Compaction Equipment for Cold Work

FIGURE 13

Vibratory Compaction Equipment for Plutonium Work

Density of 88 to 92% of the theoretical density are actually obtained depending principally on the tube characteristics. Variation in density along the tube is less than 0.5% TD as shown by the gamma scanning diagram of Figure 14. The reproducibility of the mean density is better than $\pm 0.5\%$ TD.

FIGURE 14

Gamma Densimetry of a Vibratory Compacted Oxide

Tube: Diameter: 10 millimeters

Explored length: 20 cm

The bottom of the tube corresponds to the left of the curve.

Cold Swaging Compaction³- Cold swaging is used when higher densities (94% TD) are required or when the tube has to be of small diameter (less than 7 mm or 0.3 inch).

The powder is precompacted by vibration up to 85% TD. A density of 94% TD is then reached by means of a few swaging passes.

The swager used is located in a glove box as shown on Figure 15.

FIGURE 15

Swaging Machine Installed in a Glove Box for Plutonium Work

The density uniformity is not as good as for vibration compaction and is of the order of 1.2% TD. The tube thickness variation after swaging is less than 3%. The reproducibility of the mean density is usually within 1.5% TD for both stainless steel and Zircaloy tubes, even when the diameter thickness ratio is as high as 50. Figure 16 shows a section of a swaged rod having a wall thickness of 0.2 mm (0.008 inch) without any imperfection.

171

11 X

Initial D/e = 10
Final density: 10.3 gm/cm³
Reduction: 60%

164

11 X

Initial D/e = 50
Final density: 10.2 gm/cm³
Reduction: 55%

FIGURE 16

Cross Sections of Cold Swaged Rods
(stainless steel and fused UO₂)

Hot Swaging. Hot swaging leads to densities as high as 97% TD. Nevertheless the stainless steel tubing quality after hot swaging is inferior to that obtained by cold swaging partially because of the reaction of stainless steel with oxide, even if the latter material is stoichiometric (Figure 17).

700x

166

11 X

Stainless Steel

Initial D/e = 10/1

Fused UO_2 Density: 10.7 gm/cm^2

Reduction: 60%

Hot (700 C) Swaged SpecimensFIGURE 17

Cross Sections of Hot Swaged Rod

II.1.2 Miscellaneous Studies on Plutonium Oxides and Mixed Uranium and Plutonium Oxides

a) Thermal Expansion Studies of PuO_2 and $\text{UO}_2\text{-PuO}_2$ ⁴

The thermal expansion behavior of stoichiometric and non-stoichiometric plutonium oxides and mixed uranium and plutonium oxides has been studied between room temperature and 1000 C. Figures 18 and 19 show the temperature dependence of the thermal expansion of PuO_2 , $\text{PuO}_{1.95}$, $\text{PuO}_{1.81}$, $\text{PuO}_{1.74}$, $\text{PuO}_{1.71}$ and $\text{UO}_2 - 4 \text{ m/o PuO}_2$, $\text{UO}_2 - 6 \text{ m/o PuO}_2$, $\text{UO}_2 - 10.6 \text{ m/o PuO}_2$, $\text{UO}_2 - 11.1 \text{ m/o PuO}_2$, $\text{UO}_2 - 19.1 \text{ m/o PuO}_2$, $\text{UO}_2 - 24 \text{ m/o PuO}_2$.

FIGURE 18

Dilatometric Curves of $\text{PuO}_{2.00}$ and PuO_{2-x}

FIGURE 19
Dilatometric Curves of UO₂-PuO₂

From the results obtained it can be concluded that:

- the mean thermal expansion coefficient of sub-stoichiometric PuO₂ depends essentially on the O/Pu ratio and the dilatation curves present discontinuities corresponding to phase transformation at 300 C and 650 C.

- for the same sintering conditions the mixed oxides with a composition of PuO_2 in the neighbourhood of 10 m/o have a mean thermal expansion coefficient lower than those of the other mixed oxides.

b) Effective Thermal Conductivity of Vibrated UO_2 Fuel Elements

Thermal experiments were performed aiming at properly interpreting the temperature distribution inside irradiated fuel samples and at determining the effective conductivity of the oxides. It is hoped that future work in this area will help the reactor designer to better predict the limitation of the fuel under actual reactor operating conditions.

Work has been up to now limited to UO_2 but will soon be extended to PuO_2 - UO_2 mixtures.

To date the thermal behavior of pelletized and vibrated fuel elements has been simulated (Figures 20 and 21). Results of effective thermal conductivity measurements during out of pile experiments are shown on Figure 22. From these curves it can be concluded that the UO_2 thermal conductivity decreases up to 1200-1400 C and then increases up to the melting point. This increase is due to radiant energy transmission. These results confirm and complete those previously obtained by J. L. Bates⁵ and by B. E. Murtha and W. P. Chernock.⁶

At present simulation work is being conducted on vibrated UO_2 + fission fuel elements to determine the effect of the fission product on the thermal behavior of the ceramic fuels.

II. 2. MIXED URANIUM AND PLUTONIUM CARBIDES

Two ways of preparing the plutonium carbides can be adopted. One is based on the carburization of the plutonium metal and the other on the carburization of the oxide. We have developed techniques based on the latter method which seems more economical. After processing the carbides are densified by vibration or swaging.

The flow sheet followed is shown on Figure 23. Pure plutonium carbide is sintered, ground and physically mixed with fused and ground uranium carbide during vibration. If the required PuC concentration is less than 5%, the PuC and UC powders are mixed, pelletized, sintered and ground in order to increase the amount of the plutonium fraction. The sintering of PuC-UC compounds can be obtained without any change in stoichiometry of the carbide. However, a very pure argon atmosphere is essential.

II. 3. PLUTONIUM OXIDE AND CARBIDE CERMET FUELS

These fuels are dispersions of spherical PuO_2 or PuC grains in a metallic matrix such as zirconium-niobium base alloys. They are of potential interest for reactors having a high specific power output. The PuO_2 grains are spheroidized on a vibratory screen or in a vibratory cylinder. Perfect spherical UO_2 and PuO_2 grains hence obtained are shown on Figures 24 and 25. After sintering the PuO_2 grains reach a density of 95% TD. Studies have been started on the production of spherical PuC grains by the same methods.

(b) 110 X

(c) 110 X

(d) 110 X

FIGURE 20

Corss Section Before Polishing of a Pellet After Thermal Simulation
and Metallography of Different Regions of the Heated Fuel

FIGURE 21

Cross Section After Polishing of Vibratory Compacted
Uranium Oxide Powder After Thermal Simulation

FIGURE 22

Effective Thermal Conductivity of an UO_2 Fuel Element

FIGURE 23

Flow-Sheet for the Fabrication
of Uranium and Plutonium Carbide Fuels

FIGURE 24

150 μ UO_2 Spherical Grains Prepared by Vibratory Sieving

a) Sintered PuO_2 Spherical Grains 200 μ

b) Green Spherical PuO_2 Grains 400 μ

FIGURE 25

Because of the potential reaction between PuO_2 and zirconium, experiments have been made to protect PuO_2 grains by means of inert metallic coatings.⁷ Figures 26, 27, 28 and 29 show spherical UO_2 grains respectively with Nb, Ni-Cu, Mo, Cr.

FIGURE 26

UO_2 Spherical Grains Protected with a $10\ \mu$ Nb Layer Obtained by Thermal Decomposition of NbCl_3 in a Fluidized Bed

FIGURE 27

Double Layer Coating Obtained by Kanigene Deposition of Ni and Electrolytic Deposition of Cu

FIGURE 28

Molybdenum Layer ($4\ \mu$) Obtained
by Vacuum Metallization

FIGURE 29

Chromium Deposition of $12\ \mu$ Obtained
by Vacuum Metallization

In connection with this work an important effort was made to produce a new Zr-Nb alloy showing good mechanical properties and better nuclear characteristics than stainless steel. The optimum niobium concentration in zirconium was found to be 17 w/o.⁸ However, this alloy presents a transformation point at about 600 C which somewhat limits its use as a dispersed type fuel. Irradiation experiments will start soon to determine the behavior of such matrices and dispersions under normal operating conditions.

In order to raise the transformation point, ternary alloying elements such as vanadium and chromium have been added. The amount of niobium is to be simultaneously lowered. The best results have been obtained with alloys having the following compositions:

Zr - 2 Nb - 2 V

Zr - 3 Nb - 3 V.

II. 4. FUTURE IRRADIATION PROGRAM

Our irradiation program is scheduled to start in early 1963. Samples of UO_2 - 3% PuO_2 have been prepared by vibratory compaction and swaging for preliminary irradiation tests.

II. 4. 1. Irradiation Tests in the BR2 High Flux Reactor

a) Oxide Fuels

Short irradiation (1 to 20 minutes) will be made in an hydraulic rabbit on mixed uranium-plutonium oxides to study the optimum heat removal conditions of those fuels. The plutonium enrichment will vary from 1 to 7 w/o and the heat output from 200 to 1000 W/cm.

Long irradiation experiments will be performed to determine the stability of plutonium oxide fuels at burnups up to 50,000 MWd/T and at a power output of 200 to 500 W/cm.

These tests will start on January, 1963.

b) Carbide Fuels and Cermet Type Fuels

These fuels will be tested for long burnups to define their stability under severe irradiation conditions.

II. 4. 2. Irradiation in the BR3 Power Reactor

Nine UO_2 - PuO_2 fuel rods, prepared by vibratory compaction, swaging and pelletizing will be introduced in a subassembly of the BR3 reactor in April 1963. The purpose of this experiment is to analyze the behavior of these elements in a PWR type reactor, under actual power operating conditions.

III. CONCLUSION

During the last two years we have developed various fabrication methods for the most usual plutonium fuels. We are now at the point where we are able to start fuel element fabrication for irradiation testings.

The irradiation we plan to do will give us more details on the use of plutonium as a fuel for thermal reactors, taking into account the benefit of the presence of the higher isotopes at one hand and the optimum heat output that such kind of fuels can tolerate.

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PLUTONIUM RECYCLE VALUES IN THERMAL AND FAST REACTORS AND EXPECTED PATTERNS OF USE OF PLUTONIUM

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I. THERMAL REACTOR RECYCLE

In using plutonium in a thermal power reactor we may take advantage of it mainly in two ways:

1. To reduce the amount of uranium-235 makeup required to attain any specified fuel lifetime; or
2. To increase the reactivity lifetime available for a given control strength.

Both of these characteristics are used in all low-enrichment reactors in which uranium-238 is the fertile material. This might be called "recycle in situ" since part of the newly-generated fissile material is burned in place. For example, after the initial 40 percent is discharged, the remainder of the first core of Dresden will derive about 40 percent of its power from the fission of self-generated plutonium.

Spent fuel from an enriched uranium reactor must be chemically processed. For enrichments above about 0.8 percent, uranium-235 value of the depleted uranium is greater than the expected cost of processing.* Plutonium which becomes available as a byproduct of this operation, may be recycled to decrease the amount of enriched uranium makeup required.

When recycle is used it is found that the amount of uranium-235 makeup required to attain any given reactivity lifetime is considerably smaller than for the once-through cycle of equal lifetime. The saving in uranium-235 makeup required for a given reactivity lifetime increases with the number of times that plutonium has been recycled in a given reactor. For survey purposes, it is convenient to study the steady-state of "equilibrium cycle" which is closely approached after about three recycles.

A. Method of Calculation of Plutonium Value for Thermal Recycle

A method of estimating the value of plutonium in a thermal reactor using plutonium recycle was used in a Geneva paper.¹ * This consists of equating the total cost of the fuel cycle with recycle to the cost of operation of the same reactor on a once-through cycle.

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1. Greebler, P., et al. "The Recycle of Plutonium in Low-Enrichment Light-Water Reactors", Second Geneva Conference Proceedings, 13: 251-264. 1958.

*The estimated chemical processing cost post-1970 is \$10/kg (see Section I-C for basis).

In the once-through reactor an increasing price of plutonium decreases the cost of the fuel cycle by increasing the plutonium credit. For the recycle reactor increasing plutonium value increases fuel cycle cost, since after the first two or three cycles, plutonium appears only as an inventory charge. This value of plutonium, which is defined by the intersection of the once-through and the recycle costs lines (plotted against plutonium price) has been called by Eschbach, et al., the "indifference value". At this price there is no cost difference to the reactor operator, whichever cycle he uses.

These cost relationships can be expressed for any given fuel lifetime by the following expressions:

$$C = \Delta_{25} + \bar{I}_{25} + \bar{I}_{Pu} + F + R - P \quad (1)$$

$$C' = \Delta'_{25} + \bar{I}'_{25} + \bar{I}'_{Pu} + F' + R' - P' \quad (2)$$

let

$$D = (\Delta_{25} + \bar{I}_{25} + \bar{I}_{Pu})$$

$$D' = (\Delta'_{25} + \bar{I}'_{25} + \bar{I}'_{Pu})$$

and

$$C = C' \text{ and } R = R' \text{ at equal fuel lifetimes.}$$

then

$$D - D' = \overline{\Delta FAB} + P \quad (3)$$

where

C = fuel cycle cost in dollars per kg of fuel throughput.

Δ_{25} = difference between the initial and discharge values of enriched uranium per kg of initial fuel.

$\bar{I}_{25}$ = average uranium inventory cost per kg of fuel throughput.

$\bar{I}_{49}$ = average plutonium inventory cost per kg of fuel throughput.

P = plutonium credit, per kg of discharge fuel mixture.

$F+R$ = fabrication, reprocessing and shipping costs.

$\overline{\Delta FAB}$ = additional fabrication costs per kg of recycle fuel, averaged over the core = $(F' - F)$.

D = sum of inventory and depletion costs, per kg of fuel throughput.

Unprimed symbols refer to minimum cost once-through cycle.

Primed symbols refer to the minimum cost recycle.

The quantity which can be most directly calculated for various recycle schemes is the difference $(D-D')$, the depletion plus inventory costs of once-through and recycle operation respectively for equal fuel lifetimes. This difference may be equated to the sum available (for a given mode of operation) to cover the plutonium credit and the average fabrication cost differential, $\Delta \overline{FAB} + P$.

It is manifestly not feasible to obtain precision in the calculation of plutonium value while there are still substantial uncertainties both in the values of the cross sections used in the calculations of $(D-D')$, and in the attainable minimum values of $\Delta \overline{FAB}$. Experimental confirmation is also required of the detailed validity of parts of the calculational models for deriving neutron spectra and effective cross sections as a function of reactor design and fuel composition.

Because of these uncertainties, the optimum area of design for both once-through and recycle operation of the reactor as yet cannot be defined with the accuracy desirable for commercial use. Survey calculations with conservative selections of cross-sections can at present serve mainly to estimate lower limits of the probable value of plutonium and to direct attention of further development to the areas of design in which maximum worths are likely to be found.

There is considerable flexibility available to the reactor designer in controlling the effective isotopic cross-sections through adjustment of the spectrum and of resonance self-shielding. Some of the presently used cross-sections are likely to be significantly changed by measurements which are now nearly complete.* Because of flexibility available in design, better values of cross-sections will tend to shift the optimum design points rather than to change the minimum plutonium values.

B. Fueling Patterns for Use with Thermal Reactor Recycle

Plutonium may be deployed in a thermal reactor in several different ways --each having each own advantages and disadvantages with respect to reactor design. For example, appreciable differences in plutonium value may be found in different combinations of the following:

- I. Homogeneous mixture of plutonium and uranium (such as UO_2 - PuO_2 mixtures).
- II. "Spike" loadings in which plutonium and uranium are initially segregated.
- III. Medium-lifetime cycles with multi-batch (or "graded" or "zone") loading patterns.

The main effect here is the reduction of uranium makeup and inventory costs by the addition of recycled plutonium. Similar lifetimes are attainable by once-through cycles.

*These include isotopic analyses of a Dresden fuel rod at about 5,000 MWD/T, and a VBWR fuel rod (with plutonium fuel initially containing 17 percent plutonium-240) which has been irradiated to about 2,000 MWD/T. Both are currently being analyzed (September, 1962).

- IV. Extended fuel lifetimes, and/or batch loading in which the main effect of plutonium is to permit longer reactivity lifetimes than might be attainable with uranium makeup alone in batch loading.

The work reported in this paper has selected deployment of plutonium primarily in the pattern described by (I) and (III) above. The considerations favoring the selected pattern for near-term value evaluations are as follows:

1. Uniform or near-uniform disposition of fissionable material in the reactor permits relatively constant power distributions through a refueling cycle. Highly non-uniform distributions such as "spike" or "seed-and-blanket" generally result in lower average power densities for a given reactor design because of greater variation in power distribution with burnup. While the difference may not be prohibitively costly, it adds the complexity of requiring analysis of differentials in (a) capital costs, (b) operating costs, and (c) control system design, to complete the fuel cycle evaluation.*
2. Medium-lifetimes (15,000 to 25,000 MWD/T) are selected in preference to very long fuel lifetimes (25,000 to 50,000 MWD/T) because the cost incentive for extremely long lifetimes disappears as fuel fabrication costs reach the neighborhood of \$50/kg or below (for mixed uranium-plutonium fuel).**
3. The long-lifetime, high-plutonium-concentration cycles are more sensitive than the shorter cycles to the present uncertainties in both the isotopic cross-sections, and in the methods of calculation of neutron spectra and effective cross sections.
4. The long-lived fuels have characteristically long time cycles for testing and measurement of both materials and neutron properties. (For example, a 50,000 MWD/T fuel requires a testing cycle extending from 6 to 12 years after a plutonium-fueled design is committed, depending on whether the average specific power is 20 or 40 kw/kg of fuel mixture.) In such periods of time, the growth of volume of fuel fabrication is expected to permit low fabrication costs, which produce optimum fuel-cycle costs using cycles of shorter lifetimes.

The alternative patterns of fuel deployment (II) and (IV) are not excluded from further exploration. However, the considerations given above suggest that the medium-lifetime homogeneous pattern of plutonium use may for some time give a more reliable basis for evaluation of minimum value of plutonium.

* An obvious direction of economic improvement of spike designs is to make the difference between "spike" and blanket characteristics as small as possible, thus approaching the multi-batch or graded loadings used for this analysis.

** "Spike" fuels have an apparent advantage since only a part of the fuel carries the fabrication cost penalty due to plutonium content. However, this may be largely offset in practice by the higher unit cost of fabrication of small quantities of "spiked" fuel.

Another use for plutonium recycle is the use of plutonium with natural uranium only to permit self-sustaining recycle. While such operation may be of strategic value in an isolated economy, previous work suggests that it is not of primary economic interest for reactors requiring appreciable enrichment.¹

Finally, plutonium from several thermal reactors may be used to supply the entire enriched fuel makeup of a special purpose power reactor. This approach shares many of the problems of the spike loadings discussed in (3) and (4) above. In addition, a reactor of significant size --say a 200 to 400 MWe BWR or PWR--would require one to two tons of plutonium inventory to operate advantageously. The limited availability of plutonium during the period considered as "near-term" makes this type of use impractical. For long-term use (beyond 1970 to 1973), such special-purpose thermal reactors appear less promising than fast reactors as plutonium users.

C. Throw-Away Cycles and Chemical Processing Costs

Fuel cycles can be designed which may have tolerable fuel costs without any chemical processing of fuel (D_2O -natural uranium system) or by reprocessing only part of the fuel (the "seed" of seed-and-blanket). These designs cannot be evaluated in terms of fuel costs alone, since significant added capital or non-fuel operating costs must also be considered. This limitation is also true of the fast reactor systems, which even though they promise to have very low or even negative fuel costs, nevertheless are not near-term contenders for economic power generation.

The assumption of continued high costs for chemical processing of reactor fuels appears valid only for small and isolated, or slowly-growing economies. For rapidly-growing national or regional economies, reductions in unit costs of processing are assured by the operation of the same scaling laws which improve the economics of large power units.

At least four countries have large-scale plants for solvent extraction processing of irradiated fuels for plutonium production. These plants total of the order of 50 plant-years of routine operating experience, and have produced tens of tons of plutonium.

The unit processes and equipment components are largely identical with those required for most power reactor fuels (differing mainly in the chemical flowsheet of the feed-preparation steps for different cladding materials).

At scale of operation of the order of a thousand tons per year and upwards, costs are experienced which are a small fraction of the costs of a one-ton-per-day minimum-scale plant. The economic problem concerns mainly the next three to five years, during which power reactor fuel volume is too small to support an economic size of processing operation. An estimate of the expected costs of processing in the late 1960's can be derived from the one-ton-per-day plant, and the projected fuel volume available.

1. Reference 1, page 23.1.

From the installed capacity curve derived in a later section (Figure 9), a fuel processing volume of 900 to 1000 tons/yr is expected by about 1970.

Processing costs have been estimated recently for a small privately-owned processing plant to be about \$24/kg, at an annual rate of 200 tons/yr.² Additional small-scale plants are scheduled for completion in the next few years in Belgium, India, Italy, and Japan, as well as in the United States.

Assuming only scaling-law effect (no improvement in technology), a unit cost of $(\frac{200}{900})^{.6} \times \$24.00 = \$10/\text{kg}$ is estimated for a processing rate of 900 tons/yr.

A reactor whose construction starts in the next two or three years can expect processing costs near \$10/kg toward the end of its first complete core lifetime, and still lower costs over most of its operating life.

Specialized reactor or cycle designs which compromise economic performance on the assumption that low-cost fuel reprocessing will long be unavailable appear to have only transitory worth in a rapidly growing economy.

D. Methods of Calculation

For survey purposes, the THUR burnup code was used. This is a zero dimensional burnup code (equivalent bare core model) programmed for the TRANSAC 2000. It permits direct calculation of steady-state recycle fuel isotopic compositions and reactivities and also permits calculation of once-through first cycle compositions, and stepwise calculation of recycle fuel compositions and reactivities. The code permits searching on uranium enrichments for any stage (or on the concentration of additional plutonium makeup or of burnable poison) to attain a specified fuel irradiation lifetime. Reactivity effects of non-uniform fuel burnup are taken into account in accordance with a specified fuel loading scheme. An out-in multi-batch loading pattern with five-batch loading was specified for most of the survey. Reactivities and neutron balance data are computed by a subroutine code MUGO, a multi-group diffusion theory code. (Three-group and twenty-group cross sections were used for the survey.) MUGO also computes the equivalent one-group cross sections for the calculated spectrum of each composition and these are used for the burnup calculations. A more detailed description of the THUR code and of the neutron balances used is given in Appendix I.*

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2. Runion, T. C. "NFS Chemical Procession of Plant Capability and Cost Bases", Nuclear Congress Paper, June 5, 1962. (The limiting unit rate is used excluding turnaround, since turnaround time becomes minor in large scale operations.)

* THUR is a subroutine of a more general design optimization code (HIPOP) which permits simultaneous calculation of economic optima including an interaction of thermal hydraulic, nuclear, and cost aspects of reactor design. Either the thorium or the uranium cycles may be calculated. Since the economic inputs characteristic of plutonium were themselves the objects of this investigation, the design optimization features were not used in the work summarized here.

In addition to the survey of the first cycles and equilibrium cycles, a few selected cases were followed through the first six to eight recycles to determine the rate of convergence in compositions and in economics.

An independent check on the rate of convergence of compositions and fuel cycle economics was obtained by calculating a few selected cases using the GEFU code.³ The GEFU code calculates reactivities and compositions for ten variable time steps per fuel cycle. Effective cross sections are redetermined for each time step composition, using a more precise formula for resonance self-shielding of plutonium-240 than that in THUR, and including resonance self-shielding of plutonium-242 and spatial self-shielding of thermal cross sections. The rate of convergence of compositions to the steady-state was found to be very similar to the THUR cases, as were the economic relations of the first few cycles to the equilibrium cycle.

When compared with the reactivity lifetime information from the first cores of Dresden and Yankee, both codes significantly underpredict the reactivity lifetimes observed (or overpredict the rate of loss of reactivity). This indicates a somewhat conservative (pessimistic) selection of some combination of fission product cross sections, alphas of the fissile isotopes, reactivity penalties for non-uniform void distribution and non-uniform plutonium buildup in the fuel rods, and end-of-life maneuvering control allocations. Modifications of the assumed reactivity--penalties and alpha values of fuel isotopes were made in a few selected cases to determine the sensitivity of depletion and inventory requirements to such changes.*

The thermal reactor recycle calculations summarized here have been done treating the reactivity lifetime as an independent variable. The amount of uranium-235 in the uranium makeup is adjusted at each refueling step so as to attain a predetermined reactivity lifetime. The depletion and inventory requirements of each design configuration are calculated as a function of reactivity lifetime desired for both once-through and recycle operation. Recycle calculations may also be performed keeping the uranium makeup constant or otherwise varying so that reactivity lifetimes change at each refueling step.

A variation in fuel lifetime affects all of the elements of fuel cycle costs. In variable lifetime cycles, optimization of the cycle, and the calculation of plutonium value, become very sensitive to the economic assumptions. Some of these assumptions cannot credibly be made with useful accuracy at present, and are likely to change rapidly with time. Variable lifetime calculations which credit plutonium recycle with extension of reactivity lifetimes also beg the question of the cost differential relative to the equivalent once-through cycles. The once-through cycle also can usually be made to attain long lifetimes by the use of auxiliary control systems.

When different cycles are compared at equal fuel lifetimes, the costs for shipping and reprocessing may be set equal, and fuel fabrication costs equal except for ΔF_{AB} . (This term is defined as the average increment in cost for fabrication of recycle fuel over the equivalent fuel with uranium only.)

3. Crowther, R. and V. V. Zimmerman. (Private Communication)

*To be published.

In this approach, the reactivity lifetime extension effect of plutonium is measured as a decrease in depletion and inventory costs for any given fuel lifetime. These costs may be calculated directly from the fuel cycle burnup calculations for both recycle and once-through cases. Once-through cycles with lifetimes above 15,000 MWD/T are penalized by the extra cost of an extended shim control system (like burnable poisons, fixed shim poisons, or soluble poisons). This is usually a small correction term, of the order of one to two dollars per kilogram of fuel throughput, and thus may be regarded as offsetting part of the cost of ΔF_{AB} .

The approach of comparing cycles at fixed, predetermined lifetimes, is preferred as a stable basis for optimization of designs and cycles. It is believed that the approach also provides a more conservative estimate of plutonium value.

E. Results of Equilibrium Recycle Calculations

The concentration of plutonium at the beginning of the first recycle is equal to the final concentration in a similar once-through cycle, so the average concentration in the first recycle is about twice the average of the once-through cycle. On prolonged recycle, the total plutonium concentration increases to a steady-state value about double that at the end of a once-through cycle (see Figure 1). Of this plutonium, the fissile isotopes decrease from about 75 percent in the first cycle to about 60 percent of total plutonium at steady-state recycle.*

At the steady state, the concentration of the higher plutonium isotopes, especially plutonium-242, is increased relative to non-recycle operation. The steady-state level of plutonium-242 is determined by the balance between the rate of formation in each cycle and the rate of loss through capture and by chemical and fabrication losses.

The effect of plutonium-242 is essentially the same as the effect of any additional neutron absorber in the reactor core, for example, stainless structural material. To maintain the same thermal utilization, it is necessary to add about 0.7 grams of uranium-235 for each gram of plutonium-242 present in the fuel. The effect of a steady-state concentration of plutonium-242 is measured by the increase in uranium-235 and plutonium inventory required, as compared with once-through or first recycle conditions.

For the equilibrium conditions reached in the typical light water reactor calculations, the poison effect of plutonium-240 in the steady-state is about four times as large as that of plutonium-242. However, the poison effect of plutonium-240 is largely offset by its rapid rate of conversion to plutonium-241, which has a larger fission cross section and a larger η than either uranium-235 or plutonium-239. The apparent penalty in initial reactivity due to plutonium-240 is more than overcome since for the same initial reactivity margin the reactivity lifetime is 50 to 60 percent longer than a uranium-235 cycle. The change in inventory and depletion resulting from increase in plutonium-240 reaches very nearly its steady-state value in two

*The values given are for a reference case given in Appendix II.

FIGURE 1

Recycle Fuel Discharge Isotopic Compositions

to three cycles. Although plutonium-242 continues to grow in slowly its effect on thermal utilization is small, so the cycle approaches a steady state as far as inventory and depletion are concerned relatively quickly. Another way of expressing this is to say that the economics (depletion and inventory) of the cycle converge to a steady state more quickly than do the plutonium isotopic compositions.

A conservative--that is, low--measure of plutonium value is obtained by comparing the change in depletion and inventory cost of the steady-state recycle, with the once-through cycle.

Figures 2 and 3 summarize the inventory and depletion costs as a function of $V_{\text{water}}/V_{\text{fuel}}$ and reactivity lifetime of the fuel for stainless steel clad fuel. Figures 4 and 5 show the results for zirconium clad.

The uranium-235 makeup is adjusted for each cycle to attain a desired reactivity lifetime. Recycle cases at a given fuel lifetime may thus be compared with once-through cases at the same fuel lifetime. The data in Figures 2, 3, 4, and 5, the effective fuel material costs, for depletion plus inventory (excluding fabrication and reprocessing), have been calculated using the design and economic assumptions listed in Table I. A provisional credit of \$8/g for fissile plutonium is used for some of the once-through cases (Figure 4).

Where the difference between recycle and non-recycle cases is positive and $(D - D' - P) > \Delta \text{FAB}$, the effective value of plutonium is greater than the provisional \$8/g used in the calculation of Figure 2.

TABLE I
RANGE OF DESIGN AND ECONOMIC PARAMETERS USED
IN RECYCLE CALCULATIONS

1. July 1, 1962 uranium price scale in which uranium-235 is worth \$12.01/g at 90 percent enrichment; 4.75 percent inventory charges on fuel and plutonium; provisional minimum plutonium value of \$8/g fissile used for plutonium inventory and for credit in some once-through cases. (Cases near optimum design indicate higher effective values of plutonium.)
2. Effective water-to-fuel ratio (volume of water)/(volume of UO_2) = 2.0 for reference case; range covered is 1.3 to 2.5.
3. Fuel lifetime varies from 5,500 to 55,000 MWD/metric ton of uranium content.
4. Stainless steel cladding; rod diameter-to-clad thickness ratio = 33.4. For zirconium cladding diameter/thickness ratio is 20.0.

FIGURE 2

FIGURE 3

Depletion and Inventory Cost versus Irradiation and V_W/V_F : SS Clad

FIGURE 4

Depletion and Inventory Costs versus Irradiation
and V_W/V_F : Zr Clad, Once Through

FIGURE 5

Depletion and Inventory Costs versus Irradiation
and V_W/V_F : Zr Clad, Recycle

TABLE I (contd.)

Added variables investigated which are not reported explicitly:

1. Partial recycle , 0.5, and 0.99.
2. Full plutonium makeup (unlimited recycle).
3. Density of UO_2 ; 3.0, 7.0, and 10.0 g/cc.
4. Fuel diameter (clad ID) 0.3 to 0.5 inch.
5. Coupled thermal reactors.

F. Conclusions of Recycle Design Survey Calculations

Subject to the uncertainties in plutonium cross sections discussed elsewhere, the following preliminary conclusions have been drawn from the equilibrium cycle survey calculations.

1. Water-to-fuel volume ratio

The water-fuel volume ratio for minimum recycle costs increases with the fuel lifetime from about 1.8 at 11 MWD/kg to about 2.0 at 35 MWD/kg. * However, the sensitivity to the water-fuel volume ratio is small over the region 1.8 to 2.4 for lifetimes of about 18 to 55 MWD/kg; other design requirements would be controlling in this region. With stainless steel cladding, minimum costs for recycle fuel are in the region 2.0 to 2.4 for all lifetimes.

2. Rod diameter

For zirconium cases the optimum UO_2 diameter is about between 0.4 and 0.5 inch for all lifetimes. At 16.5 MWD/kg the largest margin between once-through and recycle costs occurs at about 0.45-inch diameter with stainless steel cladding (rod diameter/thickness = 33.4) minimum costs for short lifetimes (<10 MWD/kg) favor a rod diameter of 0.5 inch. From 10 to 20 MWD/kg, cost is insensitive to rod diameters between 0.4 and 0.5 inch; above 20 MWD/kg, rod diameter of about 0.4 inch is optimum. **

3. Fuel lifetime

For both zirconium and stainless steel clad, the maximum depletion and inventory cost differential (once-through less recycle) occurs for lifetimes of 11 to 16.5 MWD/kg. When $\Delta \overline{FAB}$ is taken into account, the optimum lifetime falls in the region of 15 to 20 MWD/kg.

* For a BWR the $(V_W/V_F)_{\text{effective}}$ is defined as $A_W(1-\Delta)/A_F$ where the A's are cross sectional areas of water and fuel and Δ is the void volume fraction at full power.

** The location of this optimum is provisional pending the exploration of diameter dependence for additional V_W/V_F ratios.

4. Cladding--for minimum cost cycles

Stainless steel cladding gives larger margins for $\overline{\Delta FAB}$ than does zirconium cladding. If $\overline{\Delta FAB}$ is assumed equal for the two materials, the net plutonium value is higher for stainless steel than for zirconium by \$1 to \$2/g.

5. Minimum depletion and inventory costs for once-through cycle

The once-through cycle gives minimum depletion and inventory costs (totalling about 1.4 mills/kwh) for water-to-fuel volume ratios between 2 and 2.5, fuel lifetimes between 11 and 24 MWD/kg and 0.4-inch rods with 12 mil stainless steel clad.

6. Minimum total costs--once-through cycle

If fabrication costs of \$40/kg and shipping and reprocessing costs of \$10/kg are added, * a minimum fuel cycle cost of about 1.7 mills/kwh is obtained for the once-through cycle with stainless steel clad, without plutonium credit, for irradiations between 20 and 25 MWD/kg.

7. Minimum total costs--recycle

Taking fabrication and processing costs as in (3) above, and $\overline{\Delta FAB}$ at \$4/kg, minimum fuel cycle costs with recycle and stainless steel clad are about 1.5 mills/kwh for irradiations between 16 and 25 MWD/kg.

8. For a fixed fuel lifetime of 16.5 MWD/kg, the difference in cost between once-through and recycle increases with increasing water-to-fuel ratio (considering only depletion and inventory costs and a provisional value of \$8/g fissile for plutonium credit and inventory).

A maximum margin of 0.10 mills/kwh is available for added plutonium worth and fabrication cost differential, $\overline{\Delta FAB}$. If the fabrication cost differential is less than this the effective plutonium worth will be increased over the provisional value.

For example, with a fabrication cost differential $\overline{\Delta FAB}$ of \$6/kg, the first cycle plutonium value becomes about \$9.30/g fissile. For $\overline{\Delta FAB}$ of \$4/kg the plutonium value becomes about \$9.80/g fissile.

G. Rate of Approach to Steady-State

Figure 1 shows the composition of uranium-235 and plutonium isotopes for eight successive cycles. The plutonium isotopes from -239 and -241 approach their steady state values rapidly, while plutonium-242 continues to change slowly.

The rate of approach to the steady state compositions is shown in Figure 6 in which the percentage departure from the steady state composition is given for plutonium isotopes, and also for the change in uranium-235 occurring during each cycle. Plutonium-239 approaches its steady-state most rapidly, being within 4 percent of the equilibrium value after two recycles. Plutonium-240 and -241 are within 4 percent after four recycles, while plutonium-242 requires seven cycles.

*Estimated costs for the volume of fuel fabrication available after 1968.

FIGURE 6

Rates of Approach to Steady State Compositions

Despite the relatively slow convergence of the plutonium-242 composition, the fuel cycle cost converges rapidly. This is indicated by the departure of uranium depletion from the steady state value, being within 10 percent after the first cycle, and within about 1 percent after three cycles.

Since depletion cost is roughly half of the total cost of the fuel cycle, the cycle cost converges substantially to steady-state after two cycles, and even the first cycle departs only slightly from the steady-state values.

H. Reactivity Lifetime and Control Range Effect of Plutonium Recycle

The uranium makeup added to recycle plutonium is adjusted to get a specified fuel lifetime. For the constant lifetime condition it is found that the reactivity swing per refueling interval is smaller than when the same lifetime is obtained by uranium fueling alone. The saving in reactivity margin of recycle versus once-through increases with fuel lifetime up to about 20,000 MWD/T. The once-through to recycle reactivity difference remains approximately constant at 2 percent Δk for irradiations from 20,000 to 55,000 MWD/T (Figure 7).

The decrease in reactivity required to offset fuel burnup is largely due to the effect of plutonium-240. Capture in plutonium-240 produces plutonium-241 at about 40 percent of the rate with which plutonium-239 is formed from uranium-238.* The plutonium-240 reduces the initial reactivity of the core, but the decrease in the rate of reactivity change with depletion more than offsets the effect of initial reactivity loss on lifetime.

The "value" to be ascribed to reduction in control range required can be considered significant for some designs. If measured purely as a gain in lifetime, for example, a 4 percent reactivity allocation to burnup increases reactivity lifetime from 12,500 MWD/T on a once-through cycle to 20,000 MWD/T with plutonium recycle. In general, for once-through lifetimes above 5,000 MWD/T, a 50 to 60 percent increase in lifetime for a given reactivity allocated to fuel burnup is attainable by the use of recycle.

For reactor designs which have some inherent limitation in the reactivity or control range which can be allocated to compensate for fuel burnup, the use of plutonium recycle could result in very important extensions of the reactivity lifetime attainable. Since the fabrication and reprocessing component of fuel cycle cost are inversely related to lifetime, important improvement in fuel cycle cost for such reactors might result. The apparent value of plutonium in such reactor designs can be quite high--higher than the value of uranium-235.

For most light-water power reactor designs, however, the reactivity lifetime extension is of lower worth than the saving in depletion and inventory costs. The strong dependence of fuel cycle cost on lifetime forces the design of large power reactors to provide sufficient control range to attain reactivity lifetimes long enough so that further increases have only a small effect on fuel cycle costs. For example, assume that a reactor is designed to attain 15,000 MWD/T on the once-through equilibrium cycle. The actual reactivity

*For the reference steady-state recycle given in Appendix II.

FIGURE 7

Reactivity Change versus Irradiation for Once-Through and Recycle

range provided is usually somewhat greater to cover the loadings which make the transition from a first core (which averages the nominal lifetime) to a steady-state multi-batch reloading pattern. Further additions to the control range can be made by use of burnable poisons in the fuel, or the use of other auxiliary control means. Another factor reducing the importance of the effect of recycle on reactivity is that the economic advantage of increased lifetime diminishes both as life-time increases and as fabrication and processing costs are reduced.

As has been noted in publications by Eschbach⁴ and by Puechl⁵ reactivity lifetime extension from plutonium which is high in plutonium-240 content may be important for specialized applications such as military or space power reactors in which reactivity lifetime may be the dominant performance consideration. A premium value for plutonium--higher than that of uranium-235--appears likely for military or space-power applications for thermal as well as fast reactors. The rate of growth or possible limits of such demand is still speculative, but if it has an effect, it can only serve to increase plutonium value.

I. Fabrication Costs for Plutonium-Containing Fuels

Experience in fabrication of mixed oxide fuels containing plutonium is now accumulating at an accelerating rate. A small fabrication facility has been making irradiation specimens of uranium-plutonium oxide at Vallecitos since 1959, and has in the last few months been superseded by completion of a larger facility. A production-scale facility exists at Hanford which has made the partial oxide loading of PRTR and has scheduled a loading for EBWR. Capability for oxide fuel fabrication containing plutonium is also available on a considerable scale at Nuclear Materials Corporation.

Just as with the case of commercial nuclear power itself, economic analysis is most significant when it leads to demonstration and use on an economic scale of operation. At present, there is only small scale experience and estimates of large scale costs. These estimates indicate that the increment in fabrication costs due to the presence of plutonium can be made quite small whenever sufficient volume of fuel fabrication becomes available. Specific estimates for boiling water reactor fuels have been developed in connection with the work reported in this paper, based on detailed design and cost analyses of large-scale fabrication facilities. This study indicates that the increment due to commercial fabrication with plutonium is probably less than 10 percent of the cost of fabrication of slightly enriched uranium fuel provided that fabrication volumes are at least one ton per year of plutonium content, or about 100 tons to 200 tons per year of oxide fuel at low enrichments. The cost differential may decrease with further increases in scale of operation. A speculative estimate--\$2 to \$3/kg--has been made of the minimum value of this differential.

4. Eschbach, E. A., et al. HW-72217. February, 1962.

5. Puechl, K. H. Nuc. Sci. and Eng., 12: 151. 1962.

J. Plutonium Value Estimate

The current value of the fabrication cost ($\overline{\Delta FAB}$) is probably larger than the difference in depletion and inventory costs available for any of the cases shown in Section I-E.* However, as discussed in the previous section this is believed to be a function of the present small-volume "custom" nature of fabrication of plutonium fuel. Cost differences equal to the more favorable cases in Figures 2 and 3 (\$6 to \$7/kg margin between once-through and recycle fuel material costs) require fuel fabrication volumes of one to two tons of plutonium contained per year. As discussed previously, such volumes of recycle fuel can be foreseen as being available between 1968 and 1970.

We also see in Figures 2 to 5 that the practical operating conditions for hydrogen-moderated power reactors are found for medium fuel lifetimes in the range of 12,000 to 20,000 MWD/metric ton and rod diameters covering most of the range of interest for water (and also organic-moderated) reactors. For fuel lifetimes appreciably above 20,000 MWD/T, the differences in fabrication plus inventory costs between once-through and recycle operation become negative. For such designs it is preferable to operate a once-through cycle rather than to recycle, provided that other uses of plutonium support a price of \$8/g fissile or higher.

The results shown in Figures 2 and 3 indicate that for the fabrication cost penalty expected in the late 1960's, an equilibrium cycle with stainless steel clad fuel can obtain costs slightly lower than those of a once-through cycle which credits plutonium at \$8/g fissile. This margin in depletion costs is believed to correspond to the minimum fuel value for the industrial scale use of plutonium in thermal reactors. Several additional value-increasing factors are qualitatively evident which can improve the effective worth of plutonium to the reactor operator.

1. The equilibrium cycle calculation slightly underestimates the value of fissile plutonium in the first few recycles. Thus, for a period after the fabrication differential becomes small, but before equilibrium compositions are obtained, the average plutonium worth in a growing economy as in Figure 10 is near the first recycle value, at least through the 1970's.
2. Plutonium-242 and plutonium-240 have useful fission cross sections in a fast reactor.** A small rate of growth of fast reactors

* For the reference case, fissile plutonium values are \$8.90/g, \$8.40/g and \$8.10/g for 1st, 2nd, and equilibrium recycles with $\overline{\Delta FAB} = \$4/\text{kg}$ and neglecting the other value-increasing factors which are of the order of \$1/g.

** For a typical fast reactor spectrum with oxide fuel, the effective fission cross sections for plutonium-240 and plutonium-242 are about 15 percent of the plutonium-239 cross section. (cf. Greebler, P., et al, GEAP-3287, November 1959). The value of $\nu\sigma_f/\sigma_a$ is greater than unity for both plutonium-240 and 242.

provides a useful outlet for highly irradiated plutonium. Even while fast reactor demand is too small to control prices, it can raise the effective values of plutonium in the thermal reactor.

3. Another positive factor neglected in value calculations is the liquidation value of the final plutonium inventory from a recycle reactor. The calculations as given here take account of the inventory charge, but no credit for the increase in the amount and the value of plutonium inventory over the first cycle discharge. Assuming (a) that the final liquidation value of plutonium is the same per gram of fissile isotopes as that on which the current inventory charge is based, (b) calculating present worth on the same basis as the inventory charge (4.75 percent per year), and (c) assuming that recycle is carried on for 15 years prior to liquidation of inventory we find that the present worth of the final liquidation is about 1.6 percent of the plutonium value per annum. Taking this factor into account would increase the margin available for (ΔFAB) by about \$2.75/kg. This would correspond to an increase in plutonium value of about \$0.60/g fissile for reference case given in Appendix II.
4. Another factor increasing plutonium value is the effect on the cost of control to attain a given reactivity lifetime. Taking the conservative procedure of equating this to the cost of introducing burnable poisons, and evaluating this at \$1 to \$2/kg of uranium, the net worth (again for the same reference case) amounts to an increase of 0.25 to 0.50 \$/g.

The additional effects listed above are not readily generalized, since they will vary with the particular reactor design and fueling strategy employed. However, net fuel values up to about \$10/g of fissile plutonium* appears attainable by using the light-water reactor technology now common in the United States. This value is in good general agreement with the "near-term" value estimated by the AEC, if "reactor-grade" plutonium is taken to be about 80 percent fissile.

II. PLUTONIUM USE IN FAST REACTORS

A. Ratio of Value of U-235 and Plutonium in Fast Reactors

When plutonium is used in a fast reactor in place of uranium-235, there are two important differences: (cf. Appendix Table II B).

1. The loading of fissile material required for given lifetime is about two-thirds as large for plutonium as for uranium-235.
2. The breeding ratio is about 0.3 higher with plutonium than with uranium-235.

*Related to the July 1962 uranium price scale and economic ground rules, and assuming the attainment of a sufficient scale of fabrication of recycle fuel so that the fabrication cost penalty becomes no more than \$5/kg. This value becomes about \$7/g fissile if adjusted to a future uranium price scale in which separative work is \$25/kg and feed cost is \$18/kg.

When large fast reactors which will meet safety and reliability requirements for central station power use can be built at economic capital costs, then such reactors would preferably be started and operated using plutonium. If sufficient plutonium is not available, fast reactors could be started on uranium-235 but at higher costs for the first one or two cycles. If a fast reactor attains power costs which make it attractive to build and competition with other types of power plants (both fossil and nuclear) then the demand for new reactor inventory will determine the market value for plutonium. If the reactor is initially loaded with plutonium instead of uranium-235, it will avoid the extra cost of:

1. The higher inventory of uranium-235 required, and
2. The low conversion ratio which the first one to two cycles would produce with uranium-235.

The difference in cost of the two paths of startup of the reactor would then define the additional premium which the operator of a fast reactor would be willing to pay in order to startup with plutonium rather than with uranium-235. This type of calculation (see Appendix II) gives a limiting value of about 1.6 for the ratio of value of plutonium fissionable isotopes to the value of uranium-235. The limiting value is applicable only if operation with the uranium-235 startup cycle is itself economic. More rigorously, the calculation defines the upper limit of the value of uranium-235 relative to the value of plutonium assuming that the plutonium cycle is marginally economic. The reactor operator can not afford to pay more than about two-thirds the value of plutonium for uranium-235 in competition with plutonium.

The minimum value for plutonium is established by examination of the price which would be tolerable for a fuel cycle which is assumed to be just at economic parity with a large water-cooled reactor. The capital cost of a large fast reactor is expected to be within ten to fifteen dollars per kilowatt of capital cost of a large water-cooled reactor. (The extra cost of a multi-circuit heat transfer system is expected to be largely offset by (a) smaller physical dimensions of the fast reactor, and (b) by the relatively high thermal efficiency of a liquid-metal reactor system, 35 percent to 40 percent.) The operating and maintenance cost of a liquid-metal coolant system is also expected to be slightly higher than with water-cooled systems. To reach parity in power costs with the water reactor, and to provide an incentive for development, the fast reactor will require fuel cycle costs which are about one-half mill/kwhr lower than those attainable in a water reactor. Fuel cycle costs with water-cooled reactors are about 1.3 ± 0.2 mills/kwhr on the projected 1970-1975 uranium price scale (which is about 80 percent of the July 1962 price scale). Accordingly, the maximum value of target fuel cycle costs for the fast reactor is about 0.8 mills/kwhr (on the projected 1970 price scale). The resulting price range for plutonium is determinable as a function of the fabrication and reprocessing costs; the interest charge rate assumed; and the conversion ratio and specific power which are attained.

For the specific power of present fast ceramic reactor designs,^{6, 7, 8} and breeding ratios only slightly greater than one, the plutonium price required to give 0.8 mills/kwhr fuel cost ranges from 5 to 10 dollars per gram of fissionable isotopes.

The lower value, around five dollars, applies to near-term estimates (1965 to 1970) reflecting high unit costs associated with small-scale of operation in fuel fabrication and chemical reprocessing. The higher plutonium value would come about from reductions (by about 50 percent) in fabrication and reprocessing costs due to increased volume of production. Such values might be considered probably average values over the lifetime of a plant which came into operation about 1970.

The foregoing values represent the present trend in fast oxide reactor design in which breeding gain is considered secondary to the attainment of minimum cost of power.

For the use of U-235 in a fast reactor, we may derive the ratio of plutonium value to U-235 for a single core load.

$$D+I+F+R+S = C$$

$$D'+I'+F'+R'+S'+ \overline{\Delta FAB} = C'$$

where

C = total cost of fuel for one core lifetime, dollars

D = depletion cost or credit

I = inventory cost

F+R+S = fabrication, reprocessing and shipping costs

$\overline{\Delta FAB}$ = increment in fabrication costs due to plutonium content.

Primed symbols refer to U-235 startup cycle.

Assuming fabrication, reprocessing and shipping are equal (except for $\overline{\Delta FAB}$) we may set the depletion and inventory costs equal for equal fueling costs with U-235 and plutonium-239. The value of $\overline{\Delta FAB}$ for fast reactor fuel has been estimated as \$16/kg.⁹ Equating the costs for the U-235 and Pu-239 cycles, one may solve for the ratio of plutonium value to the value of U-235. Using the fast reactor data in Appendix II, and $\overline{\Delta FAB} = \$16/\text{kg}$, we obtain

$$\frac{V_{\text{Pu}}}{V_{25}} = 1.65$$

where

$$U_{\text{Pu}} = \$/\text{gram of (Pu-239 + Pu-241)}.$$

6. Leitz, F. "Plutonium as a Power Reactor Fuel", Am. Nuc. Soc. Proceedings, September, 1962.

7. Horst, K. M., et al. "Core Design Study for a 500 MWe Fast Oxide Reactor", Trans. Am. Nuc. Soc., 5, No. 1: 126. 1962.

8. GEAP-3721. December, 1961.

9. Collins, G. D. GEAP-3824. January, 1962.

Using U-235 at 20 percent enrichment, V_{25} is \$11/g for the July, 1962 price scale, and about \$9/g in the projected scale for the 1970's*, we find the upper limit of the value of plutonium in such a fast reactor is then \$18.15/g for the 1962 scale, and \$14.85/g for the scale of the 1970's.

B. Inventory Growth Rate Required for a Fast Reactor Economy

In order for fast reactors to become a significant part of power generation, fast reactor capacity will necessarily grow at a rate which is two or three times higher (in terms of percent growth per year of fast reactors) than the overall growth rate of power capacity in percent per year. At some starting date, say 1975, fast reactors will constitute one percent of the installed capacity. If fast reactor capacity, thereafter, grows at twice the percentage rate of the total (14 percent per year growth rate), then after twenty years fast reactors have increased only from one to eight percent of the total power economy. Section III and Figure 11 show a possible pattern of growth which is limited only by the availability of plutonium.

On this basis, it is evident that for any technology which is expected to be a significant energy supply, it must be capable of sustained growth rate for several decades with doubling times of less than five years. Additions to reactor fuel inventory are required at this rate. This kind of inventory requirement will soon grow to equal the total supply of plutonium available from both fast and thermal reactors. A reasonable consequence is that the price of plutonium for new inventory will rise until it is marginally economic to start fast reactors with plutonium rather than with uranium-235.

During the period of rising plutonium price, a remedy for the reactor operator is to seek to reoptimize the reactor characteristics to produce as much additional plutonium as possible, that is, to raise the breeding ratio and specific power as far as possible within tolerable power costs. Breeding ratios as high as 1.5 and annual net breeding gains of perhaps 15 percent are possible but usually at some expense in capital and non-fuel operating costs. For these circumstances, the fast reactor economy will also tolerate higher plutonium prices because the increase in annual credit for plutonium exceeds the increase in inventory costs.

Eventually, fast reactors may become a large fraction of total installed capacity. At such time, large breeding gains will not be required. Fast reactors will be optimized to breeding ratios and doubling times which are equal to the demand. Supplemental plutonium from thermal reactors would not be required.

Uranium-235 can also be used to start-up fast reactors during the period of rapid growth. However, the reverse of the argument on the ratio of values in the fast spectrum dictates that if the plutonium cycle were just marginally economic, uranium-235 would have to be available at about two-thirds of the price of plutonium to be useable. To the extent to which the fast reactor attained power costs better than parity with the next most competitive power source, the permissible increase in fuel costs of uranium-235

*Based on UF_6 feed cost of \$18/kg and separative work of \$25/kg.

startup cycles would increase. However, the cycle using plutonium fuel would need to have nearly 0.7 of a mill margin over the next competing power source in order to permit the use of uranium-235 for the startup cycle if both plutonium and uranium-235 were priced at \$10/g. (cf. Figure 10)

C. The Rate of Development of Fast Reactors

Although there is little question in scientific principle of the feasibility of individual elements of the technology of fast reactors, there remain formidable engineering and economic problems. Solution of these problems is expected to require the construction and operation of another generation of test and prototype reactors together with related large-scale development programs. The design and operating criteria for safety, reliability, and operation and maintenance costs in large-scale plant operation will be obtained only by many reactor-years of operating experience. Acceleration of fast reactor development is occurring not only in the United States, but also in the Soviet Union, Britain, France and Germany. In addition to the half-dozen fast reactors now completed or nearing completion, a second generation of test reactors and small scale prototypes may be expected by 1970.*

The problems of insuring safe and reliable operation of fast reactor systems will be reflected in high costs for the early prototype stations, (as has also been true of early thermal reactor prototypes).

Nominally economic operation of fast reactors may be obtained somewhat earlier in countries using low-capital-charge rates, such as used in Britain, and presumably the Soviet Union. Competitive capital costs for fast reactors in the United States are initially likely only for very large sizes (500 to 1000 MWe). The time scale for completion and demonstration of reliable operation of initial economic projects accordingly will be difficult to compress. A large-scale demand for fast reactors, based on significant operating experience with prototype economic projects, is therefore unlikely much before the mid-1970's at present rates of development. The requirement for plutonium for experimental and pre-economic prototype reactors may nonetheless consume a large part of the plutonium available prior to 1975 (in Figure 11, about sixteen tons cumulative to 1975).

D. Plutonium Values in a Growing Dual Economy of Thermal and Fast Reactors

The plutonium inventory of prototype and initial marginally economic fast reactors will probably be obtained at prices determined by thermal recycle values. Higher plutonium prices (or uranium-235 startup) could not be economically justified for such projects. As the power costs attainable by fast reactors become better than just marginally economic (mainly by reductions in capital and non-fuel operating costs) an appreciable capacity of fast reactors will be built, and these can compete with thermal recycle for the available plutonium supply.

*Existing or nearing completion: EBR I, 1 MW thermal; EBR II, 17 MWe; Bn 5, 5 MWt (USSR) plutonium core; Enrico Fermi, 60 MWe; Rapsodie, 10 MWt; Dounreay, 15 MWe. Also in design stages are: a 200-300 megawatt electric prototype station for Britain, estimated for 1964 commitment, and a 800 to 1000 megawatt electric fast reactor station in the Soviet Union. Several additional small-scale experimental reactors are scheduled in the United States, and at least one or two medium-scale prototypes are likely by 1970.

When fast reactor growth becomes sufficient to consume a large fraction of the plutonium available, plutonium price may be expected to rise until the cost of power from thermal and fast reactors equalize.

Figure 8 indicates these relationships for the reference fast and thermal reactor cases described in Appendix II. *

Fast reactor power costs may eventually improve so that even when started with uranium-235 (or with plutonium at the price determined by the intersection of line 3 and line 4 in Figure 8) the power cost is lower than that of the thermal reactor at the same plutonium cost. (That is--the intersection of 3 and 4 becomes lower in cost than the intersection of line 1 and line 3.)

The uranium price scale used for Figure 8 is a projected "mid-1970's" scale in which diffusion plant feed cost is \$18/kg uranium as UF_6 and separative work cost is \$25/kg.

The intersection of lines 1 and 2 is the indifference value for plutonium on the projected price scale, neglecting the small value-increasing effects discussed previously which are expected to increase the value by \$1 to \$2/g. The intersection of line 3 and 4 indicates the limiting value of plutonium. At higher plutonium values, it would be cheaper to operate the fast reactor with uranium-235. The limiting value would be reached only if the capital and other non-fuel costs improved so that the intersection of 3 and 4 occurred at a cost lower than the intersection of 1 and 3. The intersection of 1 and 3 is the plutonium value limit which applies during the early economic operation of fast reactors.

The cost penalty of operating the fast reactor with uranium-235 is about 0.8 mills/kwh while plutonium remains near the thermal recycle value of \$6 to \$8/g and drops to 0.3 mills/kwh at the \$12/g value of plutonium.

III. SUPPLY AND VALUE RELATIONSHIPS

A. Estimated Rate of Accumulation of Plutonium from Power Reactor Operation

The relative importance of different routes for utilizing reactor-grade plutonium will depend somewhat on how soon tonnage quantities of reactor-grade plutonium become available.

The growth of electrical power generation, in kilowatt hours per year, has averaged about 7 percent per year since 1970. The forecasts made by the Federal Power Commission and the Edison Electric Institute,

*A breeding ratio of 1.08 was used for line 3 in Figure 13. As higher breeding ratios are attained, cost parity with the thermal reactor can be reached with higher capital and non-fuel costs, for example, 0.23 and 0.45 mills/kwh higher for breeding ratios of 1.19 and 1.35 respectively. "The apparent advantage of the higher breeding gains is offset by the higher capital costs required, so that other considerations, especially safety, control the optimum breeding ratio."

FIGURE 8

Power Cost and Plutonium Value Relationships Thermal and Fast Reactors

are in substantial agreement in predicting a continuation of near 7 per rate to 1980.¹⁰

Given the rate of growth of fossil-fueled power and with a knowledge of the distribution of power costs for non-nuclear plants, it is possible to estimate the fraction of the total power market which can be nuclear. Figure 9 shows the estimated distribution in power generation costs for fossil-fueled power plants committed up to 1966 for initial operation before and including 1970.

FIGURE 9

Distribution of Fossil Plant Power Costs Relative to 1966 Nuclear Plant Costs

10. The several forecasts are summarized in Electrical World, Vol. 151, No. 7, February 26, 1962, p. 34. The actual values are: FPC 7.4 and 6.3; EEI 6.8 and 6.6 annual percent increase for the decades ending in 1970 and 1980 respectively.

It is noted that prior to 1962 the Federal Power Commission forecast showed a much larger decrease in rate with time. Other forecasters have also seen compelling reasons why an indefinite continuation of a 7 percent exponential growth was doubtful. These differences make various authorities differ from the projection of a continued 7 percent growth extrapolation by a factor of two to three for the years 1980-2000. This may also be expressed as a range of uncertainty of about ten years in the time required to reach a generation rate of 1012 kwh/yr. For simplicity, the 7 percent growth assumption is used here. It is believed to introduce only a small uncertainty relative to the other factors inherent in the calculations presented here--- at least through 1980. The increasing uncertainties with long projections are obvious, but this does not materially influence the relationships and conclusions drawn from the interaction of thermal reactors and the early growth and fast reactor growth rates.

The overlap between this distribution and the estimated distribution of power costs from a large water-moderated reactor may be estimated as shown in Figure 10. The upper curve shows the estimated growth and total installed electrical capacity (taken at the 7 percent annual rate of increase) and the lower curve shows the nuclear capacity calculated on the assumption that one-third of the new stations for which the nuclear plant would achieve cost parity or better with the competitive fossil fuel plant will actually be constructed as nuclear units. It is found that this "competitive fraction" on the cost overlap basis increases nearly linearly with time being 1 percent of new capacity in 1961, 2 percent in 1962, 10 percent in 1970, etc. The curve is extrapolated on the conservative assumption that the pattern of 1 percent per year increase is the fraction of new capacity, which is nuclear, continues to about 1990, when nuclear power is between 18 and 30 percent of total electric power production.

FIGURE 10

The growth function for nuclear power is expressed analytically in Appendix III. Figure 11 gives the total plutonium production capability of corresponding to the installed capacity of Figure 10 assuming a fuel life-time of 16,500 MWD/metric ton. Figure 11 also indicates the effects of plutonium recycle. Recycle does not greatly reduce the total amount of plutonium available. This is because the average number of recycles will be only between one and two in a rapidly growing economy.

FIGURE 11

Estimated Plutonium Production and Availability
from Thermal Reactors

As noted in Section II, the demand for plutonium by fast reactors can raise the price of plutonium above its value for thermal recycle. For the reference non-recycle thermal reactor design (see Appendix I) a change in plutonium value of \$1/g changes the fuel cost by 0.03 mills/kwhr. If the fast reactor raises the worth of plutonium by \$6/g, the fuel cost of the thermal reactor falls by 0.18 mills/kwhr.

In Figure 9, 0.18 mill/kwhr decrease in fuel cost increases by about 40 percent the capacity with which nuclear plants are competitive. This has the interesting result that the development of economically competitive fast reactors increases the competitiveness of thermal reactors.

The effect of fast reactors on the growth of thermal reactor capacity is given in Figure 12.

FIGURE 12

Projected Nuclear Capacity with Fast Reactors

The lower curve of thermal capacity is the same as the lower curve in Figure 9. The upper thermal capacity curve indicates the effect of increasing plutonium value by use in fast reactors at a value \$6/g higher than the thermal recycle value. The uppermost curve and the fast reactor curve are derived on the assumption that during this period the economic startup of fast reactors requires plutonium, and growth rate is limited only by the availability of plutonium from both fast and thermal reactors. The derivation of the curves in Figure 12 is given in Appendix III.

B. Expected Patterns of Utilization of Plutonium

The role of thermal reactor recycle in the next 10 to 15 years may be regarded as providing a cushion between supply and demand for plutonium; (1) so that there is an assured minimum value for plutonium, and (2) so that there will be no issue of interest charges on a large stock-pile of non-working

plutonium.* In addition, the technology associated with thermal reactor recycle provides the techniques and large-scale handling experience which would otherwise develop more slowly with fast reactors alone.

The plutonium inventory associated with the generation of medium-scale prototypes which should come into operation by about 1970 is considerable. For the specific powers which appear feasible during this period, each project of a few hundred megawatts or more will have associated with it roughly a ton of fissile plutonium. Nearly all of the existing and scheduled fast reactor projects in the U. S. are fueled with U-235. Since U-235 is used in each case as a "stand-in" for the intended plutonium operation, the conversion of existing and future projects to plutonium may be expected to occur as fast as fuel fabrication experience and availability of plutonium permit.

Neglecting space power or military projects, and neglecting possible overseas requests, it appears that existing and planned fast reactor projects in the U. S. could by 1970 use well over half the reactor grade plutonium then available. If fast reactor development is successful in reducing capital costs and demonstrating reliable operation, it is conceivable that all available plutonium would be earmarked for fast reactor inventory after 1975.

Figure 13 illustrates the material flow relationships of the several categories of thermal and fast reactors. For the U. S. economy, with large diffusion plant capacity already in existence and largely amortized, the pattern of use shown in the lower line of the diagram is believed to represent the lowest cost route. The natural uranium reactors are unattractive because of their higher capital costs and the larger and longer development effort remaining. The U-235 fueled fast reactors are less attractive than the plutonium fueled variety because of their higher depletion and inventory costs. The following sequence of material use-patterns may be envisioned:

1. Slightly enriched thermal reactors; small plutonium production - under one ton per year - is used mainly for experimental purposes.
2. Plutonium production from thermal reactors exceeding one ton per year; plutonium use divided between thermal reactor recycle and experimental fast reactors.
3. Plutonium production from recycle thermal reactors exceeding five tons per year. Plutonium use divided between recycle and inventory for initial large scale economic fast reactors.
4. Plutonium production from thermal and fast reactors exceeding 10 tons per year. Plutonium use mainly for inventory of growing capacity of fast reactors; some thermal reactor recycle to cushion supply and demand.

*There exists a stockpile of "weapons-grade" plutonium (low in Pu-240) which may be estimated at several tens of tons of plutonium. The timing of possible conversion of this material from military functions is highly speculative. It would involve a large loss in value relative to the cost of production. Based on the pre-1963 AEC price scale, the value of weapons-grade material is \$30/g to \$45/g.

5. Plutonium production from fast reactor approaches rate required for further growth of new fast reactor inventory. (Or cost margin of fast reactor becomes large enough to permit economic start-up on U-235.) Growth of thermal reactor capacity diminishes relative to rate of growth of fast reactor capacity.

FIGURE 13

Patterns of Utilization of Plutonium

C. Plutonium Price Formulas

Alternative pricing formulas which might be used are:

1. Dollars per gram of reactor-grade without regard to isotopic content.
2. Dollars per gram of fissile content (plutonium-239 and -241) without regard to the ratios of the other isotopes. (This is the preferred near-term formula.)
3. A four-factor formula assigning a separate value to each isotope.

4. A variable four-factor formula in which the value of each isotope is a function of the composition and the spectrum of the reactor type or design in which it is to be used.

The general value function (4) is expressed as:

$$V_{Pu} = V_9X_9 + V_0X_0 + V_1X_1 + V_2X_2$$

where

- V_{Pu} = value of specific composition of plutonium, \$/g.
 V_i = value of isotope i in \$/g (spectrum dependent).
 X_i = atom fraction of isotope i (fraction of total plutonium).

Subscripts 9, 0, 1, 2 refer to plutonium-239, 240, 241, 242, respectively.

Either 1 or 2 can represent the weighted average of the more precise formulas as long as plutonium compositions and uses are typical of the first decade of thermal recycle. However, data sufficient to permit the establishment of the more precise formulas such as 3 or 4 should become available in the next few years.

The first formula becomes inequitable if plutonium varies significantly in plutonium-240 content. A preferable formula for near-term use is (2), which prices plutonium on the basis of its content of fissile isotopes.

A two-term formula intermediate between (2) and (3) above has been proposed by Eschbach, et al.⁴ with a positive term for fissile content, a negative term for plutonium-242, and with plutonium-240 neglected except as a diluent. The accuracy of cross section data does not appear to warrant this refinement at present. When sufficiently accurate data becomes available, formula (3) might be preferred if plutonium of widely different isotopic ratios is produced. Different isotopic coefficients would be required for significantly different reactor spectra, as in formula (4). Expressions for the individual isotopic value coefficients have been developed and will be published later.

D. Some Aspects of Near-Term Plutonium Buyback

A "near-term fuel value" price of \$8/g has recently been reaffirmed by the AEC for reactor-grade plutonium as the nitrate.¹¹ Discounting later more efficient uses, the results of this study substantiate a probable fuel value for plutonium near \$10/g of plutonium-239 plus plutonium-241, or near \$8/g of "reactor-grade" material (defining this as first-cycle reactor output about 80 percent fissile). If the \$8/g formula were to remain unchanged until about five tons of plutonium have become available (about 1970 to 1971), the total cost of a plutonium buyback program would be of the order of \$40 million over this period, with an uncertainty of perhaps \$5 million.

4. Eschbach, E. A., et al. HW-72217. February, 1962.

11. Seaborg, G. T. AEC Release. August 21, 1962.

Most of the available reactor-grade plutonium can find useful employment as fuel for experimental reactor programs. Such use would displace the much more expensive low-240 plutonium, and is thus preferable from both an experimental and cost standpoint.

APPENDIX I

THUR CODE

THUR calculates reactivity and isotopic fuel concentration changes for steady-state operation with fuel recycle. It also does these calculations for once-through steady-state cycles (i. e., no recycle). Either thorium-uranium or uranium-plutonium fuel cycles may be used. (Also plutonium may be used in the thorium cycle.) For recycle operation, any desired fraction (0 to 1) of the discharged uranium or plutonium may be specified for input to the next cycle. For the present discussion, only the uranium-plutonium cycle application will be considered.

Figure A1 shows the flow of a THUR calculation. The input processor has some routines which utilize simple input data on geometry and composition to compute initial isotopic concentrations, uranium-238 epithermal cross sections from resonance integral data, thermal cross sections corrected for temperature and spectral hardening, and geometrical buckling.

The subroutine MUGO¹² is a multi-group diffusion theory calculation of flux and reactivity. It is zero dimensional and utilizes a simple geometric buckling to take into account neutron leakage. Inelastic scattering and Selengut-Goertzel treatment of hydrogen slowing down are available for many-group calculations, and they may be bypassed for few-group calculations. The output includes material neutron balance data and flux-averaged one-group fission and capture cross sections for each isotope. The one-group cross sections are used in the burnup equations of the BURNER subroutine discussed below.

Multi-group cross sections for the MUGO calculations are contained in an input cross section file. The uranium-238 epithermal cross sections in the file are modified by the input processor to give the resonance integral for the case being calculated, and thermal cross sections in the fuel are also modified by the input processor as was indicated above. The plutonium-240 capture cross section in the group containing the strong 1 ev resonance is modified during the course of the calculation by an approximate self-shielding

12. Greebler, P. and C. J. Lavios. MUGO, an IBM-653 Machine Computation Program for Multigroup Analysis of Fast and Intermediate Reactors, GEAP-3063. September 15, 1958. (This program has since been transferred to the TRANSAC 2000 Computer.)

FIGURE A1

Flow Chart of THUR Calculations

formula.^{13, 14} For other fuel isotopes, the epithermal cross section values in the file are not altered during the calculation. The code calculates the steady-state xenon-135 and samarium-group fission product poisoning.

The BURNER subroutine calculates isotopic concentrations for an irradiation $Z = \phi t$, where Z is estimated to give the desired burnup; and improved estimates are made during the course of the calculation as indicated in Figure A1. This subroutine also computes average isotopic concentrations during the irradiation Z , corresponding to an average core composition for infinite-batch (continuous) refueling, and it computes burnup in MWD/T. The burnup equations take into account steady-state recycle directly by coupling the initial and discharged plutonium isotope concentrations. To illustrate, the differential equation that describes the concentration of plutonium-241 during irradiation is:

$$\frac{dN_{241}}{dZ} = \bar{\sigma}_{c240} N_{240} - \left(\bar{\sigma}_{f241} + \bar{\sigma}_{c241} + \frac{\lambda_{241}}{f\phi} \right) N_{241} \quad (1)$$

with the initial condition

$$(N_{241})_{\text{initial}} = F_{\text{rec}} N_{241} + (N_{241})_{\text{makeup}} \quad (2)$$

where N_{240} and N_{241} are concentrations of plutonium-240 and plutonium-241 at irradiation Z ; $(N_{241})_{\text{initial}}$ is the value at $Z = 0$; $(N_{241})_{\text{makeup}}$ is the plutonium-241 supplied as additional plutonium makeup; F_{rec} is the fraction of discharged plutonium to be recycled; $\bar{\sigma}_c$ and $\bar{\sigma}_f$ are one-group cross sections from MUGO for the isotopes indicated, λ_{241} is the decay constant of plutonium-241, f is the plant load factor, and ϕ the one-group neutron flux at rated power.

The solution of Equations (1) and (2) for N_{241} involves N_{240} ; which, in turn, is obtained from two equations involving plutonium-239, etc. through neptunium-239 and uranium-238. The resulting expression is

$$N_{241} = \sum_{m=1}^5 A_m \exp \left[- \left(\bar{\sigma}_{f,m} + \sigma_{c,m} + \frac{\lambda_m}{f\phi} \right) Z \right] \quad (3)$$

-
13. The expression for the effective plutonium-240 cross section in the group containing the 1 ev resonance is

$$\sigma_{\text{eff}} = \frac{\sigma_{\infty}}{[1 + C\bar{N}]}$$

where $\bar{N}$ is the average plutonium-240 isotopic concentration and C an input parameter computed to fit the effective resonance integral calculated using the method of Reference 3.

14. Adler, F. T. and L. W. Nordheim, Tables for the Computation of Resonance Integrals, GA-377. June, 1958.

where m is the isotope index in the uranium-238 chain ($m=1$ for uranium-238, 2 for neptunium-239, 3 for plutonium-239, 4 for plutonium-240, and 5 for plutonium-241); and the coefficient A_m is given by

$$A_5 = \frac{(N_{241})_{\text{makeup}} + F_{\text{rec}} \sum_{m=1}^4 A_m \left\{ \exp \left[-(\bar{\sigma}_{f,m} + \bar{\sigma}_{c,m} + \frac{\lambda_m}{f\varphi}) Z \right] - 1 \right\}}{1 - F_{\text{rec}} \exp \left[-(\bar{\sigma}_{f,241} + \bar{\sigma}_{c,241} + \frac{\lambda_{241}}{f\varphi}) Z \right]} \quad (4a)$$

$$A_4 = \left[\frac{\bar{\sigma}_{c,240}}{\bar{\sigma}_{f,241} + \bar{\sigma}_{c,241} + \frac{\lambda_{241}}{f\varphi} - \bar{\sigma}_{f,240} - \bar{\sigma}_{c,240}} \right] \quad (4b)$$

• [Expression (4a), except 240 replaces 241 and the summation is carried only through $m=3$.]

$$A_3 = \left[\frac{\bar{\sigma}_{c,240} \bar{\sigma}_{c,239}}{\left(\bar{\sigma}_{f,241} + \bar{\sigma}_{c,241} + \frac{\lambda_{241}}{f\varphi} - \bar{\sigma}_{f,239} - \bar{\sigma}_{c,239} \right) \left(\bar{\sigma}_{f,240} + \bar{\sigma}_{c,240} - \bar{\sigma}_{f,239} - \bar{\sigma}_{c,239} \right)} \right] \quad (4c)$$

• [Expression (4a) except 239 replaces 241 and the summation is only through $m=2$.]

$$A_2 = \left\{ \prod_{m=3}^5 \left[\frac{\bar{\sigma}_{c,m-1}}{\bar{\sigma}_{f,m} + \bar{\sigma}_{c,m} + \frac{\lambda_m - \lambda_{\text{Np239}}}{f\varphi}} \right] \right\} \left[\frac{\bar{\sigma}_{a,238} (N_{238})_{\text{initial}}}{\bar{\sigma}_{f,238} + \bar{\sigma}_{c,238} - \frac{\lambda_{\text{Np-239}}}{f\varphi}} \right] \quad (4d)$$

$$A_1 = \prod_{m=2}^5 \left[\frac{\bar{\sigma}_{c,m-1}}{\left(\bar{\sigma}_{f,m} + \bar{\sigma}_{c,m} + \frac{\lambda_m}{f\varphi} - \bar{\sigma}_{f,238} - \bar{\sigma}_{c,238} \right)} \right] \quad (4e)$$

If a search on initial uranium-235 concentration, additional plutonium makeup, or burnable poison concentration is requested, the program performs a criticality test, as indicated in Figure A1. If the reactivity at batch discharge has not converged to K_{test} within a specified limit, the initial concentrations are altered in accordance with input instructions and/or estimates based on earlier iterations. K_{test} is set to differ from unity to take into

account reactivity effects not directly computed, such as maneuvering allowance at end-of-life, non-uniform plutonium buildup in the fuel rods, and effect of non-uniform void distribution on the isotopic and reactivity changes with burnup.

Cross Sections

Table IB shows neutron balance based on three-group cross sections and twenty-group cross sections for the case of 2 to 1 water-to-fuel ratio, steel clad, high density UO_2 and 99 percent plutonium recycle with uranium-235 makeup. For this case the thermal plutonium-239 alpha value is 0.66; and 0.86 and 0.85 were used as epithermal alpha values for plutonium-239 and plutonium-241 respectively. These alphas are significantly on the pessimistic side of the area of uncertainty and tend to yield a low plutonium value. (The effects on recycle economics of lowering these alpha values were discussed in the main body of the paper.) The thermal cross section hardening is based on the heavy gas model for thermalization,¹⁵ which tends to give too hard a thermal spectrum and, hence, too high a plutonium-239 alpha value.

The uranium-238 resonance integral was computed from the following fit of experimental data.^{16, 17}

$$I = 1.1 + I_0 \exp [\alpha (\sqrt{T} - \sqrt{293})]$$

where

$$I_0 = 30 \sqrt{\frac{S}{M} + 0.077}$$

$$T = T_x + 0.35 (T_{\max} - T_x)$$

$$\alpha = 0.00696 - 0.000262 \frac{M}{S}$$

The effective surface, taking into account fuel rod interaction, was computed from Bell's formula.¹⁸ The Dancoff-Ginsburg formula¹⁹ which

15. Wilkins, J. E., Jr. CP-2481. 1944.
16. Arnold, W. H., Jr. and R. A. Dannels. I, T, and α from "The Doppler Coefficient of U^{238}O_2 ", Transactions of ANS, June 1960. p. 229.
17. Hellstrand, E. " I_0 from Measurements of the Effective Resonance Integral in Uranium Metal and Oxide in Different Geometries", J. App. Phys., 28. December 1957.
18. Bell, G. I. "A Single Treatment for Effective Resonance Absorption Cross Sections in Dense Lattices", Nuc. Sci. and Eng., 5, p. 2: 138-139. 1959.
19. Dancoff, S. M. and M. Ginsburg. Surface Resonance Absorption in a Close-Packed Lattice, CP-2157. 1944.

yields a slightly higher and more accurate resonance integral, was used in a few cases to check the sensitivity of the recycle economics to this parameter.

Cross sections for long-lived fission products were obtained from Nephew's calculations.²⁰ Both thermal and epithermal fission product cross sections in the file were adjusted for neutron spectrum, fractional burnup of fissile material, and the relative contributions by uranium-235 and plutonium to the total source of fission products.

Both three-group and twenty-group cross section sets were used in the THUR calculation. For the cases using plutonium makeup in natural uranium, with plutonium recycle, the spectrum was too hard for three-group calculations, and the twenty-group set was utilized. For the uranium-235 makeup cases, the three-group set was found to be adequate when the second group cross sections were modified from one case to another to take into account resonance capture interactions by the various fuel isotopes. Table IA shows initial and final uranium-235 concentrations, discharge plutonium isotopic concentrations, and the neutron balances for a once-through cycle using three-group and twenty-group cross sections respectively.

20. Nephew, E. A. Thermal and Resonance Absorption Cross Sections of the U²³³, U²³⁵ and Pu²³⁹ Fission Products, ORNL-2869. January, 1960.

TABLE IA
THERMAL REACTOR ONCE-THROUGH CYCLE NEUTRON BALANCES

3-Group versus 20-Group
 $(W/F)_{\text{eff}} = 2.0$, $F_{\text{rec}}^* = 0$, U-235 Makeup, $\rho\text{UO}_2 = 10 \text{ g/cc}$

<u>Percent</u>	<u>3-Group</u>	<u>20-Group</u>
** e_i^{25}	2.476	2.546
e_f^{25}	1.052	1.137
e_{Dis}^{49}	0.373	0.399
e_{Dis}^{40}	0.130	0.116
e_{Dis}^{41}	0.067	0.076
e_{Dis}^{42}	0.016	0.017
U-238 captures	25.229	25.814
Np-239 captures	0.013	0.013
Pu-239 captures	6.347	6.338
Pu-240 captures	3.243	3.569
Pu-241 captures	0.457	0.478
Pu-242 captures	0.072	0.066
U-235 captures	6.100	6.015
U-236 captures	0.397	0.359
U-237 captures	0.001	0.001
Fiss. prod. captures	2.500	2.402
Sm-gp captures	0.736	0.717
Xe captures	1.854	1.786
Ox (fuel) captures	-----	0.129
Cr captures	SS { 5.214 }	0.886
Fe captures		3.376
Ni captures		0.612
Ox (water) captures	H_2O { 4.995 }	0.144
H captures		4.594
U-238 fissions	2.600	2.809
Pu-239 fissions	9.823	9.780
Pu-240 fissions	0.008	0.008
Pu-241 fissions	1.061	1.138
Pu-242 fissions	~0	0.001
U-235 fissions	28.051	27.728
U-236 fissions	0.004	0.005

* F_{rec} = fraction of plutonium recycled.

** e = enrichment (assuming fuel to be all fissile and fertile isotopes).
 Superscripts 25, 49, 40, etc., are notation for isotope U-235, Pu-239, Pu-240, etc., respectively. Subscripts i, f, and Dis. stand for initial, final and discharge, respectively.

TABLE IB

THERMAL RECYCLE NEUTRON BALANCES

3-Group versus 20-Group
 $(W/F)_{\text{eff}} = 2.0$, $F_{\text{rec}} = 0.99$, U-235 Makeup, $\rho\text{UO}_2 = 10 \text{ g/cc}$

<u>Percent</u>	<u>3-Group</u>	<u>20-Group</u>
e_i^{25}	2.140	2.211
e_f^{25}	1.128	1.267
e_{Dis}^{49}	0.453	0.561
e_{Dis}^{40}	0.250	0.223
e_{Dis}^{41}	0.167	0.206
e_{Dis}^{42}	0.146	0.174
U-238 captures	21.335	22.433
Np-239 captures	0.007	0.007
Pu-239 captures	8.512	8.983
Pu-240 captures	8.411	8.889
Pu-241 captures	2.321	2.387
Pu-242 captures	2.265	2.320
U-235 captures	4.267	4.013
U-236 captures	0.257	0.233
U-237 captures	~0	~0
Fiss. prod. captures	2.056	1.835
Sm-gp captures	0.764	0.732
Xe captures	1.675	1.503
Ox (fuel) captures	-----	0.129
Cr captures	SS { 3.878 }	0.602
Fe captures		2.229
Ni captures		0.433
Ox (water) captures	H ₂ O { 3.615 }	0.144
H captures		3.007
U-238 fissions	2.581	2.786
Pu-239 fissions	12.768	13.371
Pu-240 fissions	0.030	0.030
Pu-241 fissions	5.249	5.501
Pu-242 fissions	0.017	0.022
U-235 fissions	18.727	17.169
U-236 fissions	0.003	0.004

APPENDIX II

TABLE IIASUMMARY OF WATER REACTOR REFERENCE CASE

Cladding	0.012" SS
Reactor Power, MWe	1000
Thermal Eff., Net, %	31.2
Fuel Exposure, MWD/1000 kg	16,500
Specific Power, Th kw/kg	20
Fabrication Cost, \$/kg; U Fuel	36
(U+Pu Fuel)	40
Reprocessing, \$/kg Pu + U	
Separation	10.00
Shipping	4.00
Conversion	3.00
Losses, Uranium, %	1
Plutonium, %	1
Time, Years	8
Cooling Ship., Repro., and Fab'n	0.57
Time, Core	2.82
Use Charge or Inventory Charge, % (after 1970, 10%)	4.75, 10.0
Capital Charge Rate, Plant and First Core	14
Capital Cost of Plant, Gross \$/kwe	150
Load Factor, %	80

	<u>Uranium-235</u>	<u>Plutonium Recycle</u>	
		<u>First Recycle</u>	<u>Steady-State</u>
E_i , Atom % U-235	2.476	2.153	2.140
E_f , Atom % U-235	1.052	1.045	1.128
Pu Disch., Total, %	0.586	0.811	1.016
Pu Disch., 49 + 41, %	0.440	0.548	0.620

TABLE IIB

REFERENCE CASE FAST REACTOR

Cladding	0.015" SS
Reactor Power, MWe	500 to 1000
Thermal Eff., Net %	37
Fuel Exposure, MWD/1000 kg of Pu+U (core average)	110,000
Specific Power, kw/kg Pu+U	128
Fabrication Cost, \$/kg Pu+U	
Core and Axial Blanket	136
Radial Blanket	52
Reprocessing, \$/kg Pu+U	
Separation	20
Shipping	
Core and Axial Blanket	10
Radial Blanket	7
Losses, %	2
Time, Fab., Years	0.25
Cooling, Ship., Repro.	0.42
Core	2.14
Use Charge, % (after 1970)	10
Capital Charge, plant and first core, %, p. a.	14
Capital Cost, Gross, \$/kwe	165
Load Factor, %	80

CORE	Pu Case Initial	Atom % Final	U-235 Case Initial	Atom % Final
U-238	79.05	70.09	80.39	72.76
U-235	---	---	19.61	8.57
U-236	---	---	---	2.15
Pu-239	12.14	9.53	---	4.43
Pu-240	6.79	6.67	---	0.38
Pu-241	1.39	1.39	---	0.02
Pu-242	0.63	0.63	---	---

BLANKET Discharge Compositions

	Axial	Radial	Axial	Radial
Pu-239	2.85	1.35	2.53	1.20
Pu-240	0.17	0.036	0.15	0.032
Pu-241	0.006	---	0.005	---
Pu-242	---	---	---	---

Breeding Ratio*	1.19	0.88
-----------------	------	------

* The core design is similar to that described by Horst, et al., GEAP-3721, December 1961. Since that report was issued, improved values of cross-sections have increased the effective breeding ratio of the reference design from 1.08 to 1.19, mainly due to the decrease of alpha of 239 in the intermediate energy range.

APPENDIX III

I. Nuclear plant growth for light water reactor thermal reactor economy only.

A. Total electrical capacity

$$P = P_o e^{at} \text{ megawatts electrical}$$

B. Total electrical plant additions per year

$$\frac{dP}{dt} = a P_o e^{at} \text{ MWe/year}$$

C. Thermal nuclear plant additions

$$\frac{dT}{dt} = b a P_o e^{at} \text{ MWe/year}$$

D. Total nuclear (thermal) capacity

$$T = \left[\int_0^t \frac{dT}{dt} dt = \frac{b P_o}{a} e^{at} (at - 1) + 1 \right] \text{ MWe.}$$

$$P_o = 1.6 \times 10^5 \text{ MWe}$$

$$a = 0.07 \text{ year}^{-1}$$

$$b = 0.01 \text{ year}^{-1}$$

$$t = 0 = 1960$$

II. Plutonium from thermal reactor operation

A. Plutonium production

$$G(t) = \text{kg plutonium/year} = C_1 T(t - \delta)$$

$$T = \text{MWe of thermal reactors}$$

$$C_1 = \text{kg plutonium/year per MWe of capacity}$$

$$\delta = \text{delay from production to availability of plutonium}$$

B. Plutonium accumulation

$$\int_0^t G(t) dt = C_1 \int_0^{t-\delta} T(t) dt \quad \text{Kg plutonium}$$

$$C_1 = 0.26 \text{ kg fissionable Pu/yr if no recycle}$$

$$= 0.16 \text{ kg fissionable Pu/yr if economy averages one recycle}$$

$$= 0.115 \text{ kg fissionable Pu/yr if economy averages two recycles (for 80\% capacity factor, and 16.5 MWD/kg).}$$

III. Plant growth for mixed fast and thermal reactor economy:

Assumptions:

Plutonium priced on basis of fast reactor value after 1967,

Fast reactor economically competitive after 1973.

A. Nuclear Plant Additions

$$\frac{dN}{dt} = btaP_o e^{at}; \quad t < 7 \text{ MWe/year}$$

$$\frac{dN}{dt} = 1.6 btaP_o e^{at}; \quad t \geq 7 \text{ MWe/year}$$

B. Fast reactor plant additions

$$\frac{dF}{dt} = 0; \quad t < 10$$

$$\frac{dF}{dt} = \frac{500}{S}; \quad 10 \leq t < 13$$

$$\frac{dF}{dt} = \frac{G(t - \gamma)}{S} + \frac{500}{S} \text{ MWe/year}; \quad t \geq 13 \text{ years}$$

$$= \frac{C_1 T(t - \gamma)}{S} + \frac{(C_2 F(t - \gamma))}{S} + \frac{500}{S}$$

$$= \frac{C_1 N(t - \gamma)}{S} + \frac{(C_2 - C_1)}{S} F(t - \gamma) + \frac{500}{S}$$

$$= \frac{C_1 N(t - \gamma)}{S} + \frac{C_2 - C_1}{S^2} \int_{10}^{t - \gamma} G(t) dt + \frac{500}{S}$$

C. Thermal reactor plant additions

$$\frac{Dt}{dt} = \frac{dN}{dt} - \frac{dF}{dt}; \quad \text{MWe/year}$$

$S = 2.1 \text{ kg of fissionable Pu required per MWe for Fast Reactor}$

$C_2 = 0.134 \text{ kg per year per MWe Fast Reactor}$

$\gamma = 3 \text{ years}$

IV. Nuclear capacity in mixed fast-thermal economy

A. Total nuclear capacity

$$\begin{aligned}
 N &= \frac{dN}{dt} dt = \frac{bP_o}{a} \left[e^{at} (at - 1) + 1 \right] \text{ MWe}; t \leq 7 \\
 &= 1.6 \frac{bP_o}{a} \left[e^{at} (at - 1) + 1 \right] - 0.6 \frac{bP_o}{a} \left[e^{7a} (7a - 1) + 1 \right]; t \geq 7
 \end{aligned}$$

B. Fast reactor capacity

$$F = 0; t \leq 10$$

$$F = \int_0^t \frac{dF}{dt} dt; t \geq 10, \text{ MWe}$$

C. Thermal reactor capacity

$$T = (N - F); \text{ MWe}$$

PREPARED DISCUSSION, SESSION IVU.K. STUDIES RELATING TO PLUTONIUM BURNING
IN THERMAL REACTORS

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In the U.K. both theoretical and experimental studies have been undertaken which are relevant to the reactor physics of plutonium burning in thermal reactors. This work may be summarised briefly as follows.

Survey calculations using relatively simple physics models were undertaken to explore the trends and arrive at some indication of the value of Pu-239 as a thermal reactor fuel. The results were sufficiently encouraging to suggest that more detailed studies with methods tailored to suit individual reactor systems were justified; the techniques used in these studies may be of some interest.

The first series of calculations were made for the High Temperature Gas Cooled Reactor Concept as typified by the O.E.C.D. DRAGON reactor now under construction at Winfrith. In this work the 43 group neutron spectrum code STEWPOT was employed. This code is written to provide for either crystalline solid or free gas thermalisation. Early studies showed the effect, referred to in other papers to this meeting, that the reactivity change for a given core life can be made much smaller with plutonium fuel than with U-235 enrichment. It was also found that the life-time depended on the effective Pu-240 cross section; multi-group Carlson calculations were made to fix the values appropriate to various types of fuel element geometry. It emerged that reactors with the same total fissile and fertile loadings but different fuel configurations can have significantly different reactivity lifetime characteristics and fuel cycle economics.

Currently studies are being made for the A.G.R. (Advanced Gas Cooled Reactor), a concept which has reached the stage of a large scale experimental unit now complete at Windscale. Reactors of this type are fuelled with oxide clusters, moderated by graphite and cooled by CO₂. Lattice heterogeneity effects are very significant and the STEWPOT technique is not applicable. A suitable method for dealing with the thermal processes known as SPECTROX¹ has been described in the literature. This technique provides a multi-group representation of the mean neutron energy distribution in fuel and moderator based on a heavy gas thermalisation model. Spatial effects are dealt with by collision theory within the cluster and diffusion theory in the moderator. The burn-up version of SPECTROX requires supplementary data concerning fast neutron effects which is derived by Monte-Carlo methods.

Plutonium fuelling studies are also in progress for the Steam Generating Heavy Water Reactor concept. This is a pressure tube reactor, fuelled with Zircaloy clad low enrichment oxide clusters, cooled by boiling light water, and moderated by heavy water. In this case the presence of two different moderators at two different physical temperatures implies that fairly elaborate analytical schemes are required even for survey or initial design type calculations. To meet this need the METHUSELAH 5 group computer code has been developed. The physical model is an extension of the Bettis 4 group model using two thermal groups overlapping in energy. One group is entered through moderation in the cluster, while the other is entered through the bulk D_2O . Inter thermal group transfer cross sections² operate on the diffusing neutrons. Spatial distribution and resonance self shielding effects are dealt with by diffusion or collision theory routines as appropriate, and the programme enables changes of isotopic concentrations in the various rods of the cluster to be followed through burn-up, thus permitting studies of non-uniform cluster loadings.

Reactor physics experiments with plutonium bearing fuels are being undertaken with both graphite and water moderators in a number of experimental facilities. Experiments with 20 mil Pu/Al plates moderated by light water were described in a paper presented to the I.A.E.A. Light Water Panel Meeting in Vienna. Similar fuel has also been used with graphite moderator, the range from 2300 to 14,000 carbon to fissile atom ratio being covered. In this case, the plates were mounted in a graphite matrix in the SCORPIO II subcritical assembly. This assembly can be heated to the power reactor temperature range and measurements were made at 20 C, 200 C, and 400 C. Buckling measurements and reaction rate studies with miniature fission chambers were made. These experiments are now complete and the analysis shows good agreement with the STEWPOT-CARLSON type of calculation referred to in connection with the High Temperature Reactor.

At present the SCORPIO facilities are being used for studies with metallic bars of depleted uranium enriched with plutonium. These bars correspond approximately in composition to reject fuel from the power reactor programme. Two series of experiments are being made. In the first, the plutonium is uniformly distributed through the bars. Two lattices of this type have now been studied. Comparison measurements will be made with the same quantity of plutonium concentrated in an aluminum annulus surrounding the bar. This should enable an estimate to be made of the importance of spatial distribution effects. Analysis of these experiments is proceeding by the Monte-Carlo and SPECTROX methods.

Next year experiments with mixed PuO_2 - UO_2 fuel are planned using both graphite and water moderators.

To summarise. Preliminary survey studies are being followed up by detailed calculations related to specific reactor designs. The elaborate methods of analysis used are being checked as far as possible against

experimental data. More data for both graphite and water moderation are being accumulated with various types of plutonium bearing fuel elements. Other work which is relevant, but which could not be dealt with here, is the programme of long term reactivity change measurements related to the Calder reactors and the neutron spectra measurements made by chopper techniques with the PANTHER homogeneous plutonium assembly at Dounreay, with Zenith at Winfrith and on various assemblies at Harwell.

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1. D. C. Leslie. "The Spectrox Method for Thermal Spectra in Lattice Cells", AEEW M.211. 1962.
2. D. C. Leslie. "The Calculation of Removal Cross Sections Between Overlapping Thermal Groups", AEEW R.133. 1962.

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