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LIQUID SODIUM METAL COOLED FAST BREEDER REACTOR (LMFBR) PROGRAM
FUELS AND MATERIALS SUMMARY REPORT

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INTRODUCTION

Early in 1965, the AEC Division of Reactor Development and Technology (RDT) assembled a task force from among qualified AEC staff members to review previously generated fuels and materials data and current fuels and materials development activities pertinent to the LMFBR program, with emphasis on the U-Pu fuel cycle. This action was taken, as part of an overall LMFBR program review, to assess the adequacy of existing development activities, to obtain needed fuels and materials data for the LMFBR program on a timely basis, and to identify areas wherein program strengthening and/or reorientation might be required.

The task force undertook an intensive review of the literature, made numerous visits and held many discussions with scientists and engineers (from both AEC and industrial laboratories) associated with current development activities and/or associated with facilities of known or potential use in such activities. In mid-1965, a summary report of the task force findings and recommendations was submitted to several AEC field offices and laboratories for critical comment, and such comments have been received. Additionally, technical discussions on specific areas of the summary report were held concurrently with industrial contractors.

In mid-1965, the AEC also assembled a team of industrial, AEC laboratory and AEC fuels and materials specialists to visit the United Kingdom, France and Germany and, thereby, to become updated on the information available from the fast reactor fuels and materials development activities in those countries. Similarly, an AEC sponsored group of technical specialists previously visited the USSR to review the Soviet reactor program.

The input from the above mentioned actions has produced extensive documentation in the form of technical reports and evaluations, trip reports, minutes of meetings, and comments received from AEC laboratories and industrial contractors. All of these have been used in preparing this summary report of RDT conclusions and recommendations with respect to the LMFBR fuels and materials development program. It must be emphasized that these conclusions and recommendations are confined to the development program in the U.S., although data generated abroad (and expected to be generated in development programs abroad) have been considered in assessing the status of technology and future program needs. Additionally, although a considerable amount of foreign data has been made available to the U.S. under existing exchange programs, much of such data is preliminary and therefore is not generally included in the summary report.

ABBREVIATIONS USED IN TEXT

AEC	U. S. Atomic Energy Commission
ANL	Argonne National Laboratory
ATR	Advanced Test Reactor (Phillips Petroleum, Idaho)
CP-5	Chicago Pile-5 (ANL)
EBOR	Experimental Beryllium Oxide Reactor (NRTS)
EBR	Experimental Breeder Reactor (ANL)
EGCR	Experimental Gas Cooled Reactor (ORNL)
EOCR	Experimental Organic Cooled Reactor (NRTS)
ETR	Engineering Test Reactor (Phillips Petroleum, Idaho)
FCF	Fuel Cycle Facility (ANL-NRTS)
Fermi	Enrico Fermi Atomic Power Plant
FFTF	Proposed Fast Flux Test Facility (Battelle Pacific Northwest)
GETR	General Electric Test Reactor (Pleasanton, California)
LMFBR	Liquid Sodium Metal Cooled Fast Breeder Reactor
LTR	Lynchburg Test Reactor (Babcock and Wilcox)
MTR	Materials Testing Reactor (Phillips Petroleum, Idaho)
NRTS	National Reactor Testing Station
ORNL	Oak Ridge National Laboratory
ORR	Oak Ridge Reactor
SEFOR	Southwest Experimental Fast Oxide Reactor Southwest Atomic Energy Associates, AEC, Euratom, Gesellschaft Fur Kernforschung (Karlsruhe, Germany)
SR	Savannah River Reactors

SRE Sodium Reactor Experiment (Atomics International)
SRE-PEP Sodium Reactor Experiment - Power Expansion Program
TREAT Transient Reactor Test (NRTS)
UKAEA United Kingdom Atomic Energy Authority

ABSTRACT

As part of an AEC assessment of the liquid sodium metal cooled fast breeder reactor (LMFBR) program, the status of pertinent fuels and materials technology was reviewed and evaluated during 1965. This assessment was accomplished with full consideration of inputs by cognizant AEC laboratories and industrial contractors and involved visits abroad to the U.K., France, and Germany and results of previous visits to the USSR.

A major finding of the study was the paucity of data on properties and performance of LMFBR fuels and materials particularly in a fast flux environment, thus making it difficult to establish meaningful criteria and standards for these materials. It was uniformly acknowledged among the laboratories and the industrial contractors that a definite need exists for investigations of fuels and materials performance limitations in order to provide a basis for establishing meaningful criteria for their selection and use.

Bearing this in mind, the following were the principal conclusions of the review:

- a. Two metal fuel alloys, (U-Pu)Zr and (U-Pu)Ti, offer promise for improved performance over the previously investigated (U-Pu)Fs/Fz systems, which exhibit excessive swelling at low burnups and at relatively low operating temperatures. Work on the Zr and Ti alloys should continue until sufficient data have been collected to evaluate their potential in comparison with other candidate fuels for the LMFBR program.
- b. Ceramic fuels, primarily the uranium-plutonium oxides and carbides in that order, currently offer the best potential for safe and reliable fuels capable of achieving high burnup while operating at high sodium-metal-coolant temperatures. These fuels are currently favored over all others by U.S. industry and in all major foreign fast reactor programs. Existing irradiation data, from both domestic and foreign programs, are sufficiently favorable to warrant a considerably increased emphasis in developing and testing these fuels for LMFBR application. Uranium-plutonium nitride also looks promising, but further basic work on the performance of nitride fuel is needed and should be done to evaluate its potential.

c. The LMFBR cladding development program has been hampered by the previous lack of fast flux irradiation facilities in the U.S. Cladding alloy development should be augmented significantly, with primary emphasis on austenitic stainless steel alloys, and backup investigations on higher strength nickel alloys and refractory alloys. The study of fast neutron damage mechanisms and their effect on the properties of cladding materials must be emphasized in the augmented program. Another important area requiring further investigation is that of fuel-cladding and sodium coolant compatibility.

d. Existing thermal reactor test facilities will be sufficient for screening and comparative evaluation tests of fast reactor fuels. On the other hand, expected differences between thermal and fast neutron effects on both fuel and cladding materials are such that only fast flux irradiations can be depended upon to provide data for final selection of LMFBR fuel and cladding combinations. In this context, EBR-II and Fermi will have to be used to the maximum extent possible for fast flux test irradiations, inasmuch as they will be the only available fast reactors in the U.S. for a number of years to come.

e. The existing fast reactors (EBR-II and Fermi) have limitations as to neutron flux, temperature, environmental control, instrumentation, and amount of test space and other test conditions achievable. Accordingly, the design and construction of an adequate fast flux test facility to circumvent these limitations and to significantly improve accelerated testing capabilities is recommended on a high priority basis for the successful development of economical and reliable fast breeder fuels and materials.

METAL ALLOY FUELS

Summary and Conclusions

The uranium-plutonium fizzle (or fissium)* alloy fuels do not appear promising for LMFBR application because of excessive irradiation-induced swelling at low burnup and at relatively low operating temperatures and because of compatibility problems with iron and nickel base cladding alloys. Alloy development work is now focused upon (U-Pu)Ti and (U-Pu)Zr alloys, which show promise for improved behavior over the previously investigated metal fuel alloys. Development of the Ti and Zr alloys should continue until there is sufficient data to indicate whether or not they can compete with other fuels in the LMFBR program with respect to safety, reliability, and plant economics. There are currently no other uranium or plutonium-uranium metal fuel alloys under consideration in the U.S. for possible near term LMFBR application, nor is there currently any industrial interest expressed for the further development of these fuels.

Introduction

Historically, fast breeder reactor development in the U.S. began effectively at the Argonne National Laboratory (ANL) about 1947, with development activities leading to the world's first experimental breeder reactor, EBR-I, first operated in 1951 with uranium metal fuel. Since that time, much attention has been given to the development of metal alloy fuels as fast breeder reactor fuels. This attention has been well justified by the potential advantages of metal alloy fuels which include: (i) high fissile atom density which is conducive to high breeding ratios, (ii) high thermal conductivity, (iii) thermal expansion contributing to a negative reactivity--temperature coefficient, and (iv) relative ease of fabrication conducive to relatively simple fuel cycles and to good fuel cycle economics. Unfortunately, metal alloy fuels have thus far proved to be limited in the temperatures at which they can be operated and the burnups they can sustain without undergoing gross dimensional changes from irradiation-induced swelling.

*Fissium(Fs) is a combination of the five metallic elements: Zr, Mo, Ru, Pd and Rh, not removed in the present pyroprocessing system associated with EBR-II. Fizzle (Fz) is also a combination of the same five elements, but of a slightly different composition, principally a higher Zr content.

Uranium Alloys

The most extensive development effort on uranium alloy fuels at ANL has been on the U-5 Fs (weight percent) system, which is the present fuel (with 304 SS cladding) for the EBR-II. This fuel has operated successfully in EBR-II to date, achieving a burnup of 1.2 a/o. This probably represents a practical upper limit in this application for the present fuel design.

Uranium-molybdenum alloys have been selected as fuels for two fast reactors; the Enrico Fermi Reactor, U-10Mo (weight percent) and the U.K. Dounreay Fast Reactor, initially U-9Mo and now U-7Mo. Initial capsule investigations on U-10Mo indicated relatively good dimensional stability to burnups on the order of 2 a/o at fuel temperatures up to 600°C (1112°F). However, more recent experiments suggest that the fission rate to which the alloys are subjected, as well as temperature, determines the alloys' resistance to swelling. U-10Mo is found to be unstable when irradiated at fission rates below 6×10^{13} fission/cm³-sec at temperatures of 480 to 570°C (896 to 1058°F). At temperatures up to 460°C (860°F), fission rates of 3.6×10^{13} fission/cm³-sec are considered necessary in order for the fuel to be stable up to about 1 a/o burnup.

The instability of U-10Mo under irradiation, attributed to an irradiation-induced phase reversion, has prompted reduced temperature and power level operation of the Fermi reactor. As the reactor has operated only at power levels below one MWt and for short periods, present burnup of the Fermi fuel is negligible. Goals on the order of 0.8 a/o in the center of the core and 2.0 a/o around the edge have been established.

Performance of the U-Mo fuel in the Dounreay reactor has been satisfactory but not without difficulty. In addition to expected fuel swelling, cracking has been found in some of the examined fuel and, in certain of the higher burnup elements examined, evidence of sodium bond failure and localized melting of the U-Mo alloy has been observed. The major problem in Dounreay has been associated with corrosion of the niobium cladding material by impurities in the coolant. To reduce the corrosion, cladding temperatures have been lowered to 480°C (896°F), thus reducing the operating power levels of the reactor. Burnups in Dounreay have attained over 3 a/o of the heavy atoms by using the lower temperatures, providing void space, and changing Mo content to 7 weight percent.

The French have also performed considerable experimentation on metal fuels, including work on U-Pu-Mo, U-Pu-Ti and U-Pu-Zr. Although originally proposed as the reference fuel for Rapsodie, U-Pu-Mo has now been abandoned because of poor irradiation stability in favor of UO₂-PuO₂.

Uranium-Plutonium Alloys

The (U-Pu)Fz/Fs alloy systems have been under intensive development for many years at ANL. In many respects, however, the behavior of these alloys has proved to be disappointing. There are compatibility problems with iron and nickel base cladding alloys because of low melting eutectics between Pu and Fe, and Pu and Ni. Irradiation-induced swelling of the Fs/Fz alloys becomes excessive at temperatures above about 370°C (698°F). For these reasons, a considerable amount of work has been done to find a cladding material which will be compatible with the (U-Pu)Fz/Fs fuel as well as with the sodium coolant, and which will be capable of restraining the fuel as it swells under irradiation. Additionally, other alloy systems have been explored in search of improved performance.

In seeking a U-Pu alloy fuel with improved irradiation performance, ANL has investigated a number of alloy compositions involving U-Pu with additions of 10 a/o and 30 a/o of each of the following elements: Sc, Ti, Y, Zr, Mo, Pd, and Th. Melting temperature was selected for screening the alloys based on the hypothesis that a higher melting temperature should lead to improved irradiation swelling resistance and/or a higher temperature operating capability. From among the alloys screened the 30 a/o Ti and 30 a/o Zr alloys have emerged as the most promising.

Physical Properties and Characteristics

The following paragraphs list some properties and characteristics of the uranium-plutonium alloys for which a comparison can be made between the (U-Pu)Fz (Fs) alloys and the (U-Pu)Ti or Zr alloys. It will be apparent that relatively little information has been obtained to date on the more recent (U-Pu)Ti and (U-Pu)Zr alloys.

Mechanical Properties. Alloys of U-Pu-Fz (Fs) display mechanical properties which change rapidly in the 400-500°C (752-932°F) temperature range. The alloys are extremely brittle and strong at room temperature. As temperature is increased, the alloys lose their strength and increase their ductility.

The critical irradiation swelling temperature of approximately 400°C (752°F) corresponds to the temperature at which the alloys display drastic strength losses. Perhaps by increasing the elevated temperature strength of alloy fuels, the temperature at which swelling occurs under irradiation will be raised.

Preliminary mechanical property data on the U-Pu-Ti and U-Pu-Zr alloys show pronounced strength advantages over the Fz (Fs) alloys at elevated temperatures. As an example, U-20Pu-10Fz has a strength of about 3 kg/mm² (4,260 psi) at 618°C; the U-17Pu-12Ti alloy displays a strength of 34.3 kg/mm² (48,500 psi) at the same temperature.

Melting Point. The solidus temperature is between 820°C (1508°F) and 910 C (1670 F) for the U-Pu-Fz (Fs) alloys, depending upon Pu content. Ti and Zr alloy additions raise the solidus to around 1100°C (2012°F).

Compatibility with Clad. Iron and nickel base alloys have been eliminated for use as cladding with Fs/Fz alloys because of low melting eutectics between Pu-Fe 410°C (770°F) and Pu-Ni 465°C (869°F). Other clads which have been investigated as replacements include V-20Ti, Nb-12Zr, Mo-0.5Ti-0.08Zr, and duplex tubing. Of these candidates V-20Ti appears to have the best compatibility with the U-Pu-Fz (Fs) fuel. Tests to date indicate that the (U-Pu)Ti and (U-Pu)Zr alloys are compatible with V-20Ti. (U-Pu)Ti has been observed to react with 304 SS in tests conducted between 500 and 650°C, but (U-Pu) Zr appears to be compatible with SS at temperatures up to 800°C.

Alloy Fabrication

Injection casting is the technique developed and used by ANL for fabricating U-Pu alloy pins. Equipment required for the injection casting operation is minimal; namely, an induction furnace fitted with a zirconia-lined graphite crucible, capable of pressurization. Use of this casting technique permits the U-Pu fuel alloy to be taken from the pyrorefined cast ingot produced to a semi-finished fuel pin in a single operation.

The injection casting operation was initially developed by ANL on a laboratory basis, has gone through production development as the process used for fabrication of EBR-II, Core I and, because of its inherent simplicity, has recently been adapted to remote operation in the Fuel Cycle Facility (FCF) associated with the EBR-II reactor.

The refabrication station in the FCF receives the pyrorefined and cast ingot from the refining process and induction melts the ingot in a zirconia-lined graphite crucible under a slight pressure of helium. A number of Vycor molds are held in a position above the melt and, when the temperature of both molds and melt are proper for casting, the furnace and molds are evacuated; the open end of the molds are then submerged to near the bottom of the crucible containing the molten fuel alloy. After submersion, the furnace is pressurized

rapidly with helium to two atmospheres, which drives the molten metal into the Vycor molds. Up to 160 fuel pins, 0.144 inch in diameter by 18 inches long, have been cast in a single operation using the injection casting techniques.

For fabrication of metal fuels into small diameter pin geometries, the injection casting process is unquestionably one of the simplest of all fabrication processes. The process eliminates the need for the more conventional (and expensive) metal fabrication equipment, such as rolling mills, extrusion presses, forges, and, because of its simplicity, is ideally suited to remote operation.

Irradiation Behavior

All irradiations tests to date have been performed in thermal reactors (i.e., CP-5 at ANL and the MTR in Idaho), although tests are now underway in EBR-II. From the many thermal reactor tests available, it is clear that the (U-Pu) Fz/Fs alloys swell excessively at irradiation temperatures above about 370°C (698°F at fuel surface). The following numbers are a few data points obtained for unrestrained U-20Pu-10Fs alloy, for example:

<u>Max. Fuel Temp., °C</u>		<u>Burnup a/o</u>	<u>% Δ V a/o Burnup</u>
<u>Center</u>	<u>Surface</u>		
240	230	1.0	3.9
290	270	1.2	2.8
310	290	1.4	6.9
310	290	1.4	2.7
320	240	2.3	24
330	310	1.4	7.9
370	350	1.2	50
370	350	1.7	6.4
390	370	1.8	22
440	420	1.3	59
470	440	1.3	69
510	370	3.0	22

In spite of the extreme swelling associated with unrestrained fuel under irradiation, high burnups at relatively high temperatures have been achieved in thermal irradiations through the use of strong, compatible cladding and the incorporation of void volumes in the fuel pins to accommodate fuel swelling. Over 8 a/o burnup has been achieved, for example, with several samples of U-15Pu-10Fz clad in 20 mil Nb-12Zr alloy, operating in the CP-5 at 545°C maximum jacket temperature and 625°C maximum fuel temperature.

Only interim, non-destructive examination data is available to date for the (U-Pu)Ti and Zr alloys, although specimens in the CP-5 reactor have accumulated over 4 a/o burnup and testing is continuing. Alloy samples are also being irradiated in the EBR-II.

URANIUM-PLUTONIUM OXIDES

Summary and Conclusions

The uranium-plutonium oxides, which are at the most advanced state of development among the ceramic fuels, currently offer the best near-term potential for safe, reliable fuel, capable of achieving very high burnups while operating in sodium at high temperatures. The development of mixed oxides has received primary interest and support from industry and major foreign programs. Testing, evaluation, and analysis of the $\text{UO}_2\text{-PuO}_2$ (about 20 w/o Pu) system specifically directed towards fast reactor application in the U.S. have been underway since 1959. Thermal reactor irradiation testing of oxide fuel specimens up to approximately 100,000 megawatt days per ton of uranium-plutonium (MWD/T) in the U.S. has demonstrated irradiation stability and performance capability in a thermal flux. At the time of this survey, about 85,000 MWD/T had been achieved in one mixed oxide fuel pin in the U.K. Dounreay Fast Reactor, and also about 60,000 MWD/T in one 77-pin subassembly.

The major advantages of the mixed oxide fuel are good compatibility with stainless steel clad, excellent thermal irradiation stability, high melting temperature (4900°F), and relatively advanced synthesis and fabrication technology. The known or suspected disadvantages are non-reproducible thermal expansion characteristics, low thermal conductivity, fuel slumping at or near melting, and fuel ratcheting from thermal cycling. The significant potential fuel performance problem areas are fast flux irradiation effects, sodium-fuel compatibility, cladding integrity, fuel redistribution, plutonium migration, and concentration and transient effects.

It is concluded that uranium-plutonium oxides should be a prime fuel candidate for the LMFBR program and that the current development work should be augmented appreciably. It is recommended particularly that there be increased efforts in ascertaining the behavior and characteristics of mixed oxides in fast flux irradiations, and in improving fuel characteristics such as thermal conductivity and control of fuel redistribution. Additionally, meaningful fuel criteria and standards must be established by which fuel performance and potential will be judged and evaluated.

Mixed Oxide Composition

The procedures and equipment for compositional control and analysis are currently at an intermediate development stage. Techniques exist for determination of many of the known critical constituents in the fuel material and other parameters, such as density and O/M (oxygen to metal ratio). Most procedures, however, require destruction of the sample analyzed and involve relatively long, expensive analytical procedures. The organizations most directly involved with the mixed oxides are set up to perform laboratory scale analytical techniques. The parameters involved in obtaining proper synthesis and fabrication control appear to be relatively well understood for small batch operations and seem to offer no unique problems other than Pu containment.

The effect of initial fuel impurities on performance is generally unknown. Metallic impurities are generally kept at about 400 to 1000 parts per million initially. Total fuel impurity specifications are currently based on neutron absorption considerations rather than fuel fabrication or performance criteria.

Attention should be given to development of rapid, continuous, and non-destructive analytical techniques for prototype fuel batches. Controlled irradiation experiments are needed to determine what impurity levels affect fuel performance and to ascertain whether inadequate or unnecessary restrictions for impurity levels are currently imposed.

Mixed Oxide Characteristics

Density

The theoretical fuel density of the mixed oxides is low compared to the ceramic grade carbides and nitrides (11.08 g/cc vs. approximately 13.2 g/cc). Current fuel irradiations have employed up to 95% T.D. (theoretical density) pellets with the effective smeared bulk density controlled by varying radial gaps.

Based on UO_2 and $\text{UO}_2\text{-PuO}_2$ irradiations, the following trends are indicated.

- a. The fuel thermal conductivity increases with increased density.
- b. Fission gas release increases with decreased densities.
- c. Fuel swelling increases with increased densities due to fission product build-up.

d. Fuel particle densities increase to near theoretical values at temperatures near the fuel melting point.

e. Large (about 10%) volume increases occur when the mixed oxide fuel melts.

Thermal Expansion

The thermal expansion coefficient for mixed oxides appears not too different from the carbide and nitride ceramics. Due to the high oxide fuel central operating temperatures, which is in turn due to the low thermal conductivity of the fuel, allowance for fuel expansion must be designed into the oxide fuel elements.

Phase Characteristics

A Na-U-Pu-O compound (positive identification has not been made) has been noted in several experiments where sodium coolant ingress has occurred. Evidence is that this material has a low thermal conductivity. Long-term, controlled compatibility tests have not been performed.

Metallic agglomerates have been observed in fuel operated at high temperatures. The metallic sites appear to be metallic oxide fission product concentration sites. It is important to determine if these areas are plutonium metal concentration sites.

Based on existing thermal irradiations, the UO_2 - PuO_2 system appears to be chemically stable at temperatures of interest (below 4900°F) with no apparent detrimental phases reported. Fission product metals, primarily molybdenum, have been identified in trace quantities within the fuel matrix.

Phase diagrams of the UO_2 - 20PuO_2 (hyper, hypo, and stoichiometric) system, up to melting temperatures, should be determined.

Thermal Conductivity

The thermal conductivity of the mixed oxides is low compared to the carbide, nitride, and metallic fuels at all temperatures. Below 3000°F and in-pile, the thermal conductivity of UO_2 - 20PuO_2 is lower than that for UO_2 ; above 3000°F and to 4000°F , the thermal conductivity of the two oxide systems is nearly equal.

The PuO_2 content may affect the thermal conductivity at the low temperature range. The Battelle Pacific Northwest Laboratory reports that UO_2 and UO_2 -1 to 5 PuO_2 vibratory compacted powders exhibit near equal thermal conductivities in-pile. Other in-pile pellet experiments in the GETR indicate a UO_2 -20 PuO_2 thermal conductivity value 20% and 5% lower than UO_2 pellets at 1500°F and 3200°F, respectively.

Sodium-mixed oxide fuel compatibility, short-term, in-pile tests have shown that the effective thermal conductivity is improved by the presence of sodium in the fuel cracks and fuel-clad gap. However, conflicting evidence from other experiments indicate a poor thermal conductivity phase may be formed. Initial hypostoichiometric UO_2 -20 PuO_2 has a higher thermal conductivity than hyper-material. The effect of irradiation is not yet determined.

Melting Point

The out-of-pile melting points of coprecipitated UO_2 - PuO_2 of various compositions have been measured. The following melting points and compositions of interest include: UO_2 -10 PuO_2 : 2820°C (5108°F); UO_2 -20 PuO_2 : 2780°C (5036°F) and UO_2 -40 PuO_2 : 2700°C (4892°F).

Stoichiometry effects (O/M between 1.97 and 2.06) on the melting point have been determined with the hypostoichiometric material having a higher melting point than hyper-material. The change of melting point with irradiation exposure has not been determined. UO_2 data is currently used for guidance.

Thermal Stability

Thermal cycling of mixed oxides result in pellet cracking (both radial and circumferential). Increased thermal resistance may occur due, among other things, to the interrupted heat transfer path. At temperatures below 3000°F, fuel cracking is minimal.

Cladding Compatibility

With an initial helium gas bond between the fuel and SS-clad, excellent chemical compatibility has been demonstrated at high burnups.

General Conclusions

Some of the major development program needs for determining product characteristics are as follows:

- a. Fuel property changes as a function of irradiation.
- b. Fuel and fuel element design parameters as related to fuel performance. Items of importance include: density, fuel-clad gap, pellet surface condition, initial fuel impurities, stoichiometry, thermal cycle effects, slumping, and fabrication processes.
- c. Fuel-to-sodium and fuel-to-clad compatibility tests.
- d. Clad evaluation and development program.
- e. Fuel performance and characteristic optimization, such as thermal conductivity improvement.

Product Fabrication

Only laboratory size quantities of $\text{UO}_2\text{-20PuO}_2$ have been produced on a batch basis. Three routes of fabrication for mixed oxides are currently employed: coprecipitation, impaction of mechanically mixed, and mechanically mixing of UO_2 and PuO_2 powders. The product is ground and sized and either (1) pressed into pellets which are sintered and ground to size, or (2) vibratory packed into clad tubing. Homogeneity of the plutonium in the fuel matrix has been a major concern but recent data indicates good homogeneity resulting from any of the three methods. High pellet densities of 94 to 97% T.D. are routinely achieved. Vibratory packed densities achieve 88% T.D.

The mode of fabrication of the mixed oxides appears to be somewhat dependent on the respective fabricator's method and convenience of handling the material. Currently needed in the development program are evaluations of controlled irradiations of oxides prepared by all three methods, using controllable processes. Properly designed tests will provide means to evaluate fuel performance on a consistent basis.

Mixed Oxide Irradiations

The major portion of the mixed-oxide fuel irradiations in the U.S. has been performed in the GETR Trail Cable Facility (TCF) and the RAFT positions. Five mixed oxide capsules, plus UO_2 reference capsules, have been transient tested in the TREAT facility.

The typical flux conditions for an average GETR cycle in the TCF and RAFT positions are 8×10^{13} nv thermal and 1 to 2×10^{13} nv fast (>1 Mev). All specimens are encapsulated and tested with a static Na or NaK bond between the clad and the outer capsule. The fuel specimens have varied in length from 1.5 inches to about 6 inches.

In some instances, several small fuel specimens are combined into one capsule. This approach has been unsatisfactory because the failure in one fuel specimen has resulted in the failure of all specimens. Specimen power is controlled by sample relocation as the reactor flux profile changes.

Since the 1960-1961 irradiations of the 40 fuel specimens which achieved up to 99,000 MWD/T (calculated), no fuel irradiations have achieved over 70,000 MWD/T exposure, and failures have occurred in most instances. Failures are attributed to capsule design. Since peak exposures of about 140,000 MWD/T will occur in a fast reactor it is essential that well controlled and characterized experiments be initiated. Irradiations have been performed with the interest of measuring many parameters in one capsule. During examination, it is usually difficult to separate the variables and their specific contribution to overall performance. Future irradiation experiments should be designed to assure that the cause and effect are separable. A standard capsule design is needed.

Mixed oxide fuel irradiations are underway in the Dounreay Fast Reactor. The irradiation parameters include both pellets and powder fuel and several cladding materials to fuel burnups in excess of 5 a/o. Considerable fuel technology information on mixed oxides is generated and exchanged between the U. K. and the U.S.

URANIUM-PLUTONIUM CARBIDE FUELS

Summary and Conclusions

Uranium-plutonium carbide fuel compositions have favorable thermal conductivity, high fissionable atom density, and high melting points. They have good potential as candidate fuels for the LMFBR application.

The main problem with UC-PuC fuels is clad performance in the presence of fuel swelling and released fission gas, and clad compatibility with the fuel. Experience obtained from irradiation tests in thermal neutron fluxes indicates that a strong ductile clad with a helium gap between fuel and clad may restrain fuel swelling. Alternatively, the bond spacing may be adjusted to accommodate swelling without straining the clad; if a sodium bond is to be used, a cladding material compatible with carbide fuel needs to be found.

The present status of fuel characterizations is limited to a few selected compositions. Variables in synthesis and fabrication of fuel require development. The effects of composition, microstructure, carbon stoichiometry, and density on specific heat, melting point, vapor pressure, thermal conductivity, thermal expansion, strength, and hardness are relatively unknown.

Correlations need to be established between carbide fuel characteristics and fast flux irradiation in a high temperature sodium environment at varying degrees of burnup, including clad surface temperature, fuel center temperature, linear power, and burnup. It is necessary to know the effects of swelling, fission gas release, plutonium distribution, and fission product distribution.

Fabrication of Uranium-Plutonium Carbide

Plutonium-uranium carbide fuel specimens have been prepared for irradiation testing in the forms of sintered pellets, vibratory compacted powder, and arc cast pins. Both arc cast and sintered pellet solid solution and vibratory compacted mixed carbide fuel specimens have been irradiation tested. Cladding materials used in the irradiation tests were 20Cr-20Ni stainless steel, Nb-12Zr, type 304 stainless steel, Nb, and type 316 stainless steel. The fuel to clad bond was either argon or helium.

Uranium-Plutonium Carbide Irradiation Behavior

Irradiation of (U-Pu)C with Helium Gap

The highest average burnup attained at the time of this program review was 113,000 MWD/T in a thermal neutron flux, with 0.191 inch diameter sintered UC-20PuC fuel, using a 0.001 inch radial helium gap, a 0.030 inch wall thickness, and Nb clad. The average linear power generation was 12 kw/ft, and the average clad surface and fuel center temperatures were 555 and 925°C (1030 and 1700°F), respectively. The fuel swelling was 14 volume per cent, and the fission gas released was 30 per cent. The Nb clad remained intact but exhibited a 2 to 3 per cent increase in diameter.

The next highest average burnup reached was 101,000 MWD/T, in a thermal neutron flux also with 0.191 inch diameter sintered UC-20PuC fuel and 0.001 inch radial helium gap. The average linear power generation was 12 kw/ft and the clad surface and fuel center temperatures were 575 and 1030°C (1070 and 1885°F), respectively. The clad, type 316 stainless steel, was cracked in the longitudinal direction. The carbide in these specimens had a hyperstoichiometric structure.

A study is presently being conducted of pressed and sintered pellets contact bonded to the jacket and vibratory-compacted powder in fuel tubes. In one instance, the vibratory-compacted UC-20PuC fuel has been irradiated with negligible volume change to 6.8 a/o burnup (18×10^{20} fissions/cc) in thin (0.009 inch) Nb-1Zr alloy cladding. Clad surface temperature was 640°C. Under the same conditions, 304 stainless steel cladding showed some deformation but did not rupture.

Mixed carbide fuel irradiations are underway in the Dounreay Fast Reactor. The irradiation parameters include both pellets and powder fuel and several cladding materials to fuel burnup in excess of 5 a/o. Considerable fuel technology information on mixed carbides is generated and exchanged between the U.K. and the U.S.

Irradiation with Sodium Bond

Experience with carbide fuel sodium bonded to the clad is limited to cast or sintered uranium carbide. While there is no assurance that irradiation-effects information on the uranium carbide applies to behavior of the mixed carbides under similar conditions, some UC work is included in this discussion to illustrate the observed behavior.

Highest average burnups up to 40,000 MWD/T have been accomplished in a thermal neutron flux with 3/8-inch diameter fuel and 0.030 inch wall thickness Nb-1Zr clad. The sodium bond radial thickness was 0.020 inch. Stoichiometric, hypostoichiometric and hyperstoichiometric uranium carbide specimens were irradiated. Linear power generations ranged from 27 to 34 kw/ft. Average surface and center temperatures were 685 to 725°C and 1000 to 1120°C, respectively. The clad was intact and exhibited no dimension changes.

Cast stoichiometric or hypostoichiometric uranium carbide specimens sodium bonded to type 304 stainless steel clad of 0.010 inch thickness have been irradiated to 20,000 MWD/T in a thermal neutron flux. The 0.495 inch diameter fuel slugs had a 0.026 inch sodium bond thickness. Linear power generations were 30 to 38 kw/ft. Average surface and center temperatures ranged from 350 to 500°C (660 and 930°F) and from 615 to 870°C (1140 to 1600°F), respectively. The clad was intact and exhibited no dimensional changes.

Cast hyperstoichiometric 0.495 inch diameter UC fuel slugs clad in 0.010 inch wall thickness type 304 stainless steel with a 0.026 inch radial NaK gap were irradiated to an average burnup of 23,900 MWD/T in a thermal neutron flux. The average surface temperature was 295°C (560°F) and the average center temperature was 480°C (900°F). The clad was carburized and cracked.

General Conclusions

The fuel development program should be augmented to determine plutonium-uranium carbide fuel behavior with various clad materials and bonds. The augmented program should include study of variables such as fuel synthesis and fabrication methods, fuel-clad gap dimensions and sodium or helium within this gap. It is most important that fuel swelling and fission gas release as a function of fuel temperature and burnup be established in order that a proper foundation exists for the fuel designs utilizing the high priority fast flux testing space.

The fuel development program should include the following areas:

- a. Synthesis and fabrication.
- b. Out-of-pile properties measurement.
- c. Irradiation testing (thermal flux where appropriate and fast flux where necessary), including transient testing.
- d. Post-irradiation examinations, including irradiated fuel properties.
- e. Establishment of meaningful criteria by which to judge and evaluate fuel performance.

URANIUM-PLUTONIUM NITRIDE FUELS

Summary and Conclusions

The irradiation testing of UN-PuN to date has been minor, and the existing knowledge of the mixed uranium-plutonium nitride properties and characteristics is very limited. The available data indicate, however, that nitrides should have a higher plutonium and theoretical fuel density (≈ 14.3 gm/cc) and a higher melting point ($\approx 2840^\circ\text{C}$) than either the mixed oxides or carbides. The mixed nitride compounds have a thermal conductivity equal to that of the carbides (at 400°C , $k_{\text{Th}} = 0.18$ watts/cm- $^\circ\text{C}$). In addition to the above properties, the nitrides possess the potential, as evidenced by tests with UN and PuN, individually, of excellent compatibility with candidate cladding materials (stainless steels, nickel alloys and refractory alloys) and sodium coolant.

Based upon such preliminary information, it appears that uranium-plutonium nitrides should be given increased consideration in the LMFBR program. Development work should initially concentrate on screening tests to establish the characteristics and behavior of the fuel under operating conditions and to determine to what extent, if any, fuel dissociation and helium gas production (through the N^{14} (n, α) reaction) will impose limitations on the use of such fuel.

FAST FLUX TEST FACILITY DRIVER FUEL

As indicated in a previous section of this report, U-5Fs has had a significant role in the early metal alloy fuel development and is now serving as the EBR-II driver fuel. It is clear, however, that the U-5Fs has no prospective role in the LMFBR program other than its current one. Accordingly, considerable attention should be given to determining the value and feasibility of using other more promising LMFBR fuels in place of some or all of the U-5Fs driver fuel. To the extent that this might be accomplished, the quantity of pertinent test results generated in EBR-II would be increased.

Similarly, a 20 v/o PuO₂-stainless steel cermet has been proposed as a driver fuel for a Fast Flux Test Facility (FFTF) primarily on the basis of the large, reproducible, temperature-induced expansion characteristics of this type of fuel and the potential contribution of such expansion to nuclear safety. In a complex test reactor such as the FFTF, a cermet may prove to be essential for the reason stated. This type of cermet is not, however, a candidate fuel for LMFBR's. Accordingly, careful attention must be given throughout the continuing FFTF design studies to the possibility that an LMFBR fuel candidate, such as a ceramic, will satisfactorily perform as a driver fuel in lieu of the cermet. If such were the case, then the development involved for the driver fuel would be more consistent with, and would contribute more directly to, the development of LMFBR candidate fuels.

OTHER FUELS

Fuels such as uranium-plutonium sulphides and uranium-plutonium phosphides, on preliminary examination, offer potential for the LMFBR program. Fuels such as uranium carbide-plutonium nitride and uranium carbide-plutonium sulphide appear promising, although data is limited and irradiations virtually non-existent. The present level of effort for these and other fuels should continue.

It is recognized that other fuel concepts such as pastes and liquid plutonium alloys are potential LMFBR fuel candidates. For the purpose of this report, however, only solid fuels were considered as having a near-term potential.

The use of vented fuels is recognized as a possible need in the LMFBR program. It is recommended that such a fuel concept be explored as part of the LMFBR fuels and materials development program.

FUEL CLADDING MATERIALS

Summary and Conclusions

With respect to LMFBR fuel cladding materials, there is currently more pertinent information available for the stainless steel alloys than for any of the other materials that are presently being considered as potential cladding candidates. Continued emphasis on stainless steels in the clad materials development program appears well justified by the performance observed thus far in high temperature (i.e., up to 1200°F) sodium, by the apparently good compatibility with uranium-plutonium oxide fuel, and by its relatively low cost among the candidate materials. Much more data is needed for the stainless steels in this application, particularly in respect to compatibility with other ceramic fuels and long-term mechanical properties in the fast neutron environments of fast reactor cores.

The successful use of stainless steel as an LMFBR fuel cladding material is by no means assured at this point in time. There is a continuing need to investigate other potential cladding materials, such as the higher strength nickel alloys and refractory alloys, and there is a general need for more information on their (a) compatibility with LMFBR fuel materials, (b) compatibility with sodium, and (c) long-term mechanical properties, both out-of-pile and in-pile. It is recommended that the acquisition of these types of data be accelerated for these materials.

There is a dearth of data on the effect on materials properties of the large, fast neutron doses expected in future fast breeder reactors. Several competing neutron damage mechanisms can be foreseen which may have deleterious effects on the iron and nickel base alloys and, to a lesser extent, on other alloys. The resolution of these questions should have the highest priority among development activities, recognizing that existing fast reactors require three years or more to accumulate the needed fast neutron doses.

Stainless Steel Alloys

Stainless steel alloy technology is in the most advanced stage of development relative to other potential LMFBR cladding materials. The indicated performance with uranium-plutonium oxide fuel in 1200°F sodium coolant is good when the impurity content of the sodium, such as oxygen and carbon, is maintained at low levels.

The following commercial stainless steels offer development potential for the reasons indicated.

a. 304 is a good basic austenitic stainless steel composition which appears to be the first choice by U.S. industry as a structural material for 1200°F (649°C) sodium systems. It has received the greatest attention in the U.S. program on irradiation effects on stainless steels. For example, the data to date of in-reactor creep of annealed 304 stainless steel show that within the scatter of experimental data there is no effect during irradiation of the neutron irradiation on the minimum creep rate. There are some differences, however, between in-reactor and out-of-reactor creep characteristics. Of greatest significance is the reduced creep-to-rupture life during neutron irradiation, as illustrated in the following data:

<u>Temperature,</u> <u>°C</u>	<u>Stress</u> <u>psi</u>	<u>% Creep Strain @ Failure</u>		<u>Failure Time, hour</u>	
		<u>In-Reactor</u>	<u>Ex-Reactor</u>	<u>In-Reactor</u>	<u>Ex-Reactor</u>
650	20,000	8 to 9	25 to 26	150	550
550	30,000	3 to 4	8	770	4,500

Further discussion of the irradiation effects studies is given later in this report.

b. 316 has a higher allowable stress than 304 with apparently the same sodium compatibility. Next to 304, 316 has been receiving the greatest attention in the U.S. program on irradiation effects on stainless steels. It has been extensively used at 650°C (1200°F) by the petrochemical and chemical processing industries and currently by the steam-power industry. On the other hand, 316 stainless steel is sensitive to both carbide precipitation and sigma phase formation. Preliminary data indicate that ductility at 650°C is reduced from 60% elongation in the unirradiated condition to 16-30% elongation in the irradiated ($1 - 10 \times 10^{20}$ total nvt) condition.

c. 20Cr-25Ni (Nb stabilized) is a British stainless steel with an unirradiated strength between that of 304 and 316. After irradiation at 590°C and 750°C to 1×10^{20} nvt (fast), this alloy shows a reduction in total elongation of 50 to 80% of the unirradiated thermal control values at tensile test temperatures of 600 to 800°C.

d. Incoloy 800 is stronger than the 304, 316 and 20Cr-25Ni alloys, but it has limited service history and a less well developed technology than the commonly used stainless steels. Incoloy 800, 32% nickel and 21% chromium, offers good high temperature properties and carburization resistance. Sigma phase formation has not been a problem in Incoloy 800. Irradiations at 600°C to 10^{21} nvt (fast) indicate post-irradiation elongations of 23% compared to 34.5% for pre-irradiated samples. Higher nickel alloys are subject to more drastic reductions in total elongation.

e. Other stainless steels which merit screening for LMFBR applications include types 321, 347, and 348 from a higher strength standpoint. 19-9 DL, which is considerably stronger than the aforementioned steels at 1200°F, should be investigated to determine its potential as a cladding material. Modified compositions and metallurgical treatments should be investigated in an effort to develop a cladding material of optimum properties.

The following problems and unknowns indicate areas of future development for the above alloys:

- a. Sufficient fast flux irradiation data are not yet available for any of the above alloys.
- b. Clad-to-sodium compatibility data are needed for 19-9 DL and Incoloy 800 and more data are required for the 304, 316 and 20Cr-25Ni alloys.
- c. More fuel-to-clad compatibility data are needed for all of the candidate fuels and cladding materials, with emphasis on the effect of carburization of stainless steels by carbide fuels, particularly with a sodium bond.

Super Alloys

Super alloys, as defined for the purpose of this report, are alloys containing high nickel content ($> 35\%$ Ni) which exhibit unirradiated strength properties superior to the unirradiated strength properties of stainless steels.

A review of the limited available data on super alloys indicates that the principal advantage of these materials relative to a cladding application for potential LMFBR candidate fuels is an unirradiated elevated temperature strength which is greater than the stainless steels.

Hastelloy X is stronger than all of the stainless steels except the 19-9 DL alloy. Inconel 625 exhibits a 1000-hour stress-to-rupture of 62,000 psi at 649°C (1200°F) which is 27,000 psi greater than the 19-9 DL alloy. From the standpoint of strength alone, Inconel 625 and Hastelloy X should be studied to determine the potential of these alloys as cladding materials.

The most important unknowns associated with the use of super alloys in LMFBR's are (a) irradiation performance in a fast flux, (b) sodium corrosion behavior, and (c) compatibility with the selected fuel material.

Little data are available on the compatibility of super alloys with high temperature sodium. The sodium compatibility of Inconel 625 and Hastelloy X is questionable based on the susceptibility of these high nickel alloys to the mass transfer of nickel. The application of nickel alloys to high temperature sodium systems will require extensive compatibility tests similar to those indicated for the stainless steels. Also, extensive fuel-to-clad compatibility tests are needed to determine the effect of irradiated LMFBR fuels on super alloy cladding materials.

Vanadium Alloys

Vanadium alloy technology is in a preliminary stage of development in both this country and abroad. The most important unknowns associated with the use of vanadium alloys in LMFBR's are (a) sodium corrosion behavior, (b) general irradiation performance, (c) long-term mechanical property behavior, in- and out-of-pile and (d) compatibility with fuel materials.

The mechanical property measurements which have been reported on vanadium alloys are mostly of the short-term tensile variety. The tests which should be run to evaluate the potential of vanadium alloys to nuclear applications are of the long-term variety, e.g., creep at operating temperature. Because of the high chemical reactivity of vanadium, surface contamination at such temperatures becomes a problem and can significantly affect test results. Tests will have to be run in a vacuum or very pure inert gas. Of significance in such tests will be possible aging effects associated with vanadium alloys. Aging could lead to deviations in otherwise anticipated long-term property behavior.

The data on sodium corrosion of vanadium alloys which are presently available were apparently obtained under test conditions which were not rigidly controlled and are, therefore, subject to question. Sodium corrosion tests are needed in which the environmental conditions are rigidly controlled and carefully defined. Of particular concern is pickup by the vanadium of H, N, C, and O. Attention should also be directed toward the effect on performance, if any of the formation of

V₂O₅. It is known that vanadium forms this non-protective oxide quite readily and that V₂O₅ melts at approximately 680°C (1250°F).

Niobium and Its Alloys

More is known about niobium alloys than any of the other refractory metal alloys. Extensive development in the U. S. has brought Nb-1Zr to an advanced stage of technology. With respect to unalloyed niobium, the UKAEA was the first to use the material in an operating fast reactor. Also, considerable experience has been acquired by the UKAEA on niobium corrosion in liquid metals.

The technical bases for the use of Nb and Nb alloys in LMFBR systems are: (a) high strength and good ductility at elevated temperature, (b) excellent fabrication and welding characteristics, and (c) potentially good corrosion resistance in sodium provided that the sodium impurities are controlled. Items (a) and (b) hold for niobium, but the corrosion resistance of niobium at high temperature is yet to be established.

The principal unknowns which surround the use of Nb and Nb alloys are (a) the type and levels of impurities which can be tolerated in sodium in order to retain good corrosion characteristics, (b) the effect of fast flux irradiations on performance, and (c) compatibility with fuels.

The UKAEA has pursued extensive liquid metal corrosion studies, but significant differences between the UKAEA and the U.S. programs exist: (a) Nb (U.K.) vs. Nb-1Zr (U.S.) and (b) NaK (U.K.) vs. Na (U.S.). Extrapolations from one material to another, or from one corrosion environment to another, are risky.

Data on the irradiation behavior of Nb-1Zr alloy are meager and, with the exception of the ORNL in-pile tube burst tests on Nb-1Zr, have been conducted at low temperature ($\approx 100^\circ\text{C}$) and to low fast dosages ($\approx 2 \times 10^{20}$ nvt). No evidence of anomalous behavior is indicated by the data. However, irradiations in a fast flux at high temperature and in flowing sodium are necessary to define performance.

Molybdenum Alloys

As in the case of the vanadium alloys, data is needed in areas of critical importance. The principal questions which arise with regard to the use of Mo and Mo alloys are (a) irradiation behavior, (b) ability to fabricate and weld small diameter tubing, (c) sodium corrosion behavior, and (d) compatibility with the fuel.

The basis for LMFBR interest in Mo alloys are (a) excellent elevated temperature mechanical properties, (b) assumed good compatibility with the fuel and (c) assumed good sodium corrosion behavior. The excellent mechanical properties of Mo and Mo alloys make them attractive for elevated temperature use. Adequate tests in sodium environments are important in view of the susceptibility of Mo to surface contamination.

Compatibility data are sparse and limited to short times. Longer term data at anticipated operating temperatures are required to confirm the present extrapolations.

Present Mo welding techniques involving metal fusion may not be satisfactory. New approaches to the problem are required.

Sodium corrosion testing done to date in the U.S. has generally been in a higher temperature range than envisioned for LMFBR's. Testing under conditions more representative of LMFBR conditions is needed.

Irradiation Effects

Fuel cladding materials will reside in LMFBR cores for periods up to two to three years and will accumulate large, fast neutron doses of the order of 10^{23} nvt. While clad material specimens are now being irradiated in the EBR-II, and many further irradiations are planned, it will be some time before data can be generated in the U.S. development program on the post-irradiation properties of high exposure materials. At a peak flux of 3×10^{15} nv, for example, and an on-stream factor of 50%, it will require about four years in a peak flux position to accumulate a dose of 2×10^{23} nvt.

The data which are available on fast flux irradiations of cladding materials have been mostly generated in the U.K. at Dounreay. Some pertinent data have been received from the USSR program. The program at Dounreay involves three types of alloys, austenitic stainless steels (316, 316L, M316, FV548), nickel base alloys (PE 16) and refractories.

Much discussion and speculation has occurred in the U.S. technical community concerning the kinds and extent of materials damage that may occur in fast reactors. As an example, an interesting discussion* of the subject has been contributed by the Oak Ridge National Laboratory, and it is reproduced herewith.

*Private communication from ORNL.

"Radiation Damage Mechanisms and Effects

In Metals That Fracture at Grain Boundaries"

"Lattice Displacements

"This effect increases linearly in displacement yields per neutron in the energy range of approximately 0.1 Mev to 1.0 Mev, at which point the rate of increase of effectiveness stops with neutron energy, but the absolute damage remains at a relatively fixed high rate. Elastic collision displaced atoms generally increase the strength properties (tensile) and decrease ductility or elongation in all materials, and increase the ductile-brittle transition temperature in iron, molybdenum, tungsten, and their alloys. Displacement damage can be annealed out of most metals and alloys at temperatures above approximately one-half their melting point. (See Table I, following next section.)

"Helium Damage

"The accumulation of helium bubbles at crystal interfaces in materials that fracture by tearing apart at the grain boundaries (as do all nickel-based austenitic alloys such as stainless steels, Inconel, Hastelloy and many of the refractory alloys and metals) cause a reduction in elevated temperature ductility and strength.

"Thermal neutron irradiation of steels which contain a few to ≈ 100 ppm boron produces helium at the crystal boundaries because boron as boron carbide is resident initially as a crystal boundary impurity. We have accumulated proof of the reduction to near-zero ductility (under constant stress) of Inconel irradiated in a thermal flux to about 10^{21} nvt. The effect of this type of reaction involving boron in fast reactors is negligible in all probability, but it should be checked.

"Neutrons with energy greater than 2.8 to 3.0 Mev can produce (n, α) reactions with nickel, iron, nitrogen, oxygen, and to a lesser extent with most other elements. The helium thus produced is 'born' within the crystals and must diffuse to the crystal boundary before damage to properties is evident. This diffusion does occur in all probability at temperatures above about one-half the melting point of the metal or alloy at a sufficiently rapid rate to cause loss of ductility. The temperature where helium damage probably will be restrictive is given in the following table for various metals:

Table I

Temperature at Which Damage
Accumulates (°C)

<u>Material</u>	<u>Displacement Damage</u>	<u>Helium Damage</u>
Iron and Nickel-based alloys	RT to 400°C	> 500°C
Molybdenum and alloys	RT to 1000°C	>1200°C*
Niobium and alloys	RT to 900°C	>1100°C*
Tantalum and alloys	RT to 1100°C	>1500°C*
Tungsten and alloys	RT to 1300°C	>1600°C*
Vanadium and alloys	RT to 500°C	> 800°C*

RT = room temperature

*Based entirely on speculation; no data available.

"That effect due to (n, α) reactions in a fast flux is real is shown by very little data as yet, but is supported by Russian data (for stainless steel similar to our Type-321) for irradiations in a fast flux at 400°C (about 150°C below the temperature where lattice displacement would anneal out) to a total nvt of 1×10^{22} . The specimens were tested after removal from the reactor at the temperatures and with the results shown in the following table:

Table II

<u>Post-Irradiation Test Temperature</u>	<u>Tensile Strength, kg/mm²</u>		<u>Elongation %</u>	
	<u>Unirradiated</u>	<u>Irradiated</u>	<u>Unirradiated</u>	<u>Irradiated</u>
20°C	60	81	32	8.5
500°C	37	52	11	4.0
700°C	23.5	23	12.5	4.0
800°C	13	5.5	16	0

"Tensile specimens were about 4 cm long and strained at the rate of 4 mm/minute. This is a relatively high rate of strain and should give higher ductility results than our test standards. Thus, by our normal test standards, the 4.0% elongation results could be low, even approaching zero.

"Tests at 20°C and 500°C (below half the melting point) show the expected displacement-induced increase in tensile strength, and loss in elongation to 4.0%. At 700°C (about half the melting point), the displacement damage has been annealed out, as shown by the equal tensile strengths, but the elongation has not improved because the (n, α) produced helium bubbles have accumulated at crystal boundaries to give an essentially non-ductile metal. The loss in tensile strength at 800°C is a direct result of zero elongation; the specimen breaks before it can neck down.

"Now, referring to Table I, we can postulate the real dilemma. The nickel and ferrous alloys suffer neutron displacement damage to at least 400°C and bubble disease is operative about 500°C. There is possibly a narrow 'window' of acceptable operation between 400 and 500°C. We see no reason to trust in the existence of such a region of non-overlapping effects until established by experimental evidence. In any event, 1200°F (or 700°C) is not an acceptable temperature for stainless steels based on the above information.

"In the refractory alloys, displacement damage may be controlling for all except the vanadium alloys on which we have no data, only conjecture.

"It is believed that the development of special alloys which have 'resistance' to the bubble disease may be accomplished. ORNL, in current work, has irradiated 304L stainless steel containing 3.0 ppm B (normal for SS) to 10^{20} nvt thermal, and also with 0.2% titanium. The titanium precipitates as a carbide throughout the metal, not at the crystal boundaries, and acts as a trap site for helium in locations where the ductility is unaffected. Table III gives preliminary observations on this work.

Table III

Sample 1 -- Type 304 SS; 0.028% C, 3 ppm B

Sample 2 -- Type 304 SS; 0.028% C, 3 ppm B, 0.2% titanium

Irradiation for Both: 10^{20} nvt thermal in ORR

Test Temperature	% Elongation			
	Sample 1 - w/o. Titanium		Sample 2 - w. Titanium	
	Unirradiated	Irradiated	Unirradiated	Irradiated
700°C	36	22	41	35
840°C	46	13	71	60"

IRRADIATION FACILITIES

Summary

The LMFBR fuels development program requires the determination of the behavior of fuel and clad material, fuel elements, and fuel subassemblies under controlled but varying conditions. Closed loops in a fast flux environment, complete with coolant instrumentation and control (flows, temperature, and impurities) and fuel instrumentation capability are required to test fuel, including vented and failed fuel operation. In this manner, the variables affecting fuel performance can be separated for improved understanding of fuel behavior and the testing requirements of individual laboratories and industrial contractors can be met. Open loop test space, with an environment representative of LMFBR conditions, is required to test larger quantities of the candidate fuels. A high fast flux, of the order of 10^{16} nv, is needed to permit accelerated testing of fast reactor fuels and materials. There are no fast flux facilities in the U.S. to accomplish this purpose. Accordingly, a fast flux test facility having the capabilities mentioned is a high priority requirement for the LMFBR program.

EBR-II is the only existing AEC facility now available for fast flux testing of fuels and materials. Though the test specimens cannot be instrumented and the sodium coolant cannot be sufficiently and precisely controlled (i.e., items such as sodium chemistry, sodium temperatures, and sodium flows in a given test position), valuable and meaningful data can be obtained from this facility. The use of EBR-II should be given top priority and all reasonable means taken to increase its plant availability factor and testing capability.

Fermi, the only existing privately owned fast flux facility in the U.S., is comparable to EBR-II in its ability to undertake fuels and materials testing. Steps should be taken to use Fermi for LMFBR fuels and materials testing as a supplement to the space available for testing in EBR-II.

Thermal flux facilities should be used where meaningful data, such as comparative and screening tests, may be obtained.

Fast Flux Test Facility

The LMFBR program will require extensive use of adequately and accurately instrumented and controlled test space similar to the capability that now exists for thermal reactor programs.

A fast flux test facility should, to the extent possible, serve all U.S. fast flux testing requirements in the future to the same extent that MTR, ETR, and soon the ATR satisfy thermal reactor testing requirements, today.

The objectives for a fast flux test facility are:

- a. An adequately controlled and instrumented environment in a fast flux, representative of LMFBR reactors, for testing instrumented fuel specimens, fuel rods, fuel subassemblies, and clad materials.
- b. Capability to test fuel up to and including failure in dynamic sodium.
- c. Reliable plant performance (high and predictable plant factors).
- d. Means to test fuel for short time periods, minutes, or even seconds.
- e. Interim non-destructive fuel examination capability.
- f. A facility servicing all U.S. fast flux testing requirements.

To meet the objectives listed the following requirements should be considered:

- a. Instrumented closed loops, about 6-inch diameter for a 3-to 4-foot core, and accommodations for up to 6 or more in number.
- b. Instrumented open loops, 6 or more in number.
- c. Rabbit facilities.
- d. Provisions to meet programmatic needs for planned programs other than the LMFBR program.
- e. Facilities for interim and final fuel examination including disassembly, reinstrumentation, and reassembly of specimens, capsules or rods.

The fast flux test facility should have the following operating capabilities:

- a. An integrated fast flux $\approx 10^{16}$, 75% > 0.1 Mev.
- b. Specific power in the range of 500 to 2000 or more watts/gm Pu.
- c. Reactor core outlet sodium temperature capable of operating from 800 to 1200°F.
- d. Sodium impurity control capable of maintaining impurities to below the concentrations required to protect the stainless steel, refractories, super alloys, or other cladding and structural materials. These impurities include oxygen, carbon, nitrogen, and hydrogen.

Specific requirements needed to meet the objectives include:

- a. A minimum of two reactor primary system loops, two secondary loops and two heat sinks, designed to permit continued plant operation with one loop in a non-operating status.
- b. Closed loops accessible and removable external to the reactor.
- c. Adequate provision for closed-loop coolant control instrumentation and monitoring equipment including all equipment for controlling and monitoring the test specimen, capsule, rod or sub-assembly.
- d. Adequate provision for open-loop instrumentation and control equipment.
- e. Accessibility from reactor exterior to disconnect (make and break) open-loop instrumentation.
- f. Plant reliability and availability factor as high as reasonably practical.

The AEC should initiate the design, construction, and operation of a Fast Flux Test Facility (FFTF) at the earliest date practicable. The objectives and requirements for such a facility listed above indicate that its successful undertaking and timely accomplishment will require a substantial and competent development and engineering effort from the best of the AEC-Industry-Laboratory complex. Moreover, it will require a more thoroughly planned, disciplined, and extensive effort than has been characteristic in the past of successful development of facilities that are similar, but not as complex.

Detailed procedural plans should be developed for undertaking the FFTF compatible with the magnitude and importance of the task. This projection of the development, design, and construction requirements will be dependent upon the present conceptual design efforts, as augmented by review and inputs from competent test reactor and liquid metal reactor plant designers. Planning should be directed to the means for selecting the most competent AEC contractors and for insuring their satisfactory performance in conjunction with the best efforts from the AEC, the AEC laboratories, and industry.

EBR-II

EBR-II, an operating AEC owned facility, offers, by the use of experimental subassemblies, the best available fast flux testing facility. Substantial volumes of fast flux testing space (volume equal to over 10 standard driver core subassemblies) could be made available with 1000°F sodium temperatures and a fast flux to about 3.6×10^{15} . The major questions to be considered in connection with the use of EBR-II are: (a) what actions are needed to achieve and maintain an increase in its plant availability factor, and (b) what reasonable steps are available to overcome the current lack of instrumentation capability (particularly fuel temperature measurement) on test specimens. The use of EBR-II to meet the needs of industrial and AEC laboratory participants for LMFBR fuels and materials testing should be given a high priority.

Fermi

Fermi, a fast flux facility, is built and due to operate at base load in 1966. By the use of experimental subassemblies, it offers the only alternate for a fast flux testing facility in the immediate future. Fast flux testing space (estimated up to 12 subassemblies) could be made available to 1000°F sodium temperatures and a fast flux to about 3×10^{15} . As is the case with EBR-II, Fermi has no capabilities for individual subassembly failure detection, fuel instrumentation, closed loops, or high temperature (i.e., 1200°F or higher) sodium. Present indications are that Fermi will not be available for irradiation testing until late 1966. Further considerations include the following:

- a. Only 50,000 megawatt days of operation available in the U-10 Mo fuel on hand. This would give approximately 100,000 MWD/T burnup in test specimens using a fully enriched fuel.
- b. Present plans call for a $\text{UO}_2\text{-PuO}_2$ core after the U-10 Mo is removed.

SEFOR

SEFOR, a privately owned facility now under construction, and designed for fast neutron dynamic studies, has been considered for possible use as a fast flux test facility. It is considered unavailable as an irradiation facility for the LMFBR program primarily because of low flux and lack of adequate fuel test capabilities.

Dounreay and Rapsodie

Information from these projects will be useful as a supplement to the U.S. program. The AEC is discussing arrangements to obtain the maximum information from Dounreay and Rapsodie and the possibility of direct utilization of irradiation space in Dounreay.

Thermal Flux Facilities

Although some data is now becoming available from Dounreay, present limitations on the extent of fast flux testing has impaired developing a correlation between fast and thermal flux fuel element testing. As a result, thermal flux data must be considered as only a tentative indication of what fuels and materials performance will be in a fast flux environment. A fast neutron flux will probably have a different effect on cladding materials than a thermal flux due to changes in vacancy and interstitial production rates, thermal spikes, and neutron-proton and neutron-alpha reactions. Until results can be obtained of effects in a fast flux, instrumented thermal flux test results should be used with caution. Attempts to find correlations between fast and thermal flux should be made as soon as possible and applied to any testing of fast reactor fuel and cladding materials in thermal flux facilities.

The available thermal flux reactors were examined for use as test facilities for the LMFBR program. Thermal reactors, such as the MTR, ETR, ATR, CP-5, GETR, B&W LTR, SRE-PEP, SR and ORR, can and should continue to be used to the extent practicable for screening and comparative fuel and clad materials testing.

The installation of sodium loops in thermal reactors, such as the ATR and ETR, with flux filters appear expensive both in money and reactor power and would provide only a small amount of usable testing space. The flux available in these loops is usually a compromise between fast and thermal and, in all cases investigated, the resultant flux level was low by factors of five to ten when compared with LMFBR requirements.

Several existing U.S. thermal facilities (i.e., EOCR, EBOR, SRE, EGCR) designed for purposes other than fast flux testing have been proposed for conversion to fast flux testing. Modifications to these proposed facilities would be costly and complicated stop-gap measures which would not provide the required volume of flux testing space. It is concluded that such proposed modifications should not be undertaken.