REACTOR ENGINEERING LECTURES

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INTRODUCTION

The twenty-eight lectures contained in this report were prepared from notes used in a classified course in Reactor Engineering given at Argonne National Laboratory from September, 1953 to June, 1954. The lectures were presented to Argonne employees for the purpose of broadening their understanding of the engineering problems involved in the design of nuclear reactors. An attempt was made to present the principles involved rather than to cover all of the facts relating to the various subjects. Some important items were not written down in the notes, but were covered in actual presentations. Other subjects such as nondestructive testing were hardly mentioned because of lack of time. In general, instrumentation as well as the basic physics of reactors were omitted since these subjects were covered in other courses.

Subsequent to the presentation of the classified lectures, the notes were declassified by deleting classified items. They were then rewritten to remove the inconsistencies and references to classified reports and for use in the International School of Nuclear Science and Engineering. For convenience, all the lecture material on fluid flow and on heat transfer has been combined into single lectures although three lectures were actually devoted to each subject.

Lectures 24 and 25 were prepared by R. S. Voris, and Lecture 27 by H. M. Feder. References to classified reports are presented in Appendix A. No attempt was made to include complete references.

The opinions expressed in the lectures are those of the writer and not those of Argonne National Laboratory. The lectures have not been reviewed by the Laboratory as a whole.

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LECTURE 1

NUCLEAR ENGINEERING

HIL-5000

Reference: "The Design of Nuclear Power Plants," J. A. Lane,
S. McLain, Chem. Eng. Prog., 49: 287 (1953).

Introduction

Nuclear engineering covers the applications of the broad fields of fluid flow, heat transfer, ore refining, metallurgy, mechanical design, remote handling, electronic and mechanical instruments, physics, and process engineering to the preparation of materials, design, construction, operation, and utilization of the products of nuclear reactors. The discussions in this series of lectures are limited to just a few topics. The basic physics calculations of reactor design, flux distributions, critical masses, etc., and electronic and mechanical instruments are not discussed.

Nuclear Reactors

The nuclear reactor is the central device or tool of the entire nuclear energy field. It is essentially a device for transforming the potential energies of fission of uranium, U^{235} or U^{233} , and plutonium atoms into heat with or without the simultaneous production of new fissionable material. As a research and development tool, the heat is usually neglected and it is the excess neutrons that are important. As a furnace or furnace and boiler for a nuclear power plant, it is the heat that is important and the excess neutrons are utilized to produce more fissionable material from U^{238} or Th. The heat produced in the nuclear reactor is available at a constant temperature as high as the materials of construction and the coolants can stand. There is no low temperature heat to recover or utilize. The central problem in nuclear design is, therefore, heat removal.

The nuclear reactor may be utilized for various heat cycles as shown in Figure 1.

The nuclear reactor consists of fuel in a suitable form, coolant, fertile material to produce new fuel, cladding materials, control materials and devices, structural materials, usually moderating materials, shielding materials, means for loading and unloading, means for handling and controlling the coolant, and sensing instruments for neutron density and temperature. Since each requirement may be met in several ways, there are a large number of possible reactor types. It is this fact that makes a discussion on nuclear reactors so complex.

WATER COOLED REACTOR

LIQUID METAL COOLED REACTOR

GAS COOLED REACTOR

Figure 1 - Simplified Heat Cycles

Existing and Planned Reactors

In the 12 years since the first self-sustaining reactor was achieved by Fermi's group on December 2, 1942 at the University of Chicago, there have been built and plans have been made for many reactors. Some of these are mentioned below. This list is probably far from complete.

A. Hanford, Richland, Washington - Three war built, Pu production reactors plus some built since the war and under construction. In addition, there is a test reactor for checking materials at Hanford.

B. Savannah River - Several production reactors are under construction or in operation. In addition, there are three test reactors of various types at this installation.

C. Arco - There are two research reactors, MTR and EBR-I, and the land-based prototype of the submarine reactor in operation. The Borax-I tests have been completed and Borax-II is under operation. A reactivity test reactor has been constructed in the canal of the MTR. An aircraft test site is under construction. A fast critical assembly building has been built and a fast reactor is planned as a part of the Commission's Five Year Plan.

D. Argonne National Laboratory - The two research reactors, CP-2 and CP-3', have been taken out of service. The research reactor CP-5 is in operation; in addition, there have been three critical assemblies used at Argonne. A test reactor and a boiling reactor are being designed for construction at Argonne in the near future.

E. Oak Ridge National Laboratory - One war built reactor, plus the homogeneous reactor experiment, the aircraft reactor experiment, the bulk shielding test facility, the LITR, and an aircraft shielding test facility have been built, and a new research reactor to replace the LITR, and another homogeneous reactor are under construction. A larger, intermediate scale homogeneous reactor is under design. The original homogeneous reactor, HRE-I, has been shut down and dismantled.

F. Schenectady, New York - The land-based prototype submarine reactor is under construction at West Milton. At least two small research reactors have been used at various times.

G. Los Alamos, New Mexico - Two small research reactors, one of which was the first to use Pu, the first to be liquid metal-cooled, and, at the same time, the first fast reactor. This fast reactor has been dismantled. The other reactor was the first homogeneous reactor. A larger reactor of the homogeneous type is under construction.

H. Brookhaven - One large research reactor.

I. Westinghouse - Three zero power reactors.

J. North American Aviation - One research reactor.

K. Santa Susanna, California - A pilot, sodium-cooled graphite moderated reactor will be started soon.

L. Fort Belvoir, Virginia - An army, packaged power reactor is under contract.

M. Several universities and research institutions such as Pennsylvania State University and the University of Michigan are planning reactors. North Carolina State has one in operation. Armour Institute, Westinghouse Electric Company, and others have plans for privately owned reactors.

N. Reactors are either planned or are under construction at Convair, Lockland, and elsewhere.

O. Industrial Groups - Several industrial groups have stated definitely that they desire to construct nuclear power reactors. These include the Nuclear Power Group (Commonwealth Edison Company and others), Yankee Atomic Electric Company (New England Power Company and others), Detroit Edison Company and others, and Consumers Public Power District of Columbus, Nebraska. The Newport News Shipbuilding and Dry Dock Company have proposed that they be permitted to construct a ship reactor.

P. Shippingport - A pressurized water reactor is under development by Westinghouse for construction at Shippingport by the Duquesne Electric Company.

Q. The first real utilization of a reactor for power is the submarine "Nautilus" at New London. Additional submarine reactors have been authorized.

Canada, England, France, Russia, Norway - Netherlands, and Sweden have operating reactors, and these countries and several other European and non-European countries are planning additional reactors. Most of the European programs are directed toward power production with little emphasis or interest in weapons materials. The Indian program is of interest in that in the view of their AEC Chairman, Professor H. J. Bhabha, "There is not enough coal or oil remaining in the ground in all the world to raise the living standard of India's 357,000,000 population to one comparable to that enjoyed by the people of the United States." They are primarily interested in thorium breeding. The British program is the most extensive of the power production programs announced to date.

The European public seem to be more reactor-minded than the American public. This may be due to the emphasis on weapons here with its security problems. In addition, European industries are contributing directly to the general reactor development programs.

Most of the above reactors have been built or are being built primarily for research and development work; the production reactors at Hanford, Savannah River, the Submarine Reactors, and some United States, British, and Russian (?) reactors are exceptions to this. The Experimental Breeder Reactor and Homogeneous Reactor, along with the Submarine Reactor, are the only ones that have produced electrical power, and the Submarine Reactor is the only one that is capable of producing significant amounts of power. However, the cost of power plants in the planning or early construction stages is well over one billion dollars.

Several types of nuclear reactors are under development in this country since each one of them gives promise of ultimately producing electrical power at a competitive price, provided development projects can be worked out successfully. Many of the problems in this course will deal with the known methods of attack of these development programs. In general, the technical problems of nuclear power production have been shown to have feasible solutions. Cost analyses indicate that only by continuing development over a period of years will we be able to produce competitive power. Reactors may be used for the production of chemicals, such as various intermediates, plastics, and petrochemicals.

Basic Information

Basic information such as heats evolved during fission and the physical properties of the materials can be obtained from various sources. References of interest as background are:

"The Elements of Nuclear Reactor Theory,"
S. Glasstone and M. C. Edlund, D. Van Nostrand, 1952

"Introduction to Nuclear Engineering,"
R. Stephenson, McGraw-Hill, 1954

"Introduction to Nuclear Engineering,"
R. L. Murray, Prentice-Hall, 1954

'Atomic Energy for Military Purposes,"
H. D. Smyth, Princeton University Press, 1945

'Report on the Atom," G. Dean, Knopf, 1953

"Materials and Equipment for Reactors,"
Nucleonics 11 18-48 (June 1953)

"The Science and Engineering of Nuclear Power,"
Vol. II, C. Goodman, Addison-Wesley, 1949

"Hearings Before the Joint Committee on Atomic Energy 83rd Congress First Session on Atomic Power Development and Private Enterprises," 1953; Super. of Doc., U. S. Gov. Print. Of., Washington 25, D. C.

"Reports to the U. S. Atomic Energy Commission on Nuclear Power Reactor Technology," by the four study groups, U. S. Atomic Energy Commission, May 1953; Super. of Doc., U. S. Gov. Print. Of., Washington 25, D. C.

"The Reactor Handbook," AEC, 1952 is the most complete information source on reactors. This four volume handbook has been classified, but a declassified version is planned.

"Introduction to Reactor Engineering," S. Glasstone, to be published.

Design Procedures

The engineering design problems encountered when one attempts to establish the detailed procedures for design of a particular type of nuclear reactor begin with the calculation of critical mass, neutron fluxes, etc. These are a part of the preliminary work in reactor design and are frequently made by physicists.

The following steps would be followed in the design of a nuclear reactor or power plant:

- A. Establish power level
- B. Select components for the reactor system
- C. Establish size limitations of the reactor core
- D. Estimate amounts of fissionable materials required
- E. Select coolant
- F. Calculate heat removal system
- G. Determine optimum arrangement of fuel and moderator
- H. Design fuel elements and control system
- I. Establish operating conditions
- J. Design reactor structure, including pressure vessel, if any
- K. Design shielding and external facilities.

These various factors are discussed in more detail below:

A. Establish power level - The power level obviously is based on the requirements and function of the reactor and is supplied by the user. Stationary power plants will generally have power levels from 100 Mw to 1,000 Mw of heat energy, which again is dependent on the electrical output desired and the thermal efficiency of the reactor system finally adopted for construction. For the near future, most types of nuclear reactors will be relatively costly to construct, but will use cheaper fuels than conventional power plants. The high capital and low operating costs mean that reactors will be expected to run continuously at the full load rating or they will be base load plants. Thus, the reactors must be designed for continuous operation for many years at their full load ratings. Actually, the power level must be adjusted from time to time as the reactor system is designed because the heating rating will vary somewhat, dependent upon the thermal efficiency of the entire system.

B. Select components for the reactor system - The selection of the most suitable components for the reactor is dependent upon the particular reactor type chosen for construction. The materials which we must consider are listed in Table I.

Table I

NUCLEAR REACTOR COMPONENTS

Component	Material	Function
Nuclear fuel	U^{233} , U^{235} , Pu^{239}	Sustains fission reaction
Fuel diluent	U^{238} , Th^{232} , Al, Zr, Fe	Provides heat transfer surface or absorbs excess neutrons to generate new nuclear fuel
Moderator	C, H_2O , D_2O , Be, BeO	Reduces neutron energy
Coolant	Air, He, H_2O , D_2O , Na, Bi	Removes heat
Control element	Cd, B, Hf	Adjusts neutron level
Structure	Al, Zr, Fe (s.s.)	Supports and contains fuel elements
Shield	Fe + H_2O , Concrete	Protects personnel from radiation

The choice of actual materials will be dependent on the type of reactor selected for construction. It appears at the moment that we have several reactor types which may produce economical power if the engineering problems can be solved so that we have economical systems.

These are listed in Table II. It will be noted that U^{233} and U^{235} are interchangeable in thermal reactors and that an alloy of U^{233} or U^{235} in Th can be used in place of natural U or the slightly enriched U. Obviously, U^{233} or Pu can replace U^{235} . In the fast breeder, either U^{235} or Pu may be used, but Pu gives a much higher conversion ratio. This will be discussed in a later lecture.

The choice of system depends essentially on economic balances of all costs between the various systems. At the present time, only a limited number of these systems have been developed technically to the point that construction of large-scale reactors could be undertaken with assurance, and very little cost information on these reactors is available. The cost data we now have indicate that all the systems potentially have promise of being economical, provided the known development problems can be solved satisfactorily.

Table II

REACTOR SYSTEMS

<u>Reactor Type</u>	<u>Fuel</u>	<u>Moderator</u>	<u>Coolant</u>
Pressurized water	Natural U	D ₂ O	D ₂ O, H ₂ O
Pressurized water	Natural U	Graphite	H ₂ O
Pressurized water	Slightly enriched U	H ₂ O	H ₂ O
Boiling water	Natural U	D ₂ O	D ₂ O
Boiling water	Slightly enriched U	H ₂ O	H ₂ O
Sodium graphite	Slightly enriched U	Graphite	Na
Homogeneous	Natural U	D ₂ O	D ₂ O
Homogeneous, solution or slurry	Slightly enriched U	H ₂ O	H ₂ O
Homogeneous	Highly enriched Pu	None	Liquid Metals
Fast breeder	Highly enriched Pu	None	Na

C. Establish size limitations of the reactor core - The size of a reactor core will depend on the specified power level and power density and upon the critical mass. Normally for power reactors operated with thermal neutrons, more than one critical mass will be required so that it is only in the fast reactor design that we need seriously to consider critical mass in the engineering calculations of heat removal. In general, even in the fast reactor, the heat removal will be the limiting condition upon the reactor. Reactor core dimensions are surprisingly small for the enriched reactors and may be quite large for the graphite moderated natural U reactors. Normally, the core of a power reactor of reasonable size would be a right

cylinder of diameter equal to the height of 3 to 15 feet. For any particular system, excepting the fast reactor, the reactor dimensions can be varied over rather wide limits by variation in the degree of enrichment or the amount of absorbing materials present.

The power density is usually stated as kw/kg of fissionable fuel and is a function of the design or type of fuel element and the efficiency of the coolant. Because of the high cost of the fuel, particularly in fast reactors, effort is made to keep the power density as high as possible. In the MTR the power density is approximately 10,000 kw/kg.

D. Estimate amounts of fissionable materials required - The amount of fissionable material required is usually based upon the heat transfer rate and total power output required. Due to the critical mass considerations, a given minimum amount of material must be present, and due to cost considerations, the minimum enrichment required would be used. Even here, we sometimes have difficulty; for example, the Savannah River reactors have approximately 10 critical masses present. The question of use of enriched fuel is one which is subject to economic analyses since the separations plants for U^{235} at Oak Ridge, Paducah, and Portsmouth can be operated to produce uranium of any U^{235} enrichment required. Pu or U^{233} produced in the Hanford or other reactors may also be utilized as they become available. Insofar as possible, U^{235} as it occurs in natural U should be used, as it is the cheapest fissionable material.

E. Select coolant - In order to obtain minimum holdup with fuel, with the subsequent economic charge for this investment, it is essential to obtain the highest possible power density or specific power possible. This statement, of course, applies to all large reactors built for power or for production of fissionable materials. Thus, in power reactors the heat removal problem in the reactor core is the most important engineering problem and the solution of this problem is dependent primarily on the choice of coolant.

The obvious coolants are air, helium, H_2O , D_2O , sodium and bismuth. The availability of air makes it an obvious choice as a coolant. However, it is a poor heat transfer medium in that it has relatively low specific heat and very low rates of heat transfer. Thus, pumping charges are excessive. In addition, air which is discharged from a reactor must either be circulated or discharged through a stack. In the latter case there is the hazard of the possibility of active material getting into the air stream in the form of particulate matter and, in addition, radioactive argon will always be generated in air used as a reactor coolant. A total of 1850 #/min of air is drawn through the MTR to cool the graphite, thermal shield, and experimental plugs. About 1500 curies/day of argon-41 are produced. Helium has the objections of high pumping charges and the necessity for recirculation. A large external heat transfer surface would be required to cool the helium due to low heat transfer rates. Because of the pumping charges,

gases have not been considered for power reactors if power is to be produced through a steam cycle. The use of gases combined with gas turbines may, however, prove attractive, see Figure 1.

Ordinary water is particularly attractive because of its cost; however, in power reactors where high temperature operation is necessary, water cooling, both H_2O and D_2O , becomes less desirable because of high pressures. Water also becomes radioactive and emits very strong gamma rays of short life on leaving the reactor. Impurities, such as sodium in the water, become radioactive and the system must be completely leakproof. The water decomposes into hydrogen and oxygen in the radiation field and this gaseous mixture must be diluted or recombined to prevent explosive mixtures. Light water is a very efficient moderator, but hydrogen has a rather high neutron absorption. The neutron cross sections of interest are presented in Table III.

H_2O as moderator and coolant cannot be used with natural U and it is necessary to use graphite or D_2O moderators or slight enrichment. The cost of the slight enrichment must be balanced against the high neutron absorption, high pressures, and tight lattices required for H_2O reactors. The main objection to H_2O and D_2O is the fact that water is a very reactive and, therefore, corrosive material.

Organic coolants have been studied, but they undergo rapid decomposition under the irradiation in power reactors. The use of D_2O as a coolant and moderator is advantageous from a neutron viewpoint and because natural U can be used in a D_2O -cooled and moderated reactor. The cost of D_2O requires that the volume of the system be a minimum and that losses be held low. In addition, the investment charged for this water must be balanced against the increase in neutron economy. Obviously, a D_2O system must be leakproof, decomposed gases must be recombined, and great care must be taken in handling the water.

The use of Na as a reactor coolant is extremely attractive for high temperature operation because of the low vapor pressure (no pressure vessel), low corrosive properties, and high heat transfer rates. The entire system must be completely leakproof because of the danger of reaction between the Na and air or water. The handling of Na is difficult because of its melting point, $97^\circ C$, and radioactivity after neutron exposure. All Na surfaces must be blanketed with inert gases to prevent fires. All portions of the equipment must be provided with heaters to prevent freezing, and a purification system must be provided to remove Na_2O and other impurities.

The use of Bi has been under study at Brookhaven National Laboratory. In this case the U is dissolved in the Bi. A reactor in which the U is present as a part of a liquid metal solution used as a coolant fuel material is known as a liquid fuels type of reactor.

TABLE III

CROSS SECTION, BARNS ($10^{-24}\text{cm}^2/\text{nucleus}$)

Isotopes or Element	Thermal			Fast				Slowing Down, ξ
	Non-Fission Capture	Scattering	Fission Capture	Non-Fission Capture	Scattering	Inelastic Scattering*	Fission Capture	
H	0.33	20 - 80	-	5×10^{-5}	4.5	0	-	1.000
D	0.0005	7.5	-	-	-	-	-	0.725
He	0	1.3	-	-	-	-	-	-
Be	0.009	-	-	-	-	-	-	0.206
C	0.005	5	-	$10^{-5}(?)$	3	0	-	0.158
Na	0.49	3.5	-	0.0014	3	0.3	-	0.083
K	2.0	2.0	-	-	-	-	-	-
Fe	2.4	11	-	0.005	3	1	-	-
Bi	0.03	9	-	-	-	-	-	-
Th ²³²	7.0	13	-	-	-	-	>0	-
U ²³³	-	-	533	-	-	-	-	-
U ²³⁵	107	-	580	-	-	-	>0	-
U ²³⁸	2.75	8.3	<0.5 mb	-	-	-	>0	-
Pu ²³⁹	315	-	750	-	-	-	>0	-
Pu ²⁴¹	400 $\pm$ 50	-	1080 $\pm$ 100	-	-	-	-	-

*Inelastic scattering occurs with heavy elements by neutron capture with neutron emission at lower energies.

NOTE: 16% of the U²³⁵ atoms that capture thermal neutrons do not fission and 30% of the Pu²³⁹ atoms that capture thermal neutrons do not fission.

F. Heat removal system - Cooling can be carried out in a number of ways such as forced circulation, natural convection, or boiling or flash vaporization. In power reactors, we would expect to either use forced circulation or boiling with some flash vaporization. It will probably turn out that water or liquid metal cooling by forced circulation will become the most economical system. The utilization of flash vaporization is advantageous because of minimum pumping charges but introduces the possibility of hot spots and burnout of the fuel assemblies.

G. Arrangement of fuel and moderator - In order to obtain the maximum heat transfer per unit weight of fissionable material, the surface area must be very large, relatively high velocities of coolant must be used, and the temperature drops must be high. The design of the fuel elements themselves is dependent upon the metal core systems used, the methods of bonding or maintaining high heat transfer rates between the core and cladding, and the rates of corrosion of the cladding at the elevated temperatures. Aluminum and zirconium can be used as U fuel element cladding materials since they have satisfactorily low neutron absorption rates. The use of stainless steel has been considered for Na-cooled, graphite-moderated reactors and for fast reactors. Tantalum and molybdenum have also been considered for fast reactors. Later lectures discuss how some fuel elements of U and Th and their alloys with Pu and U, respectively, may be clad for use in reactors.

H. Design fuel elements and control system - In order to keep the temperature in the center of fuel elements within reason, the fuel elements must be formed in as thin plates or thin rods as possible. The cladding thickness must be a minimum.

I. Establish operating conditions - The choice of cladding material and coolant with the corrosion rates of the cladding material in the coolant fixes the maximum temperature of the surface of the fuel elements, and thus the temperature of operation. The temperature drop across the film is chosen on the basis of economic considerations of capital investment, pumping costs, and efficiency of the system.

J. Design reactor structure, including pressure vessel, if any - Obviously the reactor core must be put together with structural materials in order to hold the fuel assembly and moderator in place. It is only after the major part of the reactor has been designed that the structural parts are seriously considered. In addition, control elements must be used either in the core or near the core or part of the reflector around the core must be removed to control the power level of the reactor. The design of the pressure vessel for water cooled and moderated reactors is a major problem.

K. Design shielding and external facilities - When all the core parts have been designed, the thermal shielding and the external systems can be designed and matched to the reactor. Of course, in an actual design all parts must be checked to give the most economical over-all system.

LECTURE 2

THERMAL VS FAST REACTORSReferences:

"Basic Problems in Central Station Nuclear Power," W. H. Zinn, Nucleonics, 10 8-14 (Sept. 1952)

"The Failure and Disassembly of the Los Alamos Fast Reactors," E. T. Journey, Amer. Inst. Chem. Eng. Mon., 13: 191 (1954).

Introduction

It is the purpose of this lecture to review briefly the differences between thermal and fast reactors and to mention some variations of the two types.

Comparison of Thermal and Fast Reactors

The fundamental difference between the two types of reactors is the average energy of the neutrons at the time of absorption. In the thermal reactor, this energy varies from 0.025 ev at room temperature to 0.085 ev at 925°C for completely thermalized neutrons. Actual average energies of the thermal neutrons are not much higher in graphite and D₂O moderated reactors. The average energy of all neutrons is, of course, much higher. In the fast reactor the energy is of the order of 0.5 to 1 Mev. At first, one would think that these energy differences would not be too important and the reactors would be quite alike. This is not the case. The reason for the important differences are the variations in cross sections with the resultant effects on critical mass and heat removal.

As noted in Table III of Lecture 1, the cross sections for fission, absorption, elastic scattering, and inelastic scattering differ in varying degrees for the different neutron energies. The absorption and fission cross sections for fast neutrons are low for all isotopes, but increase rapidly at lower neutron energies. The elastic and inelastic scattering cross sections depend on the atomic weights of the elements involved. For low atomic number materials, elastic scattering results in slowing down and in thermalizing the neutrons. For heavy elements, inelastic scattering is relatively more important and such elements are to be avoided in fast reactors. Thus, for fast reactors, we need elements for coolants and structural materials of intermediate atomic number and for thermal reactors we need elements of medium or low atomic number and low thermal cross section.

The ratio of non-fission neutron absorption to the total absorption (fission plus non-fission neutron absorption) varies with neutron energy.

The number of neutrons emitted for the various fissionable isotopes are presented in Table I.

Table I

NEUTRONS PER FISSION WITH THERMAL NEUTRONS

Isotope	Average Neutrons per Fission, ν	Ratio: Non-Fission, α Fission	Net Neutrons per Fission, η
U^{233}	2.50	-	-
U^{235}	2.50	0.184	2.12
Pu^{239}	3.00	0.42	2.11

The cross sections for non-fission capture for all elements tend to decrease with increased neutron energies. However, many isotopes have high resonance capture bands in the intermediate zone, but these disappear at high neutron energies. There result several facts of interest. First, the critical mass is high for fast reactors as compared with thermal reactors. In general, for a power reactor only one critical mass would be present in a fast reactor while there might be several critical masses present in a thermal power reactor. Second, the inventory cost of maintaining the high critical mass of the fast reactor is appreciable. Third, due to the high velocities of the neutrons in fast reactors, neutron leakage from the core becomes large as the volume and surface area is increased. Nevertheless, the core must be diluted for large power reactors in order to provide cooling surface and volume for the coolant.

In the minimum sized fast reactor, the core consists of undiluted enriched fuel elements with the minimum amount of coolant. Such a core as used in the Experimental Breeder Reactor I and the Los Alamos Fast Reactor is about the size of a gallon can. For a large size power reactor producing several hundred thousand kw of heat, the core is made larger by dilution of the fuel with U^{238} . The U^{238} is slowly converted to Pu and partially fissioned by the fast neutrons. An optimum design would permit the production of Pu in the core as rapidly as the fuel is consumed. The core might be a right cylinder a few feet in diameter and height.

On the other hand, a thermal reactor may be built with natural uranium containing the U^{235} diluted with U^{238} as it occurs in nature. This represents the lowest cost fuel per gram of fissionable material (U^{235} in natural U at

\$10/lb is worth \$3.1/gm as compared with an estimated value of \$20/gm for fully enriched U^{235}). For heavy water and graphite-moderated reactors, the critical masses of natural U run from 20 to 200 tons. Light water and Na-cooled, graphite-moderated reactors require from 10 to 100 tons of slightly enriched U (U containing 1.0 to 1.5% U^{235} as compared with 0.711% U^{235} in natural U). Fully enriched H_2O and D_2O -cooled and moderated reactors require from 1 to 5 kilograms of U^{235} .

In order to utilize the neutrons which leak out of the core of the fast reactor, the core is always surrounded by a blanket of a heavy element. The normal material is U^{238} . Part of the neutrons are utilized in forming Pu^{239} , part are reflected into the core, part cause fast fission, and part leak out of the reactor. The excess neutrons are utilized in the blanket to form additional Pu. Outside of the blanket, iron may be used as a pure reflector since the iron nuclei reflect many of the neutrons without appreciably slowing them down. Part of the neutrons are slowed down by inelastic scattering, part are reflected, and part pass through the iron.

In a thermal reactor, the fuel is diluted with moderating materials and surrounded by a reflector of moderating material. Here the reflector both slows the neutrons down and "reflects" them to the core. Due to the high cross sections of the fissionable atoms, the fuel may be very dilute.

We define the conversion ratio of a reactor as the ratio of the number of new fuel atoms produced per atom of fuel destroyed. In thermal power reactors using fuel diluted with Th^{232} or U^{238} , the conversion ratio varies from 0.7 to 1.0, while in fast reactors the conversion ratio varies from 1.0 to 1.5 or more. Some people use the term breeding ratio to refer to the production of new fuel by fission of the same kind of fuel. Other people use the term breeding ratio only to designate a conversion ratio over 1.0. Actually, the term conversion ratio is usually used in reference to thermal reactors, while the term breeding ratio refers to fast reactors, and there is little confusion.

Xenon-135 is formed by decay of I^{135} which is formed from Te^{135} (a fission product) in reactors. The thermal cross section of the Xe^{135} is about 3.5×10^6 barns and so it acts as a serious poison. A second fission product, samarium-149, has a high cross section in thermal reactors. Both of these materials and the other fission products have low cross sections in fast reactors and are not troublesome.

The control of thermal reactors is carried out by addition or removal of materials which have high neutron absorption cross sections such as Cd, Hf, and B. In fast reactors, control must be carried out by movement of the reflector or by some other change that increases or decreases the neutron leakage. The amount of excess reactivity and the control required is much higher in a thermal than in a fast reactor since the change in poisons present is rapid and large in thermal reactors.

Thermal reactors usually lose reactivity with increase of temperature. They are said to have negative temperature coefficients or are non-autocatalytic. Less is known about fast reactors. The life of a neutron in a fast reactor is of the order of 0.1 to 0.01 that in a thermal reactor.

U^{238} may undergo fission when it absorbs a neutron with an energy of 1.1 Mev or greater. In heavy water and graphite-moderated reactors fueled with natural U, the fast fission effect is about 1.03, or about 3% of the fissions occur with fast neutrons in the U^{238} . In the slightly enriched U, light water-cooled and moderated reactor, the fast fission effect may be higher (up to 10%) depending upon the H_2O/U volume ratio. The lower the amount of moderator, the greater the fast fission effect will be. In a fast reactor using a diluted fuel core, about 1/5 of the heat production is in the blanket. This heat production is due mainly to the fast fission effect.

As noted above, in thermal reactors resonance absorption is very important. For example, U^{238} has resonance absorption bands at about 6.5 and 30 ev. Th^{232} has a resonance absorption band at about 23 ev. Resonance absorption is most important in that it removes a portion of the neutrons while they are being slowed. This has led to clumping the fuel with its partial segregation from the moderator. In homogeneous reactors, clumping is impossible and slightly higher enrichment is required than for heterogeneous reactors. The resonance absorption leads to Pu^{239} or U^{233} so the over-all conversion ratio is not altered appreciably.

Thermal Reactors

The core of thermal reactors consists of the fuel, fertile material, moderator, coolant, control elements, and structural materials, if any. Fully enriched reactors contain no fertile material and the core and critical mass may be very small. Power reactors will have cores of larger sizes depending upon the moderator and the number of critical masses present. The sizes of the cores of different types of reactors are indicated in Table II.

The reflector consists of 1 to 3 feet of H_2O , D_2O , or graphite. Usually, graphite would be used in place of H_2O because of the high cross section of the water.

Some two feet of iron and H_2O and 8 to 10 feet of concrete are used for shielding of all reactors. A cross sectional view of Argonne's Research Reactor, CP-5, which illustrates the various components of a thermal reactor is presented in Figure 1.

Figure 1. Cutaway Drawing of CP-5

Table II

REACTOR CORE SIZES

<u>Fuel</u>	<u>Moderator</u>	<u>Coolant</u>	<u>Approximate Diam & Ht of Core, ft</u>
Fully enriched U	H ₂ O	H ₂ O	1 - 2
Fully enriched U	None	Na	1
Slightly enriched U	H ₂ O	H ₂ O	4 - 6
Natural U	D ₂ O	D ₂ O	8 - 12
Slightly enriched U	Graphite	Na	10 - 15
Natural U	Graphite	H ₂ O	20 - 30

Fast Reactors

An important consideration in fast reactors is the rate of increase of the fissionable fuel. The breeding ratio may be defined as:

$$BR = \frac{\nu - 1 - \alpha - \ell + F (\nu^1 - 1)}{1 - \alpha}$$

in which

ν = neutrons released per fission

ν^1 = neutrons released per U²³⁸ fission

α = neutrons wasted in non-fission capture of fuel

ℓ = neutrons wasted in capture by structural materials, coolant, fission products, and leakage. ℓ depends on the design and operating history of the reactor.

F = Fraction of fissions in U²³⁸

The doubling time in days is approximately given by the following equation:

$$D.T., \text{ days} = \frac{10^6}{\text{Breeding gain} \times \text{Specific power}}$$

The specific power of a reactor is defined as the kw/kg of enriched fuel. We must include the fuel undergoing recovery and refabrication in computing breeding gain.

Fast reactors may be heterogeneous, homogeneous, or fluid fuel. In heterogeneous and fluid fuel reactors the central problem is one of heat removal with the minimum critical mass. In the homogeneous reactors, the core is separated from the blanket by a containing wall.

For heterogeneous reactors we may use either natural or depleted U and 10 to 20% Pu as the fuel. The fuel cladding may be either Zr or stainless steel and Na coolant.

In fluid fuel reactors the fuel is pumped through fixed fuel channels and the reactor cooled by liquid sodium or other coolant just as if the fuel were fixed in the fuel elements. No really suitable liquid fuel has been suggested to date. Cr or Fe alloys of U can be used in a Ta system, but the melting point of the UCr or UFe alloy is high, near 800°C. Corrosion and mass transfer studies have not been completed.

For the homogeneous reactors a large number of fused salt and liquid metal systems have been suggested. The most interesting are the Bi-Pb-U alloys and Bi-U alloys. Solubility is low. For example, a solution of 4.6 wt % U in Bi freezes at 655°C.

Los Alamos Fast Reactor

The first fast reactor was a low power reactor which was developed and built at Los Alamos and known as the Los Alamos Fast Reactor. This reactor was unique in three ways: it was the first fast reactor, the first Pu-fueled reactor, and the first liquid metal-cooled reactor.

The reactor consisted of a 6 in. high by 6 in. diam cylindrical array of plutonium fuel elements in mild steel jackets in a mild steel tank. Mercury was pumped in through the bottom of the tank and out through the top by an external electromagnetic pump to a heat exchanger system. Some natural U rods were interspersed with the Pu rods to give symmetrical flow to the Hg.

The core was surrounded by 6 in. of silver-plated natural U blocks.

This assembly was surrounded by shielding through which there were many experimental holes. The reactor was controlled by interchanging some of the U reflector with boron-10 rods. In addition, a large section of the reflector could be dropped 3 in. The temperature coefficient was negative and amounted to $2.6 \times 10^{-5} \delta k$ per °C. The nominal operating power level was 25 kw with a fast flux of 10^{13} .

The reactor was brought to power in March 1949, and operated about 3 years when it was shut down permanently, due to fuel rod failure, and dismantled.

EBR-I

The Experimental Breeder Reactor, EBR-I, was developed at Argonne National Laboratory. The core of the reactor was designed in detail and built by Argonne and the building and facilities were designed by the Austin Company. The reactor was built at Arco.

The core of the reactor was made from enriched U^{235} . The core and inner blanket are cooled by NaK, an alloy of sodium and potassium which is liquid at room temperature. Surrounding the core is an inner natural U blanket which is NaK cooled and an outer blanket which is air cooled.

The reactor has operated almost trouble-free for three years at 1400 kw of heat power. It was the first reactor to produce heat for electrical power and the first reactor used to demonstrate breeding, or a conversion ratio over 1.0.

The NaK enters the reactor at 250°C and leaves at 350°C. The heat is transferred through an intermediate heat exchange system to a boiler which produces 400 psig steam. The heat transfer rate in the core is about 210,000 BTU/ft²hr with a coolant flow rate of 210 gpm. The flux is 1×10^{14} neutrons/cm². Control of the reactor is carried out by movement of part of the reflector.

The major parts of the reactor system are illustrated in Figure 2. Figure 3 illustrates the outside of the reactor including the shielding and top plug.

The reactor has been operated for much of its life at low power as an experimental facility for the study of the basic physics of fast reactors. Detailed studies of the ratios of non-fission to total capture cross sections and measurements of the average energies of the neutrons present at various points in the reactor have been made.

EBR-II

In view of the excellent results obtained with EBR-I, it has been decided to build a larger version of the Fast Breeder Reactor of an intermediate or pilot plant size for power production as a part of the Commission's Five Year Plan. This reactor, known as EBR-II, is under development and design at Argonne. The essential differences between EBR-I and EBR-II will be the dilution of the enriched core, a higher power level, and a higher rate of heat transfer. The purposes for construction of this reactor are: (1) to gain experience in operation of fast reactors which have diluted cores, (2) to obtain further experience with liquid metal cooling of reactors, (3) obtain

Figure 2. Components of EBR-I Reactor System

Figure 3. A View Vertically Downward of the Reactor Shield and the Top of the Reactor Tank.

economic data on a pilot plant scale reactor for the use of fast reactors for production of civilian power, (4) to obtain further data on control of fast reactors, and (5) to obtain further knowledge of breeding and fast neutron physics. The reactor will produce 62,500 kw of heat in the core and blanket. Some 15,000 kw of electrical energy will be produced.

The reactor is illustrated in Figure 4. The fuel elements of the core, not shown, will be made from either Pu or U^{235} . Surrounding the core will be a blanket of depleted or natural U which will absorb the excess neutrons that leak out of the core.

Na, which melts at 97°C , will be pumped through the core and blanket by the electromagnetic pump shown in the illustration. The operation of this pump will be discussed in Lecture 20. The Na will circulate through a heat exchanger in which the heat will be transferred to NaK.

The loading and unloading mechanism is shown above the reactor core. The irradiated fuel will be simply removed to a storage rack in the reactor tank for several weeks for decay of part of the fission products.

The NaK will transfer the heat, in turn, to boiling water. Steam at 600 psig will produce electrical energy in a regular turbine and generator complex.

It is planned to build the reactor at Arco near EBR-I. Experimental fuel element manufacturing facilities and a fuel recovery plant may be built as a part of the reactor plant.

British Fast Reactor

The British have announced that they are designing a fast reactor to be built in the North of Scotland. No details of the design are available.

Figure 4. Cutaway Drawing of EBR-II Reactor System

LECTURE 3

LIGHT AND HEAVY WATER REACTORSReferences:MTR

"Three Declassified Reports on the MTR," N. F. Lansing, ORNL-CF-53-12-21, Feb. 10, 1954. Reports were prepared by John R. Huffman and L. J. Weber

Packaged Power

"A Ten-Megawatt Pressurized Water Package Reactor and Associated Power Plant," A. L. Boch, W. R. Gall, and W. H. Jordan, ORNL-CF-53-10-106, March 18, 1954

NRX

"Canadian Facilities for Isotope Production and Bombardment," F. W. Gilbert, Nucleonics, 10, No. 1, 6 (1952)

"The NRX Pile at Chalk River," W. B. Lewis, Physics Today, 4, No. 11, 12, (1951)

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CP-5

"A Description of the Argonne National Laboratory Research Reactor CP-5," J. T. Weills, Amer. Inst. Chem. Eng. Mon., 11: 213 (1954).

Power

"A Current Design for a Full Scale Reactor Power Plant," T. G. LeClair, Amer. Inst. Chem. Eng. Mon., 13: 122 (1954)

Introduction

It was early recognized in the Manhattan District that light water could not be used as a moderator coolant for natural U reactors because of its high cross section. The resonance absorption of the U makes it desirable to open up the lattice so that nearly all neutrons will be completely

thermalized without coming into contact with U^{238} . If this is done with H_2O , the absorption in the H atoms becomes excessive. For C and D atoms the cross sections are low enough to permit their use as moderators with natural U. In other words, k for any H_2O -natural U lattice is always less than 1.0.

The use of light water has so many benefits in respect to availability and coolant properties that it is used to cool the Hanford graphite reactors and the Chalk River D_2O -moderated reactor. In these reactors, the amounts of light water present are kept low and the graphite and D_2O account for most of the neutron moderation.

Professor Eugene Wigner in 1946 suggested the use of light water-cooled and moderated reactors for use in research and development since such reactors would give an important gain in fast flux over heavy water-moderated and cooled reactors. Subsequently, the Materials Testing Reactor became a heterogeneous, enriched, light water-cooled and moderated reactor. The fuel elements were thin Al plates with enriched Al-U alloy cores separated by narrow cooling passages.

Beginning in the fall of 1946, active development work was carried out at Oak Ridge on light water as a coolant. Flow tests for pressure drop, vibration, corrosion, and heat transfer were carried out for a period of years. These tests demonstrated that high heat transfer rates using Al-clad fuel elements could be obtained for the expected life of two weeks under the conditions of deionized water and temperatures up to $100^\circ C$. Corrosion films built up rapidly for the first two weeks and then the rate of corrosion fell off nearly to zero under the conditions of test - deionized water at about the boiling point. Methods of manufacture of Al-U fuel elements were developed to produce satisfactory plate assemblies having high heat transfer area per unit volume. The basic physics were studied. A control philosophy and a control system were developed that would permit control of reactors having very short periods.

One of the fears was that bubble formation would cause fluctuations in the neutron flux. Another fear was that the reactor would continue to increase in power level until the fuel plates were melted in case an accident occurred and the reactor became prompt critical. Extensive, but inconclusive, calculations were made on this latter point. The reactor required various materials of construction and extensive development of the external system.

During the two or three years from 1949 to 1952, the use of slightly enriched U for power reactors was discussed at length. The use of slightly enriched U with light water and sodium-cooled, graphite-moderated reactors may be cheaper than other fuels and moderators under certain conditions. Reactor size, rate of burnup, shipping costs, chemical costs, etc., all have their effects on over-all costs. No categorical statement can be made that will hold under all conditions. Each reactor type must be examined in detail for each use considering the local conditions.

In 1952, the Fluor Corporation was asked to make a more complete cost estimate of the use of the light water-cooled and moderated reactor as a production tool and power reactor. It is this design which has been stated by Fluor to be able to produce power at rates which are nearly competitive with fossil-fueled fired plants.

The Borax tests run at Arco in 1953 and 1954 were another logical development of this reactor type. Further studies of H_2O vs D_2O for use in boiling reactors are still under way at Argonne.

Light water has been used in the Swimming Pool Reactor and is being considered for other research reactors.

Some of the advantages and disadvantages of light water-cooled and moderated heterogeneous reactors are listed in Table I. Some of these also apply to heavy water.

Table I

ADVANTAGES AND DISADVANTAGES OF H_2O REACTORS

<u>Advantages</u>	<u>Disadvantages</u>
Good heat transfer	Requires slightly enriched U
High heat capacity as a coolant	Reactive or corrosive
Cheap	High vapor pressure
Known technology	Heavy equipment required
Low viscosity	High cross section
Small reactors	Low conversion ratio
Excellent moderator	Partially decomposed in reactor
High fast fission effect with U^{238}	Cannot be made to breed with U
Inherent safety (This is discussed in Lecture 7)	Short neutron life

The research reactors such as the Swimming Pool Reactor all are similar to the MTR in core design. CP-5 has similar fuel elements.

Description of the MTR

The purpose of the Materials Testing Reactor was to provide varied irradiation facilities at the highest practical fluxes. The fast flux was set as 10^{14} and the thermal flux in the reflector then became 2×10^{14} . To achieve these fluxes in a reactor to be used for varied experiments meant that the core had to be small or the power level had to be high. A D_2O reactor is much superior to the H_2O reactor in respect to volume in which there is a high thermal flux.

So the minimum critical mass was chosen which would permit reasonable operation and still allow excess reactivity for experimental work.

The reactor was originally designed for construction at ORNL. When the reactor program was transferred to ANL on January 1, 1948, the reactor was considered for construction at ANL. When the possible hazards were considered, it was finally decided to build the reactor at Arco.

The core, shielding, and control facilities were designed at Oak Ridge and the remainder of the plant was designed at Argonne. Detailed design was carried out by Blaw-Knox Construction Company and the main construction was carried out by Fluor Corporation. Phillips Petroleum Company has been the operator. Construction costs were about \$18,000,000.

Most of the development work was conducted at Oak Ridge. This work culminated in the full-scale flow tests and critical assembly. This critical assembly is still in operation as the Low Intensity Test Reactor, LITR, at Oak Ridge. Presumably it will be disassembled after completion of the Oak Ridge Research Reactor, recently authorized.

The fuel is enriched U and is alloyed with Al. The alloy is clad with 2S Al. The plates are then assembled into fuel elements. End fittings are riveted on and the assemblies given a final machining consisting of grinding down four small contact plates on the bottom fitting to give a vertical alignment to the fuel element.

The core is 9 x 27 x 24 in. and either a slab or square loading may be used. At present a slab loading is being used with 23 fuel elements and 4 shim rods in a slab with the shim rods located at the (2,2), (2,4), (2,6), and (2,8) positions. Each fuel element contains about 160 grams of U^{235} or a total critical mass of about 3.7 kg. Each fuel plate is 0.064 in. thick and the water passages are 0.113 in. wide, see Figure 1.

The bottom grid is a heavy Al casting and the top grids are removable castings which support the fuel elements and guide the shim rods. Be is used to fill the space around the core and completely fill the Al core tank which is about 54 in. ID. The Be has a minimum thickness of 30 cm.

Above and below the Al tank are other tanks which act as water inlet and outlet ducts. 20,000 gpm of deionized water are run through the tank with water velocities of 30 ft/sec. The water temperature rises from 100 to 111°F. The maximum design heat transfer rate is 540,000 BTU/ft²hr, see Figure 2.

Outside of the Al tank there are several feet of graphite. The portion of this graphite near the tank was so irregular due to the experimental facilities that 1-in. diam balls were used. The graphite is cooled by air drawn into the reactor from the reactor building and exhausted through a 200-ft stack without filters.

Figure 1. Pictorial Cross Section of MTR Core and Reflector

Figure 2. Vertical Cross Section of the MTR

The thermal shield consists of two 4-in. thick layers of steel, see Figure 3. Except for the top, this is cooled by the air as it is drawn into the graphite. The top of the thermal shield is cooled by air bypassed through it. The facilities are cooled by air leaking around them. The main beam holes have water cooling facilities for the plug tips which extend into the reactor tank.

The biological shield is barytes concrete placed by the Prepakt method.

The central Al tank has been so designed that it can be removed with difficulty through the top of the reactor.

Loading and unloading is carried out by means of hand tools that pick up and drop the fuel elements from the core into a discharge chute which leads to the canal below the reactor. This discharge chute is equipped with a hydraulic operated valve to prevent flow of reactor water to the canal. In addition, the canal fuel element handling device seals the lower end of the discharge chute while the regular valve is open. The top plug contains about 12 in. of Pb. During shutdown, this plug may be removed, the top of the reactor flushed with fresh water, and unloading and loading carried out by hand. Reloading is carried out at about 21-day intervals.

Two control rods are located in the Be just outside of the core. Normally, only one rod is used with one in standby. Two additional rod spaces are available. The rods are Cd and control about 0.005 k. The rods are driven through amplidynes.

Eight shim rod places are provided and four ordinarily used, see Figures 1 and 4. Each rod is made up of a poison section and a fuel section. During shutdown the poison sections are in the core and the rods then act as safety rods. During operation the poison sections are withdrawn and as much fuel drawn in as is required. Xenon override for 1/2 hr only is provided. Each shim rod contains about 100 gm of U^{235} .

The shim rods are operated by induction motors through gear trains. The scram feature consists of an electromagnet on each rod drive that will hold the rod under normal operations, but will release the rods on scram shutting down the reactor in some 15 to 20 milliseconds. These are shown at the lower end of the rods in Figure 4.

The reactor instrumentation is described in the following paragraphs:

The neutron-flux signals for operating the control circuits come from 10 to 14 instruments located in the 10 instrument holes provided in the reactor structure. Three ionization chambers are reserved solely for the safety function to furnish scram signals at 1.5 to 3 times the operating level and to

Figure 3. Horizontal Cross Section of the MTR

furnish a slower signal to initiate reversible reduction of the operating level. Other signals provide either visual indication to the operator or signals to the servomechanism and other automatic devices. For visual indication from the operating level to 10^{-5} of the operating level, a direct-reading galvanometer is available. Another galvanometer shows the logarithm of the intensity factor from the operating level down to 10^{-11} of the operating level. In the lower part of the range the signal arises from a fission chamber and counting-rate meter.

There are three groups of instruments furnishing information to the operator and to the control system; (1) the reactor instruments measuring neutron flux or power level, (2) the rod position and motion indicators, and (3) the instruments for the auxiliary facilities.

Reactor Instruments

It is necessary to make a rather arbitrary distinction between reactor instruments and instruments on the auxiliary facilities, particularly on the cooling water. Reactor instruments include radiation instruments measuring the neutron level in the reactor and radioactivity in the water, temperature instruments measuring the distribution of exit water temperature across the active lattice and temperatures in the thermal and biological shields, and pitot tubes measuring the distribution of water flow from the bottom (exit) of the fuel assemblies.

The reactor radiation instruments are listed in Table II.

The importance of having proper instruments for control purposes is made evident by consideration of Figure 5, which shows the relation of the power (or neutron) level considered in four definite ranges to the choice of instrument and control action. These ranges serve as convenient divisions of the entire range of reactor operation. At the lowest level is the source range, which lies below the range of the counting rate meter and in which control rod withdrawal should be prohibited. Until the level is raised above 10^{-11} of full power by a suitable source, the effective k of the reactor should be kept less than 1.0, i.e., rods should not be withdrawn. Next comes the counter range, in which the combination of fission chamber and counting rate meter is the source of a reliable signal. For the MTR only intermittent rod withdrawal is permitted in this range. The lower limit of the period meter makes the transition to the period range. By the time this range is reached, the cold, clean reactor will have become critical and the level will be rising on a stable period. Finally comes the power range, over which an appreciable fraction of full power may be measured.

Figure 5. Relation of Power Level to Instruments

Table II

REACTOR RADIATION INSTRUMENTS

<u>Instrument</u>	<u>Use</u>
Parallel circular plate ionization chamber	Safety and servo signals
Compensated ionization chamber	Log N* and period signal; galvanometer signal
Fission chamber, 12-ft travel	Counting rate signal
Air wall ionization chamber	Water flow; N ¹⁶ monitoring
Water monitor ionization chamber	Water sample monitoring
Boron thermopiles	Neutron flux in graphite

* N here means essentially neutron flux (nv)

Experimental Facilities

A few of the experimental facilities are listed in Table III.

The horizontal beam holes penetrate to the lattice. While these are costly to use, due to the necessity for continuous water cooling, they can be used for loop tests as they are completely equipped with various kinds of service facilities, see Figures 2 and 3. The horizontal through hole is the only hole that penetrates completely through the reactor. The difficulty in the use of such holes is the thermal expansion of the loop components. Thick lead doors are provided in the main test holes to permit change of plugs by means of a coffin. The most useful facilities have been the removable Be pieces. Small irradiation samples are placed in the center of such pieces and irradiated at the higher fluxes. Already the results have indicated what alloys are expected to be stable and usable in power reactors.

The MTR cooling water is drawn through a flash evaporator in which the bulk of the water is cooled by flash evaporation and the steam formed, is condensed in turn by circulation through cooling towers.

All the experimental holes are filled by plugs during normal operation. Experimental plugs are built to include special test devices and samples at their tips. The sequence of loading the experimental plugs follows: with the reactor shut down, a heavy lead coffin or shield is attached to the outside of the reactor shielding and locked in place; the regular plug is pulled out into the coffin; a thick door in the reactor end of the plug closed; a heavy

Table III
EXPERIMENTAL FACILITIES

Type of Hole	Facility Designation	Size, in.	Approach	Estimated Flux (max)	
				Thermal	Fast (>1 mev)
Through	VT2	2-3/4 x 2-3/4	Vertical thru shim rod hole	2.5×10^{14}	1×10^{14}
Through	HT1	4-7/8 x 4-7/8	Horizontal thru reactor	1.8×10^{14}	3×10^{13}
Beam	HB1,2,3	6 ID in Be	Horizontal to core edge	5×10^{14}	7×10^{13}
Beam	HB4,5,6	6 ID in Be	Horizontal to Be in active sect.	2.8×10^{14}	2×10^{11}
Beam	HG9	10 x 16	Horizontal to tank wall	8×10^{13}	4×10^{12}
Beam	DB1,2,3,4,5,6	6 ID	Inclined down to tank	4×10^{13}	5×10^{10} (DB2 only)
Thermal	T2	6 ft x 6 ft	Horizontal to Pb window	1×10^{12} max.	
Pneumatic Rabbit	HR1,2	1 ID	Horizontal thru reactor	1.6×10^{14}	4×10^{13}
Pneumatic Rabbit	HR3,4	4 ID	Horizontal to reactor tank	8×10^{13}	
Hydraulic Rabbit	VH1,2	1 ID	Vertical from basement	2.4×10^{14}	
Hydraulic Rabbit	VH3,4	1-1/2 ID	Vertical from top	2.5×10^{14}	10^{13}
Fast Flux	~40 Be Ass'ys	3 x 3	Vertical from top tank*	5×10^{14} max.	7×10^{13} max.
Fast Flux	~15 Be Ass'ys	3 x 3	Vertical from top tank*	5×10^{14} max.	7×10^{13} max.

*Charged or discharged only when reactor is shut down and top plug is removed.

Note: These estimated neutron flux values are based on beryllium or graphite (according to material penetrated) in the experimental holes. Actually, experimental apparatus and samples will cause local "sinks" and reduce the flux attainable for a particular experiment.

12 in. thick lead eccentric door within the concrete shield is closed shutting off the gamma radiation from the core; the coffin is removed and replaced by a coffin containing the experimental plug; and the sequence of operations reversed.

Test facilities and the regular plugs that extend into the reactor next to the core must be water-cooled at all times during operation. The outer portions of the plugs are cooled by air drawn into the reactor between the plugs and hole liners. The smaller lower-flux holes can be cooled by the cooling air passing through the graphite.

As noted above, the 55 removable Be assemblies have proven to be of exceptional value for irradiations of small samples at very high fluxes. The centers of the Be pieces are drilled out and samples placed in them. The normal cooling water provides ample cooling and the test pieces can be dropped into the canal for reloading.

Research Reactors

The Low Intensity Test Reactor at Oak Ridge was built to test the hydraulic characteristics of the MTR. For this purpose, only the reactor tanks and a mocked-up core with external circulating pumps and piping were provided. Al was used as a stand-in for the Be and the fuel elements were dead, i.e., they contained no fuel, but in other respects the core followed the MTR design. Later, design changes in the orificing were made for the MTR. This mockup was used to develop loading and unloading techniques for the MTR.

After completion of the hydraulic tests, the reactor was loaded with regular fuel assemblies and used for a check of the neutron characteristics of the MTR. When these tests were completed, shielding was built around the reactor and it has been used as a test facility. The reactor produces a flux of about 10^{13} .

The Bulk Shielding Facility was designed and built at Oak Ridge for the specific purpose of providing a low intensity but portable neutron source to test various shielding materials in various arrangements. This reactor uses an enlarged MTR core in an open tank with H₂O reflector.

The use of H₂O is being considered for various research reactors such as the University of Michigan and Pennsylvania State University Reactors.

STR

The design studies of a Submarine Thermal Reactor were started at Oak Ridge. After transfer of the reactor program to Argonne in 1948, the decision was made to use pressurized H_2O in an enriched lattice. Design and development work were carried out at Argonne and Westinghouse at Bettis Field. While the reactor core was mainly designed at Argonne, Westinghouse carried out part of the development of Zr-clad fuel elements, designed most of the auxiliary equipment, and built both the land-based prototype at Arco and the Nautilus reactors.

The work at Argonne and Westinghouse involved an extensive program of development and test of materials, heat transfer, control and zero power reactor tests. The Arco prototype has operated very successfully.

Power Reactors

Several studies have been made by industrial groups of the use of light water-cooled reactors for power production. As noted above, such reactors require slightly enriched U. The additional fissionable material may be added by removing slightly enriched U from the diffusion plants, adding fully enriched U either by mixing with natural U or by making separate fuel elements, or by recycling Pu.

In comment on the above procedures, it has been noted that slightly enriched U may be removed from the diffusion plants. However, this requires the preparation of UF_6 for the diffusion plant and several steps to obtain metal from the slightly enriched UF_6 removed from the separation plant. These steps are costly. The use of fully enriched fuel in separate fuel elements permits the recovery of the unburned fuel in the Arco Chemical Separation Plant. A reactor which is operated primarily with natural U and with the addition of some enriched fuel elements is known as a "spiked" or "seeded" reactor. The use of Pu is advantageous in that it may be mixed with natural U and recovered in the Hanford Separation Plants. Use of Pu requires that the fuel elements be prepared in glove boxes since Pu is toxic.

One advantage of the light water reactor as fueled with slightly enriched U or with natural U spiked with enriched U is the high fast fission effect that may be obtained. With tight or close lattices in which the ratio of volume of H_2O to the volume of U is kept low, the fast fission effect may be as high as 1.10. This means that an appreciable percentage of the fissions occur in the U^{238} with neutrons of over 1.1 Mev energy. This high fast fission effect results in a conversion ratio that may be between 0.9 and 1.0. In reactor designs evolved to date, the lowest H_2O/U volume ratio is 2.0 to 3.0 which will give a fast fission effect of about 1.05, see Figure 6.*

* BNL-Log No. C-7580, "Lattice Parameters for Light Water, Slightly Enriched Uranium Reactors." (Nov. 3, 1953)

Figure 6. Fast Fission Effect for Light Water Moderated Reactors

U^{233} may be used in light water reactors with Th as the source material.

The first of the five authorized reactors under the Five Year Plan for developing power reactors is a full scale, not a pilot scale, pressurized light water reactor. It appeared to be the only reactor which, at the time the decision was made, could be built in full scale without the necessity of going through the pilot plant stage. In addition, some of the problems involved in its design such as large pressure vessels could best be solved by a full scale model. In this reactor, H_2O at high pressure is pumped through the reactor and a boiler to produce steam.

The development and design of this reactor was begun as a ship reactor and the work was continued at Westinghouse as a logical extension of their work on the Submarine Thermal Reactor and the ship reactor. The Duquesne Electric Company will supply the turbine and electrical equipment and purchase the steam produced in the reactor heat exchangers. The reactor will be built at Shippingport, Pennsylvania. Steam at 600 psig will produce 66,000 kw or more of electrical energy from the 300,000 kw of heat. This reactor will use slightly enriched U-Zr-clad fuel plates.

The steam produced will be saturated. In order to prevent excessive erosion in the turbine, it will be necessary to remove this steam at 50 psig or so from the turbine for removal of the water. Three reheaters will be used.

It should be noted that, even though the pressure of the inlet steam to the turbine is lower than in regular central station practice, the steam may be more corrosive to the turbine blades due to the lack of superheat.

The General Electric Company has announced interest in building a pressurized light-water, graphite-moderated reactor at Hanford. This is an extension of their experience in operation of the Hanford Reactors. Actually, if only a small fraction of the heat produced in the present production reactors could be converted to low pressure steam, it would add significantly to the Northwest power resources.

The Fluor Corporation has been carrying out the design of a power reactor based on the use of Al-clad fuel elements. Their design has been stated to be low enough in cost to be nearly competitive with fossil fuel-fired steam plants. They estimate a plant to produce 1,000,000 kw of heat and 200,000 kw of electrical energy could be built for \$170 per electrical kw or \$34,000,000.

Army Packaged Power Reactor

The Government has awarded a contract to the American Locomotive Company to develop a "packaged power reactor." This reactor was designed by Oak Ridge National Laboratory for construction at isolated posts. The advantage of a reactor over a conventional oil fired and diesel power plant is the saving in fuel shipping charges. Enriched U^{235} fuel elements for one year's operation, clad and ready for use, will weigh only a few hundred pounds at the most. The reactor will produce about 10,000 kw of heat.

Heavy Water Reactors

There are many advantages of heavy water when compared with light water for moderating a reactor. Some of these advantages are:

- (a) Low neutron absorption cross section, as indicated in Table III of Lecture 1.
- (b) Good moderating properties.
- (c) Longer neutron life. The long neutron life is an important advantage from the operation viewpoint in respect to safety.
- (d) Use with natural U. The neutron absorption is low enough that a natural U- D_2O reactor will go critical.
- (e) High conversion ratio. Conversion ratios over 1.0 are possible.
- (f) In addition, heavy water has many of the advantages of light water, including inherent safety.

The disadvantages in the use of heavy water are real. Some of these are:

- (a) Cost. Heavy water occurs to the extent of 0.015% in ordinary water. Its separation results in a high cost.
- (b) Operation of a heavy water reactor requires extreme care not to loose or contaminate the heavy water.
- (c) D_2O has the disadvantages of light water such as high chemical reactivity, high vapor pressure, heavy equipment required at high temperatures, and partial decomposition in a reactor.

The above factors balance one another fairly well so that the use of D_2O appears at present to be competitive with graphite and H_2O for reactor moderators. If further reductions in the price of D_2O occur in the future, the use of D_2O may become widespread. In those countries which do not have U^{235} enrichment plants, the use of D_2O has the overriding advantage that they are independent of outside sources once a reactor is in operation. The cost of construction of a heavy water plant is only a small fraction of

the cost of an U enrichment plant. Small scale heavy water plants are practical. Several of the European countries have built natural U-D₂O reactors and others are planning to build such reactors. India has just purchased 10 tons of D₂O for use in a research reactor.

CP-3

The first heavy water reactor was built at Argonne during the War under the direction of Dr. Walter H. Zinn. The first step was the measurement of the cross sections of deuterium and oxygen in the original graphite reactor. These measurements confirmed the low cross sections of both materials. During the fall and winter of 1943 - 1944, the design and construction of the heavy water reactor were carried out. D₂O was made available and the reactor became critical on May 15, 1944. The reactor was designated CP-3 or Chicago Pile No. 3.

The general features of the reactor are illustrated in Figure 7. The reactor consisted essentially of a cylindrical tank containing D₂O in which Al-clad U rods were hung. The D₂O was circulated through an external heat exchanger and the system's D₂O was blanketed with He. The He was circulated through a catalytic O₂-D₂ recombination chamber and the recombined D₂O was returned to the reactor.

The reactor tank of 2S Al was about 5-1/2 ft diam by 9 ft high. The top of the tank was expanded to form a shoulder for support of the cover plate and space for the pipe connections. The fuel rods, He circulation pipes, one of the D₂O pipes, and a central thimble were supported or connected through the stainless steel cover plate by means of Al liners. Several of the fuel rods contained thermocouples and these were also connected through the top plate, see Figure 8.

The original reactor contained 121 natural U rods, 1.1 in. diam by 6 ft long, with a weight of approximately 3 tons of U. By 1950, the cladding on a few of these rods had failed. The reactor was reloaded at that time with Al-covered, enriched U rods made up of 2 weight per cent U²³⁵ in Al. These rods were 7/8 in. diam and 5.5 ft long. With the enriched fuel, the reactor was designated as the CP-3' reactor.

A graphite reflector and concrete shield surrounded the reactor tank. A sealed cover plate and removable shielding were placed above the reactor. Nine experimental beam holes and a thermal column were constructed through the shield. The reactor is illustrated in Figure 9.

The reactor was operated at 275 kw heat with a maximum thermal flux of 3.4×10^{12} and a maximum fast flux of 3.5×10^{11} .

Figure 7. Isometric Sketch of CP-3

Figure 8. Top of CP-3 Showing Fuel Rods, Central Thimble and Connections

Figure 9. View of CP-3' in Use

The reactor was controlled by movement of Cd rods in the reactor tank. Large changes in reactivity were compensated for by adding enriched U or Cd slugs to the core. Two safety circuits, which received current from independent ionization chambers located in cavities in the concrete shielding, shut the reactor down in case of a power surge. A third ionization chamber supplied current to a rate-of-rise meter or a "Pile-Period" meter. This circuit incorporated a recorder and sensitive relays to "Scram" the reactor if the rise of power on start up was too rapid. A third power level trip, safety circuit, utilized current from still another ionization chamber. This chamber served an additional function of supplying current to a galvanometer and photo cell circuit for automatically maintaining the reactor at a given power level. A hand-operated dump valve and a storage tank were provided for emergencies. Significantly, the D₂O was never dumped from the reactor by use of this dump valve until May 15, 1954, when the reactor was removed from service because the Palos Park site was being abandoned.

NRX

For several years the NRX reactor at Chalk River, Canada, had the honor of having the highest flux and being the most advanced development reactor in existence. The reactor was designed by the Canadians with help from the British during the last year of the war and the years immediately following the war.

The reactor was designed for a flux of 6×10^{13} at the normal operating power of 10,000 kw. The reactor was placed in operation in 1947. A schematic diagram is shown in Figure 10. The reactor tank is an Al calandria about 8 ft diam by 10 ft high. One hundred seventy-six natural U rods, 1.360 in. diam, Al-sheathed, are hung in this calandria and cooled by pumping river water over the rods through a 0.10 in. annulus. Outside of the cooling rods are the calandria tubes holding the heavy water in the Al tank. Surrounding the Al tank are many experimental facilities and shielding. Both above and below the calandria are placed heavy thermal shield plates.

Control rods and the level of the D₂O are used to maintain constant power level.

The choice of the Chalk River site, 125 miles northwest of Ottawa, was made on the basis of isolation in case of failure of the reactor. An accidental power surge occurred in December 1952, causing failure of the Al cladding of about 10% of the U rods. Part of the U was melted and part was oxidized or corroded away by the water. Ten thousand curies of radioactivity were carried into the basement and some gaseous reactivity escaped. The ceiling, walls, and floor of the main building were contaminated to about 50 mr/hr and the basement water left a contamination of 10 r/hr. Immediate steps to recondition the reactor were started and successfully completed by February 1954. Since then, the reactor has been in continuous operation, at over 20,000 kw of heat.

Figure 10. Diagram of Chalk River Reactor (NRX)

Several countries have D_2O , natural-U research reactors similar to CP-3 in principle. These are France, Norway, Holland, and Sweden. Other countries are planning such reactors.

CP-5

The Argonne Experimental Reactor, CP-5, is a D_2O cooled and moderated reactor which operates at 1000 kw using enriched U^{235} in Al-clad fuel plates similar to those described for the MTR. The core is a rough cylinder 2 ft diam by 2 ft high. Ten fuel elements are ordinarily used with a critical mass of about 1500 grams.

The D_2O is forced upwards through the fuel elements at 5 ft/sec. The water spills out into the moderator space in a 6-ft diam Al tank and is drawn off for cooling and recirculation.

The control system is of interest in that four signal arm rods are used at the top of the D_2O above the fuel for shim safety. These rods tend to suppress the neutron flux in the top of the moderator but do not change the flux appreciably at the center and bottom of the core. These safety rods are spring loaded so that they drop into the core rapidly in case of power surges or other rapid shutdowns. A single control rod is used for fine control. The reactor is inherently safe since boiling of the water in the fuel elements would reduce the reactivity sufficiently to shut the reactor down. A quick opening valve is provided for dumping the D_2O from the reactor.

About two feet of D_2O surround the core on all sides. The Al tank is 6 ft diam and the water level is maintained at 6-1/2 ft depth during operation. The flux in the core is about 2×10^{13} at 1000 kw. The reactor cross section is shown in Figure 1 of Lecture 2. Eight beam holes extend into the reactor tank. Four of these are provided with heavy eccentric doors for use in changing the plugs.

The Al tank is surrounded by two feet of graphite which extends outward to form two thermal columns. Outside of the graphite is 3-1/2 in. of Pb and about 5 ft of iron aggregate concrete shielding.

Heat exchangers and auxiliary equipment are placed in the basement. The heat is transferred to H_2O in an exchanger and dissipated in a cooling tower. Potentially, the reactor can be operated at higher power levels as desired by addition of more heat exchange equipment.

The reactor is housed in a concrete building which is sealed to provide retention of the fission products in case of release of some of these materials by accident. This building is illustrated in Figure 11.

Figure 11. CP-5 Building

LECTURE 4

HANFORD AND SAVANNAH RIVER REACTORS
GRAPHITE REACTORSReferences:

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Introduction

Detailed descriptions of the Hanford and Savannah River Reactors have not been made public. The Hanford Reactors are natural U-fueled, graphite-moderated reactors of large size, producing hundreds of kw of heat. The Savannah River Reactors are natural U, D₂O-moderated reactors. Both projects were designed, built, and placed in operation by the E. I. du Pont de Nemours Company with assistance from many industrial firms and research and development organizations.

Hanford Reactors

The choice of H₂O-cooled, graphite-moderated reactors for construction during the War was natural, since this was the only combination of materials that could be assembled quickly and that would operate. Development work was carried out primarily at the Metallurgical Laboratory at The University of Chicago. The du Pont Company carried out a great deal of development and test work in their own laboratories and also in the Hanford plants themselves. The X-10 or Oak Ridge Laboratory (then known as the Clinton Laboratory) was built to provide some Pu and serve as a pilot plant for the Hanford separation plants. Other groups assisted with the production of the U and graphite for the reactors as well as to supply materials and equipment. The Mallinckrodt Chemical Company and the carbon companies made very noteworthy contributions.

The reactors were built on the Columbia River because of the isolation and supply of cold, high purity cooling water. Construction of the first reactor was started June 7, 1943, and the third reactor placed in operation early in 1945.

The reactors are similar to the Oak Ridge Graphite Reactor discussed below, except that they are larger and light water cooled. The U fuel rods are clad with Al. One of the reactor buildings with its associated water treatment and steam plants is illustrated in Figure 1.

At the end of the War, the General Electric Company took over operation of the Hanford Project including operation of the reactors and associated plants. There has been continuous expansion of the number of reactors and the chemical separation plants since 1949. Large, well-equipped development laboratories have recently been built.

Savannah River Reactors

In 1950, the du Pont Company was asked to design and build a group of reactors which, with their associated fuel fabrication and chemical plants, were constructed at the Savannah River site near Augusta, Georgia. Considerable development work was carried out at Argonne and other sites. D₂O separation plants were built at Dana and Savannah River. The du Pont Company is continuing to operate the D₂O and reactor plants.

Both the Hanford and Savannah River Projects were very large and rate among our largest industrial plants both in size and cost.

Graphite Reactors

Several reactors have been built based on the use of graphite as the moderator and natural U as the fuel. These include the first operating reactor, the Oak Ridge Graphite Reactor, the Brookhaven Research Reactor, the Hanford Reactors, and the British Reactors. A graphite, natural U reactor is under construction in Belgium. A sodium-cooled, graphite-moderated reactor, using slightly enriched U, is being built at Santa Susanna, California.

CP-2

The first controlled self-sustaining nuclear reaction occurred in a "pile" of graphite and natural U on December 2, 1942, under the direction of Dr. Enrico Fermi at the University of Chicago. This reactor was soon disassembled and set up about 25 miles southwest of Chicago in a forest preserve site near Palos Park.

Figure 1. General View of Hanford Reactor Buildings.

The pile as set up at the Palos Park site was named CP-2 or Chicago Pile Number 2. It contained about 10 tons of U metal, 42 tons of uranium oxide, and 472 tons of graphite in a pile 20 x 22 x 21 ft. Five feet of concrete shielding was placed on all sides and the top as illustrated in Figure 2. The flux was about 10^{10} at a maximum power of 100 kilowatts. As no cooling was provided, steady operation was limited to 20 kw.

The reactor was operated until May, 1954, when it was permanently shut down and the site abandoned. It is hoped that the National Park Service will declare the site of CP-2 and CP-3 a national monument, so these historical reactors will not have to be dismantled.

Oak Ridge Graphite Reactor

The Oak Ridge Graphite Reactor, or X-10 Reactor, was built during the War to provide some Pu for use in the pilot plant for chemical separations. The reactor was air-cooled and, therefore, not a pilot plant for the water-cooled Hanford Reactors. The reactor consists of a 24 ft cube of graphite through which there are 1248 fuel holes with 8 in. between centers. The natural U fuel slugs are 1.1 in. diam by 4 in. long and canned in Al. 39 to 54 slugs are used in each fuel channel. Outside of the graphite there is the usual concrete biological shield.

100,000 cfm of air are drawn through the reactor. The maximum fuel jacket temperature is 245°C at a 3800-kw power level. The maximum flux is 1.2×10^{12} . The air comes into direct contact with the graphite and in case of a slug failure may pick up fission products and Pu. For this reason, it is necessary to filter the air through roughing and fine filters before discharge to the stack.

The air is drawn in between the graphite and the front shield, through the reactor and out into a rear plenum. The fuel slugs are loaded through holes in the front shield by removal of wood plugs from these holes. Tubes extend almost to the graphite to carry the slugs into the graphite holes. At the rear, the slugs drop through the plenum chamber into a canal.

This first really good sized reactor is still in very active operation as a producer of most of the radioisotopes distributed in this country and as a research and development tool. It has operated about 90% of the time since it was built. The reactor was designed to operate at 1000 kw but it has been operated at about 3800 kw since 1944.

Figure 2. General View of CP-2

Brookhaven Reactor

The Brookhaven Reactor was built from graphite and natural U in order to have a large volume of experimental space and a large number of experimental facilities. Essentially, it is a larger and higher power version of the Oak Ridge Graphite Reactor and it operates at 30,000 kw. The graphite is a 25 ft cube compared with the 24 ft cube of the Oak Ridge reactor. The outside dimensions are 38 x 55 ft. The fuel rods are clad with Al tubes having several ribs for increased heat transfer surface. The Al is not bonded to the U, but the space is He-filled and the pressure of the He is continuously checked for leaks.

The air is drawn into the reactor through a central plenum and discharged on two sides at 300°F. After leaving the reactor, the air is cooled by water sprays and blown up a 350-ft stack. Due to the large amount of A^{41} formed, very careful surveys of activity in the surrounding areas must be made continuously.

The top shield of the reactor is made of removable slabs of heavy concrete to permit shielding tests on a large scale. Many holes penetrate the shield and graphite for use primarily in research and isotope manufacture. The control rods are horizontal and are placed on one corner to limit their interference with the test facilities as much as possible. Heavy steel flywheels are kept in motion at all times to provide inertia energy to drive the control rods into the reactor in case of a power failure.

SGR

A logical extension of the Hanford experience with water-cooled, graphite-moderated reactors is to increase the temperature by using high pressure water or liquid metal cooling. Designs of such reactors have been prepared by North American Aviation, Inc. and the Monsanto Chemical Union Electric industrial team (see Lecture 6).

North American Aviation has been developing this reactor concept for several years. Their design was chosen for one of the pilot plants to be built as a part of the Government's Five Year Program. The reactor is now under construction at Santa Susanna, California, about 25 miles north of Los Angeles.

The reactor will use slightly enriched U rods, clad with stainless steel, and Na cooling. A number of small rods will be clumped together to form a fuel assembly. Arrangements will be made to clad the graphite moderator in large sections for replacement in case this is required. The Na will leave the reactor at 875°F.

The reactor will be placed in a tank to prevent loss of coolant in case of power outages with danger of melting the fuel elements. Loading and unloading will be carried out through the top shield by means of a heavy coffin.

The heat will be transferred to NaK in the primary heat exchangers. The heat will then be dissipated to air in a NaK to air heat exchanger. Thus, no attempt will be made to produce any electrical power even though 7,500 kw could be produced from the 20,000 kw of heat produced. The reactor is illustrated in Figure 3.

The sodium-cooled, graphite-moderated reactor has the advantages of higher surface temperatures of the fuel elements due to lower corrosion rates and thus high over-all thermal efficiencies due to the high coolant temperatures, high heat transfer rates, and low pressures in the primary reactor system. The disadvantages are the increased U enrichment required, the impossibility of operating on natural U, low heat capacity of the coolant, the additional heat transfer and pump equipment required, the difficulties of removing Na-coated fuel elements from the reactor and cleaning them before immersion in the canal, the induced radioactivity of the Na, and the non-compatibility of Na and NaK with H_2O and O_2 .

The General Electric Company has stated that it is interested in building a pressurized water, graphite-moderated power reactor as a part of the Hanford project. This suggestion has not yet been given favorable action by the Government.

Figure 3. Sodium Reactor Experiment

LECTURE 5

HOMOGENEOUS REACTORSReferences:

"Criticality of the Water Boiler and Effective Number of Delayed Neutrons," F. de Hoffman, AECD-3051, December 8, 1944.

"Distribution and Power Measurements in the Water Boiler," L. D. P. King, R. E. Schreiber, AECD-3054, October 19, 1944.

"An Enriched Homogeneous Nuclear Reactor," AECD-3059, January 25, 1951.

"High Power Water Boiler," F. L. Bentzen et al., AECD-3065, September 19, 1945.

"The Homogeneous Reactor Experiment," S. E. Beall, C. E. Winters, Chem. Eng. Prog., 50: 256(1954).

"Liquid Metal Fuel Reactor Systems for Power," C. Williams, F. T. Miles, Amer. Inst. Chem. Eng. Mon. 11: 245(1954).

"Liquid Metal Fuels and Liquid Metal Breeder Blankets," R. J. Teitel, D. H. Gurinsky, J. S. Bryner, Amer. Inst. Chem. Eng. Mon., 13: 11(1954).

Introduction

A homogeneous reactor may be defined as any reactor in which the fuel is dissolved in or carried as a slurry by the coolant. Homogeneous reactors may be fast or thermal.

There are a very large number of reactor systems that could be considered. These include water solutions of salts, hydroxides, acids, etc., fused mixtures of salts, hydroxides, acids, fused metals, etc. The only moderator that may exist as a fused fluoride, liquid metal, or hydroxide is beryllium. All hydroxides and acids, of course, contain hydrogen. Most nonaqueous systems suggested to date either have high melting points or low U solubility. As U is soluble as the nitrate, sulfate, phosphate, and carbonate, several aqueous or acid solutions are feasible.

Many advantages are claimed for homogeneous reactors. Some of these appear to be real. These advantages are:

A. Simplified fuel preparation. The fuel is used as a sulfate, phosphate, oxide, or metal and the solutions or slurries are easily and inexpensively prepared.

B. Chemical costs are less. The irradiated solutions or slurries are readily prepared for chemical treatment. Reactor solutions in D_2O require complete evaporation and drying of the residues in order to recover the D_2O . The residues are then dissolved in H_2O for chemical separation processes. At the end of the chemical processing, the steps of evaporation, drying, and solution in D_2O must be followed.

C. Xenon is easily removed. By continuous degasification, the xenon and much of the iodine may be removed during operation so the loss of neutrons to xenon burnout and the xenon problem are removed.

D. Simplified reactor systems can be used. Since the reactor tank does not have to be opened for loading and unloading and no fuel elements need be handled, the tank may be built with a very small top opening and the building can be much more compact.

E. Materials of construction can be selected primarily on the basis of corrosion and creep resistance rather than neutron properties, since there need be no solid materials in the reactor tank except the control rods which are themselves poison rods.

F. Flexibility of operation. Most homogeneous systems have very large negative temperature coefficients and the plants may be operated entirely on the concentration of the solution and temperature coefficient. This means that to start up sufficient enriched fuel is pumped into the slurry or soup that the reactor will go critical. As the flux and temperature rise, the power will level off at the proper temperature, and the entire plant will then operate on the power demand. More power output lowers the temperatures which increases the reactivity, flux, and power until a new balance is reached.

G. High power densities may be obtained. The power density is limited by the rate of pumping and heat capacity of the soup rather than by heat transfer as in the case of heterogeneous reactors.

H. Radiation damage can be held to that involved in the core sphere in core-blanket reactors.

In addition to the advantages, there are several disadvantages:

A. Only part of the fuel is in the reactor where it can contribute to the power production. Perhaps half of the total solution or slurry will be present in the external lines, heat exchangers, and pumps. In heterogeneous reactors, an additional fuel loading may be required in the processing plants for recovery of the used fuels and in preparing new fuel elements or in storage awaiting loading.

B. Delayed neutrons will cause the external system to become radioactive, making maintenance very difficult. This is a very serious disadvantage.

C. Corrosion may be greater in a salt, acid, or liquid metal system than in a heterogeneous system. Since corrosion is a limiting factor in heterogeneous as well as homogeneous reactors, comparisons between the two systems must be made with care.

D. Gas formation may be greater in water systems than in heterogeneous systems, since the water is mixed with salts, acids, etc.

E. Leaks in the coolant heat transfer system are much more serious than in the corresponding heterogeneous systems since U, Pu, and fission products may be lost and contaminate the heat exchange areas.

F. Slightly higher enrichment is required due to the resonance absorption and lower U density. No fast effect is present. The resonance absorption forms Pu from U^{238} , so the over-all conversion ratio is not altered.

There have been at least three homogeneous reactors built and operated more or less successfully. Two of these were very small, the Los Alamos and North Carolina State College Reactors, and the other was experimental in size, the Oak Ridge Homogeneous Reactor Experiment. There have been several others considered, and suggestions for power plants have been based on several types. Brookhaven has carried out extensive development work over several years on a liquid metal system.

Los Alamos Water Boiler

The Los Alamos Water Boiler was built during the War. It was under construction from August, 1943 to May, 1944. The water boiler consisted of a steel sphere, 1 ft ID, containing cooling coils. The soup was made up of UO_2SO_4 dissolved in ordinary water. The critical mass was 565 gm U^{235} . A power of 1 kw was used at a flux of 5×10^{10} with a minimum of cooling elements.

By December, 1944, the reactor was revised to give a higher power. It operated as a boiling reactor at 10 kw at a flux of 3×10^{11} . A number of experimental facilities were built in.

A foot of BeO surrounds the core followed by several feet of graphite.

The North Carolina reactor is similar in size and manner of operation.

HRE # 1

The Homogeneous Reactor Experiment No. 1 was started in 1948 at Oak Ridge National Laboratory. It was essentially a small circulating $\text{UO}_2\text{SO}_4\text{-H}_2\text{O}$ reactor which operated at 1000 kw with production of 100 to 200 kw of electrical energy. The reactor was designed to:

- A. Demonstrate the operational feasibility of a circulating fuel at a fairly high power level.
- B. Study radiation decomposition and corrosion behavior in a homogeneous chemical reactor at flux densities similar to those encountered in a large production reactor.
- C. Demonstrate electrical power production.
- D. Obtain operational experience in the handling of a homogeneous circulating reactor.

One of the problems considered serious was the gas evolution. For this reason a very extensive program of tests on various core shapes was carried out. It turned out that gas separation was not difficult and gas bubbles not harmful, so the problem has become a normal engineering design problem.

The core itself was a stainless steel cylinder containing 50 liters of 30 grams enriched UO_2SO_4 per liter in light water.

Control of the reactor was carried out by changing the concentration of the solution by evaporation to concentrate the solution to increase power and by dilution to decrease power. Provision was made for movement of control plates in the reflector and for changing the reflector level. Actually, other than dilution changes in startup and shutdown, the negative temperature coefficient took care of control. $\delta k/^\circ\text{C} = 0.001$ which was about 100 times the amounts normally present in heterogeneous reactors. The mechanical control was not used.

Operation of the reactor was remarkably stable and straightforward. Since the reactor system had been designed on the basis of minimum cost, everything was kept compact and means of cleanup were limited. Many devices were included which were not needed. When leaks occurred, they were very troublesome as the hot soup soaked into the concrete, making recovery and decontamination very difficult and costly.

The reactor went critical on April 17, 1952, and reached full power on February 24, 1953, at 1000 kw. The reactor has been shut down and dismantled.

ISHR

Oak Ridge National Laboratory is developing a larger version of the Homogeneous Reactor to be known as the Intermediate Size Homogeneous Reactor. This is planned as a two-core type of reactor in which the second core or blanket surrounds the central core with the two separated by a Zr sphere. The reactor is planned to produce 65,000 kw of heat and 16,000 kw of electricity. The present concept makes use of UO_2SO_4 in D_2O for the central core.

At present a smaller experimental reactor of this general design, HRE-II, is being built to check out the design. It is obvious that the ISHR will not be started for two years or so.

Liquid Metal Fuel Reactors

All reactor concepts which make use of a solution or suspension of the fuel in a liquid metal are included in liquid metal fuel reactors. Such reactors may be fast or thermal. They have many advantages and disadvantages. If a completely satisfactory reactor system in respect to corrosion and solubility of fuel could be developed, a liquid metal fuel reactor would promise to produce economical power.

The liquid metal fuel reactor has high temperature operation at low pressure, high burnup, removal of gaseous fission products, high heat transfer rates, and other advantages of the homogeneous reactor. Possibility exists of inexpensive chemical processing of the fuel.

The main difficulties are low solubility of U in a liquid metal of low cross section and corrosion. Solubility of U in Bi increases with temperature as follows: 0.5 w/o at 480°C , 1.0 at 550, and 2 at 640°C . This amount of solubility can be utilized by use of external blankets in thermal reactors or possibly in fast reactors.

The use of slurries is feasible from the carrying standpoint of the liquid metal but the danger of separation exists. Extensive experimental work is required before a suitable design will be demonstrated.

The construction of a liquid fuel reactor was not included in the Five Year Program because, at the time of the decision on what reactors would be built, the liquid metal fuel reactor appeared to be a longer range project. Rapid progress in developing suitable systems has been made recently.

Fused salt systems are possible but no practical system for use in a civilian power reactor has been suggested.

LECTURE 6

INDUSTRIAL STUDY GROUP REPORTSReferences:

"Reports to the U.S. Atomic Energy Commission on Nuclear Power Technology," Commonwealth Edison Public Service, Dow Chemical Detroit Edison, Monsanto Chemical Union Electric, Pacific Gas & Electric - Bechtel; Gov. Print. Off., Washington, D.C.

Introduction

In 1951, the Commission authorized four groups of companies to make a survey and study of reactor technology to appraise the prospect for private industrial participation in joint production of electric energy and fissionable materials from reactors. The studies covered both technical and economic features. The four groups carried out studies over a period of one year and submitted reports in 1952.

The Commission studied these reports but took some time to decide just what action should be taken. Actually, no real action was taken until after passage of the Atomic Energy Act of 1954 and an evaluation of what could be done under that act. The recent announcement of the desire to join forces in the so-called Reactor of the Year Plan is the first concrete result of these studies. Under this plan it is possible that several full-scale reactors will be built.

After submittal of their reports, the PG & E Bechtel group dropped active work but continued interested at the administrative levels. The Monsanto Union Electric group dissolved but the Monsanto Company continued a small group in study and design work. This group is still active. The Dow Detroit Edison group continued active both in studies and development work. The group was enlarged by addition of some 25 companies but the Dow Chemical Company withdrew from the group. The Commonwealth Edison Public Service group has continued studies of various types of reactors. Several other companies including American Gas and Electric Co., Bechtel Corp., Pacific Gas & Electric Co., and Union Electric Company have joined this group now known as the Nuclear Power Group.

Several groups have announced definite specific plans for construction of power reactors.

Dow Detroit Edison Reports

The Dow Chemical Company Detroit Edison group submitted a non technical report, all of which was published in the declassified version of the reports.

Their report listed the important specifications for an economical power reactor as:

A. It should use low cost fuel such as depleted U or Th, i.e., it should be a breeder and produce more fuel than consumed,

B. The reactor should have a high breeding gain, e.g., it should be a fast reactor.

C. The reactor should operate at high temperature and reach 500 to 600°C in the core.

D. The reactor should permit integration of liquid metal or fused salt extraction permitting fast, low cost processing and recovery of the fissionable materials.

E. The reactor should require a minimum exclusion area. Continuous separation and segregation of the fission products indicate the possibility of small areas.

F. The reactor should use uncanned fuel, preferably mobile or fluid fuel.

G. The reactor should be inherently self-regulating.

On the bases stated, the group carried out design and development work on a liquid-metal cooled, fast reactor.

Their report pointed out that power is desirable, but power production interferes with high plutonium production, so no power would be developed.

They suggested building the reactor on a barge or ship and anchoring some miles off shore in order to obtain a suitable exclusion area.

The Dow Detroit Edison group has been working in their own laboratories and with a number of subcontractors on development of fuel elements and other problems. The Government has been spending about \$700,000 yearly on the Brookhaven liquid-metals work. In 1953, the Dow Detroit group spent about \$250,000, and presumably more in 1954, on reactor work. The group holds frequent meetings with AEC representatives as sort of information meetings.

PG & E - Bechtel

The PG & E - Bechtel group chose the Na-cooled, fast heterogeneous reactors as most promising for plutonium and power after a study of water-cooled and Na-cooled reactors. They looked at other types only briefly.

The group concluded that a dual purpose reactor would be feasible; the fast breeder is more attractive; the dual purpose reactor is economical with weapon-grade Pu prices; the reactors would not be economical for power only until technological improvements are made to reduce the capital and operating costs of the plant, including fuel preparation and chemical separation; a water-cooled or fast breeder reactor can be built, and the prime risk on continued expansion of the Pu facilities should remain with the Federal Government.

Since there is not much difference in their report from other studies, no details of the group designs are given here.

Monsanto - Union Electric

This group based their studies on an extrapolation of Hanford experience on the premise that this would lead to an operating plant in the shortest possible time. While the group recognized many development and operational problems, they believed they could be solved in a reasonable period.

The group examined the graphite-moderated, sodium-cooled reactor using stainless steel cladding and tubes with slightly enriched U. Large reactors at high power levels, 10^6 and 3×10^6 kw of heat, were considered.

Their report presents the design in some detail. They assumed that reloading during operation was a requirement. The fuel elements were to be hollow and clad inside and outside by stainless steel tubes, with Na used as the bonding agent.

In the design of the 10^6 kw reactor, the Na enters the reactor at 300°F and leaves at 650°F .

After submission of their report, the Monsanto group continued in active design. The group looked at the sodium-cooled, graphite-moderated reactor and concluded that the reactor type would be the one they would choose for power only design and, more importantly, that the sodium-cooled, graphite-moderated reactor might be economical as a power producer in the very near future.

Several changes in their basic design were made. First, Zr was considered in place of s.s. as the cladding for the fuel. Second, Zr canning of the graphite was considered as a means for permitting removal and replacement of the graphite in case of excessive damage either by irradiation or Na.

Commonwealth Edison - Public Service Group

The Commonwealth Edison Public Service group studied two reactors in their original study. These were the He-cooled and the D₂O reactor for Pu and power. After submitting their initial report, the group studied the light water and then made briefer studies of the Na-cooled, graphite-moderated, Knolls Fast Power Breeder, aqueous homogeneous, and boiling reactor concepts. The initial work was directed at reactor types using natural U, but later reports included slightly or fully enriched types.

The gas-cooled reactor does not look promising because of the high pumping costs. In the Commonwealth design for a 350-Mw (heat) reactor, the gross electrical power output was 61,700 kw, but auxiliary power is 12,000 kw of steam driven and 15,000 kw electrically driven equipment, giving a net electrical of 45,000 kw. The reactor which they considered was a graphite-moderated, natural-U, He-cooled device with the He at 10 atmospheres. The U was in the form of a star-shaped slug with 8 thin points or wings.

The economy could be improved by higher pressures and higher temperatures, but the coolant temperature rise used was 400°F to 700°F with a circulation rate of 3.2×10^6 #/hr. A He-to-boiling water heat exchanger was used with a conventional turbine. A gas turbine was not studied so far as is known. In any case, due to the low heat transfer coefficients, gas coolants will be expected to be inefficient until much higher temperatures can be tolerated.

The liquid-cooled reactor was a D₂O-cooled and moderated, natural-U pressurized reactor designed for reloading during operation. The reactor was designed for 1064 Mw of heat, coolant velocity of 25 ft/sec, maximum Zr-clad temperature of 465°F. The flux was a maximum of 1.1×10^{14} . Gross electrical power output was 225,500 kw or 211,500 kw net. Steam pressure was 175 psia and 371°F.

Light Water Reactor

The Commonwealth group next studied the H₂O-cooled and moderated reactor. While the design was interesting, again we do not have time to go into details. Compared to the D₂O reactor, the cost was reduced from \$335 to \$298/kw exclusive of D₂O costs.

Additional Studies

The group has reviewed the SGR, FBR, liquid homogeneous, and boiling reactors. They list them tentatively for power only in order as:

FBR

SGR

Homogeneous or Boiling

Foster Wheeler Pioneer Service & Engineering

The group reviewed the design of all the various reactors that have been suggested and compared the results on the basis of a different type of financing in which approximately 10% return on the investment was added. The net result was that the costs of all the plants went up considerably. They concluded that none of the reactor systems could produce economical power today. They also concluded that the reactors that have the greatest possibility of economic power production are the thermal, U^{233} , circulating fuel type and the fast Pu breeders.

They suggest a fluidized solids reactor.

Their studies were based on somewhat different bases than others, e.g., the requirement of private risk capital. They also assumed that the supply of fuel is adequate.

Consolidated Edison Company

On February 10, 1955, the Consolidated Edison Company of New York announced that it planned to construct a power reactor near Peekskill, New York, to supply 100,000 to 200,000 kw. The cost is estimated as \$ 30,000,000 to \$40,000,000. It is understood that the Babcock and Wilcox Company will design the reactor.

LECTURE 7

BOILING REACTORSReference:

- "U. S. Atomic Energy Commission Seventeenth Semiannual Report,"
Gov. Print. Off.
"Evaluating Reactor Hazards from Airborne Fission Products,"
R. B. Mesler, L. C. Widdoes, Nucleonics, 12: 39, Sept. (1954).

The pressurized water reactors discussed in a previous lecture are designed to operate at such pressures that no boiling will occur in the reactor cores. For several years following the War, it was feared that bubble formation in the core would change the density and therefore the reactivity of the reactors. This, in turn, might lead to fluctuations and difficulties in control.

In 1952, Mr. Samuel Untermyer pointed out that formation of steam in a reactor core might lead to increased safety in operation since the formation of steam would always lead to increased neutron leakage and decreased reactivity - thus tending to shut the reactor down. If the steam could be used directly, the heat exchanger and primary circulation pumps could be eliminated. Also, the steam would be available at a higher temperature since no temperature drop across a heat exchanger would be required. Since the steam is always radioactive due to the $O^{16}(n,p)N^{16}$ reaction and radioactivity from the impurities in the water, the turbine and condenser would become gamma and beta ray active.

In order to determine what would happen, Argonne built a small experimental reactor at Arco in 1953 known as the Boiling Reactor Experiment or Borax I. This was a light water cooled and moderated, fully enriched, non-pressurized reactor. The fuel elements were identical with the MTR fuel elements. Conditions were imposed that normally would be expected to result in an excessive power rise. When excess reactivity was added very quickly, rapid power excursions occurred, but these were quenched by the decreased reactivity resulting from the increased neutron leakage due to the increased steam formation. Extrapolations of the test data indicate that instantaneous additions of excess reactivity up to 3.8% would be self-limiting without serious damage when the water in the reactor is at its boiling point. With the water in the reactor at room temperature, instantaneous additions of excess reactivity up to 1.9% would produce similar results. With 3.8% excess k , the power level of Borax-I rose by a factor of e every 0.0024 second and with 1.9% excess reactivity the power rose a factor of e every 0.0065 second.

A final short period excursion was performed on July 22, 1954. In this power excursion sufficient reactivity was added to permit the power to rise very rapidly to a high level. This excursion resulted in destruction of the reactor core and some associated equipment. The possibility of destruction of the core was considered in planning the reactor - in fact this was the reason for locating the reactor at Arco and placing the control instruments at a distance from the reactor. Care was taken to make certain that appreciable amounts of fission products would be avoided.

No reactor would ever be designed to permit additions of over 0.75% excess reactivity in short periods. Excess reactivity of 0.75% corresponds to the delayed neutrons. These experiments then demonstrated that pressurized and boiling water reactors have an inherent safety or self-quenching characteristic. Of course, the data quoted hold for only the particular reactor under test but, qualitatively, it may be said that all reactors cooled and moderated by bulk H_2O are inherently safe.

Immediately after the final power excursion, a second boiling reactor experiment was constructed near the original boiling reactor at Arco. It began operation October 19, 1954. The reactor operates at 6000 kw of heat and 300 psi.

Based on the favorable results with Borax-I, it was decided to proceed with a pilot size boiling reactor as a part of the Commission's Five Year Plan. This reactor, designated the Boiling Experimental Reactor, BER, is being designed by Argonne for construction at Argonne. Sargent and Lundy are designing the building and supporting facilities, while Allis Chalmers is designing and building the steam turbine and electrical equipment. The steam turbine is being designed for a very low leakage rate.

The reactor system is illustrated in Figure 1. As indicated, the reactor will be loaded by removal of the top cover of the pressurized tank and the control rods will be operated from below the reactor. The fuel will be partially enriched U or spiked with fully enriched plates. Steam at 600 psig will be generated in the reactor and led directly to the turbine. The turbine and condenser must be shielded. Since no neutrons will be emitted in the turbine, the only radioactivity present will be that due to the radioactivity carried by the water. Since the steam will be wet as it leaves the reactor, it will be run through a dewatering device. Nevertheless, corrosion of the turbine may be expected to exceed that of turbines operating at higher temperatures with superheated steam. It is probable that the steam will be removed from the turbine for dewatering at about 50 psig and 15% moisture.

Figure 1. Cutaway View of the BER

The reactor building will be a sealed steel shell which is designed to retain the radioactive gases and particulate matter released if there should be an accident with a resulting power surge. The pressure vessel will be lined with concrete to prevent danger of punctures occurring in the steel shell in case of accident.

The Borax II and BER reactors will be used to determine not only the inherent safety of this type of reactor, but also to obtain data on the use of boiling in power reactors. While the boiling reactor is basically simple, it is not certain that the rate of heat production can be sufficiently high to permit economical operation of large sized reactors. These reactors will not be equipped with pumps, so heat production under natural circulation only can be studied. It is believed that the problems associated with operating a turbine with radioactive steam and corrosion products can be solved at reasonable costs.

LECTURE 8

FLUID FLOWReferences:

- "Unit Operations," G. G. Brown and Associates, John Wiley and Sons, 1950.
- "Chemical Engineers Handbook," J. H. Perry, McGraw-Hill, 1950.
- "Liquid Metals Handbook," R. N. Lyon, Navexos, P-733, Government Printing Office.
- "Prediction of Pressure Drop During Forced Circulation Boiling of Water," R. C. Martinelli, D. B. Nelson, Trans. Am. Soc. Mech. Eng., 70: 695 (1948).
- "Thermodynamic Properties of Steam," J. H. Keenan, F. G. Keyes, Wiley, 1936.
- "Heat Transmission," W. H. McAdams, McGraw-Hill, 2nd Ed., 1942.
- "Dimensional Analysis," P. W. Bridgman, Yale University Press, 1946.
- "Mechanical Vibrations," J. P. Den Hartog, McGraw-Hill.
- "Accelerated Cavitation Research," W. J. Rheingans, Trans. Am. Soc. Eng., July, 1950.
- "Reactor Handbook," 1953, Vol. 2, Cp. 1.5.

Units

In fluid flow and in heat transfer, care must be exercised that units are used with the same dimensions. This is not too difficult except in respect to viscosity and force units. In force, we use poundals, which is the energy required to accelerate 1 lb to a velocity of 1 ft/sec in 1 sec. Since mass (or weight) is 32.17 times the force, we frequently have to use the term g in our equations as the conversion factor between mass and force. Weight is never used since it varies with height above sea level.

Viscosity is measured as mass/(length)(time) and is reported in grams/(cm)(sec) as the poise, or more frequently, the centipoise equal to 0.01 poise. In English units, we use the viscosity in pounds mass/(feet)(seconds). This unit has no name. To convert poises to English units it is necessary to multiply by 30.5/454 or 0.0672. Viscosity μ , in

centipoises $\times 6.72 \times 10^{-4}$ = viscosity in English units. For viscosity in English units and hours, we multiply centipoises by 2.42. Some data are presented in Table I.

The density of D_2O is 1.1. In general, we use the common steam tables for D_2O as well as H_2O .

Additional data needed in fluid flow calculations may be found in most of the standard texts on heat transfer.

The discussion presented below is quoted almost without change from Brown, "Unit Operations." Similar derivations are presented in most Mechanical and Chemical Engineering texts, see reference to Bridgman.

Dimensional Analysis

The procedure followed in deriving the equations for friction loss in fluid flow and the equations for expressing heat transfer data is that known as dimensional analysis. Dimensional analysis simply indicates what variables are important and indicates the relationships of these variables to one another. The constants which always occur in dimensional analysis equations must be derived by fitting the equations to the plotted data.

The essential principles of dimensional analysis are:

1. Each and every one of the physical quantities or measurements may be expressed as a product of the powers of a very few fundamental dimensions.
2. Equations of physical quantities are homogeneous in the net dimensions of all additive terms. All terms added together must have the same dimensions when expressed in terms of the fundamental dimensions selected.
3. Any general relation between physical quantities can be expressed in a generalized manner independent of any particular units to involve only the dimensionless products of all the physical variables and the necessary dimensional conversion constants. This principle follows from the others and is known as Buckingham's "pi theorem." It is the key to dimensional analysis.

The energy dissipated in overcoming friction is some function of the properties of the flowing fluid and the confining pipe and their relative motion. The significant properties of the pipe may be its length L , its internal diameter D , and the roughness or depth of its surface irregularities F . The properties of the fluid are its mass m , its density ρ , and its viscosity μ .

Table I

PROPERTIES OF LIQUIDS

Material	Viscosity		Density		Sat. Liquid Specific Heat		Thermal Conductivity*			
	°C	Poise x 10 ⁻²	°C	gm/cm ³	°C	cal/gm °C	°C	cal/(sec)(cm ²)(°C/cm)	°F	Btu/(hr)(ft ²)(°F/ft)
H ₂ O	0	1.7921	0	1.000	0	1.008	0	0.00132	32	0.319
	20.2	1.0000	20	1.002	20	.998	93	0.00162	200	0.392
	40	0.656	40	1.008	40	.998	205	0.00158	400	0.382
	60	0.4688	80	1.029	60	.999	316	0.00121	600	0.293
	80	0.3565	100	1.043	80	1.002				
	100	0.2838			100	1.006				
	150	0.13			150	1.030				
Sodium	103.7	0.684	100	0.928	100	0.3305	100	0.204	212	49.3
	167.4	0.506	250	0.891	200	0.3199	200	0.193	392	46.6
	250	0.381	400	0.854	400	0.3055	350	0.175	662	42.4
	400	0.269	550	0.817	600	0.3002				
	700	0.182	700	0.780	800	0.3040				
Lead	441	2.116	327.4	10.686	327	0.039	330	0.039	626	9.4
	456	2.059	450	10.536	400	0.037	400	0.038	752	9.2
	551	1.700	650	10.302	500	0.037	500	0.037	932	8.9
	703	1.349	750	10.188			600	0.036	1112	8.7
	844	1.185	850	10.078			700	0.036	1292	8.7

*Btu/(hr)(ft²)(°F/ft) x 0.00413 = Cm cal/sec cm² °C/cm

The relative motion between the fluid and pipe is the fluid velocity v . Therefore, the energy dissipated in overcoming friction is some function of all these variables or

$$f' = \Phi_1(D, L, \epsilon, m, \rho, \mu, v) \quad (1)$$

For a given point condition the unknown function, Equation 1, may be written in exponential form.

$$f' = z D^a L^b \epsilon^c m^d \rho^e \mu^n v^r \quad (2)$$

where the dimensionless coefficient z and all the exponents are of given values only at the point condition. If the condition is changed in any way, as by changing the velocity, z and all the exponents may change. Equation 2 is simply another way of writing Equation 1. It is definitely not to be inferred that the true relation between the variables is exponential.

Application of the first principle of dimensional analysis by substitution of the fundamental dimensions is presented below, and the units are presented in Table II

$$\frac{mL^2}{t^2} = z L^a L^b L^c m^d \left(\frac{m}{L^3}\right)^e \left(\frac{m}{Lt}\right)^n \left(\frac{L}{t}\right)^r \quad (3)$$

Table II

DIMENSIONS OF VARIOUS QUANTITIES IN TERMS OF MASS, m:
LENGTH, L; TIME, t; TEMPERATURE, T

<u>Quantity Units</u>	
<u>Quantity</u>	<u>Mass, m</u> <u>Length, L</u> <u>Time, t</u> <u>Temperature, T</u>
Temperature (T)	T
Time (t)	t
Length (x, y, z, D, L,)	L
Mass (m)	m
Force (F)	mL/t^2
Energy (E)	mL^2/t^2
Velocity (v)	L/t

Table II (Contd.)

Quantity	Mass, m Length, L Time, t Temperature, T
Acceleration (a)	L/t^2
Density (ρ)	m/L^3
Pressure (P)	m/Lt^2
Surface tension (γ)	m/t^2
Mass flow rate (W)	m/t
Mass velocity (G)	m/L^2t
Viscosity (μ)	m/Lt
Heat capacity	mL^2/t^2T
Specific heat (C)	L^2/t^2T
Thermal conductivity (k)	mL/t^3T
Rate of heat transfer (q)	mL^2/t^3
Coefficient of heat transfer (h)	m/t^3T

By application of the second principle, the net dimension of the product on the right side must be the same as on the left, mL^2/t^2 , and the summation of the exponents of each fundamental dimension on the right is equal to the exponent of that dimension on the left, as follows:

$$\text{For } m \quad 1 = d + e + n$$

$$\text{For } L \quad 2 = a + b + c - 3e - n + r$$

$$\text{For } t \quad -2 = -n - r$$

Here are three equations and seven variables or unknowns. The energy dissipated as friction is directly proportional to the mass of fluid transported, and d is therefore unity. This leaves six unknowns for three equations. If the three equations are independent, they may then be solved in terms of three arbitrarily chosen variables, such as b , c , and n , giving

$$e = -n \qquad r = 2 - n$$

$$a = 2 - b - c - 3n + n - 2 + n = -b - c - n$$

If these values are substituted for a , e , and r in the exponents of Equation 2,

$$f' = zD^{-b-c-n} L^b \epsilon^c m^{-n} \mu^{n/2} v^{2-n}$$

The groups into which these variables will be assembled in the functional relation are indicated by the exponents. The variables are assembled according to the appearance of each letter in the exponent to assure dimensional homogeneity. For convenience the groups are usually made dimensionless as follows:

All variables having a numerical exponent yield the dimensionless group f'/mv^2 .

All variables having the exponent b yield the dimensionless group L/D .

All variables having the exponent c yield the dimensionless group ϵ/D .

All variables having the exponent n yield the dimensionless group $\mu/Dv\rho$.

These dimensionless groups constitute a function of some description to express the interrelation of variables.

$$\frac{f'}{mv^2} = \Phi_2 \left[\frac{(L)}{(D)}, \frac{(\epsilon)}{(D)}, \frac{(\mu)}{(Dv\rho)} \right] \quad (4)$$

If exponents other than b , c , and n had been chosen for retention in the establishment of dimensional homogeneity, a different set of dimensionless groups would have resulted. Intermultiplication of the dimensionless groups to yield different dimensionless groups is permissible. In this way some other function of different dimensionless groups may be obtained which may be more convenient than the first.

The best choice of exponents for retention, or the manner of intermultiplication to yield the most convenient groups, depends on the use to be made of them.

This is all the information that can be obtained from the application of dimensional analysis to this problem. The nature of the function Φ_2 is not indicated in any way. The analysis simply states that a function exists between these various dimensionless groups, however complicated it may be, provided all the significant physical quantities have been included. This is an important guide to the procedure of determining the function empirically from experimental data. For example, a series of measurements of the friction energy f' taken under conditions of constant L/D and constant ϵ/D must yield a single function when f'/mv^2 is plotted against the Reynolds number $Dv\rho/\mu$. All experimental points lie on the same curve, regardless of the velocity, density, viscosity, or mass of the fluid. If the experimental points do not lie on a single curve within the experimental error, one or more significant physical quantities or measurements were omitted in writing the first Equation 1.

Equation 4 may be written as follows, with energy given in foot pounds (or gram centimeters).

$$\frac{f' 2g}{mv^2} = \Phi_3 \left[\frac{(L)}{(D)}, \frac{(\epsilon)}{(D)}, \frac{(Dv\rho)}{(\mu)} \right] \quad (5)$$

In the derivation, the friction energy was assumed to vary directly with the length of pipe, and L accordingly appears as the first power. It follows that L/D appears as the first power, as this is the only dimensionless group containing the quantity L . This assumption has been found valid for all values of L/D greater than about 50. Therefore,

$$f = \frac{f' D 2g}{mLv^2} = \frac{f' D 2g}{Lv^2} = \Phi_4 \frac{(\epsilon)}{(D)} \frac{(Dv\rho)}{(\mu)} \quad (6)$$

The accurate estimation of the friction energy from correlation of experimental data has been a goal of engineers for many years. Early work neglected the character of pipe surface, omitting the ratio, ϵ/D . The data could not be correlated, particularly at high values of the Reynolds number, until this variable was properly included.

As the Reynolds number is increased above 2000, as by increasing the quantity of fluid flowing, the flow will not remain laminar. Near $Re = 2000$, eddies and turbulence may develop in the core of the stream which has the greatest velocity. These eddies dissipate more energy and increase the friction losses. Since the eddies are local, this increased friction loss is local, tending to decrease the local velocity (decrease Re), and the flow may again become laminar. But the local decrease in velocity causes corresponding local increases in velocity elsewhere, with formation of other eddies. This region of incipient turbulence, from $Re = 2000$ to about 4000, is therefore unstable.

As the Reynolds number is increased to values above 4000, the turbulent flowing core becomes well developed and the velocity distribution across a diameter of the pipe stabilizes. The properties of the fluid now have less effect upon the flow conditions than in strictly laminar flow, and the roughness becomes of significance, particularly if it is sufficient to affect the turbulent core in the central part of the stream. Friction losses are stabilized and can be computed.

A small twig or other floating object tossed into a river or small stream running rapidly, as in flood, offers an excellent opportunity to see the laminar flow in the slowly flowing water next to the shore and the turbulent flow in midstream.

As the Reynolds number is increased further, the turbulent flowing core expands at the expense of the laminar flowing annulus until the turbulent flowing core occupies the entire cross section of the pipe right down to the protuberances which make up the roughness. In the completely turbulent region the roughness of the pipe is the most important consideration, and the properties of the fluid, viscosity and density can be neglected in estimating friction losses.

The energy dissipated by a fluid emerging from a pipe into a vessel or space of large cross-sectional area cannot be computed accurately. In no event will this loss exceed the kinetic energy possessed by the fluid at the exit of the pipe. This kinetic energy may be taken as the maximum possible loss.

When the conduit through which the material is flowing is of a cross section other than circular, the use of the simple diameter D in the Reynolds number and friction factor is no longer possible. However, if the volume of flowing material in a specified length of conduit V is divided by the wetted surface for the same length of conduit σ , a linear dimension is obtained which may be used as $D/4$ for estimating friction losses for fluids flowing in annuli, and, for lack of a better correlation, for other conduits, whether running full or not. This use of such linear dimension is not consistent with the principle of dynamic similarity; rather it is an empirical procedure for estimating friction losses.

For a cylindrical conduit running full, this linear dimension reduces to the diameter of the conduit.

$$\frac{4V}{\sigma} = 4 \frac{(\pi D^2)}{(4)} \frac{(1)}{(\pi D)} = D \quad (7)$$

Equation 6 may be written as

$$\frac{2gDf}{v^2L} = \Phi_6 \left(\frac{\epsilon}{D} \right) \left(\frac{Dv\rho}{\mu} \right) \quad (8)$$

and the Reynolds number $\frac{Dv\rho}{\mu}$ plotted for various pipe roughnesses, ϵ/D , versus a factor which may be equated to $\frac{2gDf}{v^2L}$ to calculate f , the actual

friction loss. Such a plot has been prepared using all the data collected on pressure drops of gases and liquids, see Figure 1.

Most texts and Handbooks do not include data at the low pipe sizes of interest in some nuclear reactors. Work done in the various Atomic Energy Laboratories has indicated that the data may be extrapolated to these low pipe sizes, see Figure 2.

Figure 1 - Friction Factor vs. Reynolds Number

Figure 2 - Relative Roughness vs. Equivalent Diameter

The method of analysis presented here does not hold for very small capillaries or gases near or above the velocity of sound.

One point that should be considered in fluid flow is the velocity to use. Regardless of the amount of eddy currents and the laminar film along the sides, we use the average or bulk velocity.

If we look at a fluid such as water flowing in a pipe, we differentiate the laminar and turbulent flow as indicated above. In turbulent flow there is actual movement of material transverse to the direction of flow. We might consider this flow as rather a rolling motion with eddies continuously forming and disappearing. Along the sides of the pipe we will always find a layer of varying thickness in laminar flow. This film, as the liquid is heated or cooled, is very important in heat transfer since the heat must flow through this layer by conduction.

Enlargement Losses

There are losses when pipes change size, since the velocity changes, causing extra eddying. For enlargements,

$$F_e = K_e \frac{v_1^2}{2g} = \left[1 - \frac{A_1}{A_2} \right]^2 \left[\frac{v_1^2}{2g} \right]$$

$$\frac{A_1}{A_2} = \text{ratio of upstream area to downstream area.}$$

Values of K_e are given in various texts in chart form. Note that $K_e = 1.0$ for infinite downstream diameter.

Contraction Losses

$$F_c = K_c \frac{v_2^2}{2g}$$

K_c is given by Figure 3.

Fittings

Pressure drops due to fittings and valves can be added to the friction losses by adding various equivalent pipe diameters to the length of the straight pipe.

Figure 3 - Loss Coefficient for Sudden Contraction

Example 1:

A reactor fuel assembly has openings 2.620 in. wide and 0.118 in. thick. The plates are 25 in. long. Square end pieces fit over the assemblies of fuel plates in the form of an extension of the side plates. Calculate the pressure drops in psi between the end pieces for the fuel assembly for water at an average temperature of 100°F and sodium at 400°F assuming that the surface roughness of the plates is equivalent to drawn tubing and that fluid velocities of 20, 25, 30, 35, and 40 ft/sec are used.

Solution:

$$v = 20, 25, 30, 35, 40 \text{ ft/sec}$$

$$D: \text{Perimeter} = 2 \times 2.620 + 2 \times 0.118 = 5.48 \text{ in.} = 0.456 \text{ ft}$$

$$\text{Area} = 2.62 \times 0.118 = 0.309 \text{ in}^2 = 0.00214 \text{ ft}^2$$

$$V \text{ per foot} = 0.00214 \text{ ft}^3$$

$$\text{Wetted perimeter/ft} = 0.456 \text{ ft}^2$$

$$D/4 = 0.00214/0.456 = 0.00469$$

$$D = 0.0188 \text{ ft} = 0.226 \text{ in.} \text{ Note that this is nearly equal to 2 times the actual thickness.}$$

$$\rho \text{ H}_2\text{O} = 62.1 \text{ lb/ft}^3 \qquad \rho \text{ Na} = 0.901 \text{ gm/cc} \\ = 56.2 \text{ lb/ft}^3$$

$$\mu \text{ H}_2\text{O} = 0.69 \text{ centipoises} \qquad \mu \text{ Na} = 0.428 \text{ centipoises} \\ = 0.000464 \text{ lb/ft sec} \qquad = 0.000288 \text{ lb/ft sec}$$

$$400^\circ\text{F} = 204^\circ\text{C}$$

$$\text{H}_2\text{O: } \frac{Dv\rho}{\mu} = \frac{0.0188 v 62.1}{0.000464} = 2520 v$$

$$\text{Na: } \frac{Dv\rho}{\mu} = \frac{0.0188 v 56.2}{0.000288} = 3670 v$$

H₂O

f	Re	$f' = \frac{2gDf}{v^2L}$	v^2	$\frac{f}{\text{ft of head}}$	$\frac{\text{ft}}{\text{in psi}}$	Contrac. & Exp. Loss		Calculated Total Loss	Measured* Length	
				ft	psi	ft	psi	psi	24in.	Total
20	50,400	0.0235	400	16.1	6.91	9.3	4.01	10.9	7.0	9.5
25	63,000	0.0220	625	23.6	10.2	14.6	6.31	16.5	9.5	12.0
30	75,500	0.0210	900	32.5	14.0	20.0	8.65	22.6	16.0	20.0
35	88,100	0.0200	1225	42.1	18.2	28.5	12.3	30.5	-	-
40	101,000	0.0196	1600	54.0	23.3	37.3	16.1	39.4	-	-

Na

20	73,500	0.0215	400	14.8	5.78	9.3	3.63	9.4		
25	91,800	0.0199	625	21.4	8.35	14.6	5.60	14.0		
30	110,000	0.0194	900	30.0	11.7	20.0	7.81	19.5		
35	128,000	0.0188	1225	39.7	15.5	28.5	11.1	26.6		
40	147,000	0.0185	1600	50.9	19.9	37.3	14.6	34.5		

*In somewhat different geometry

$$\epsilon = 0.000,005$$

$$\epsilon/D = 0.0003$$

$$L = \frac{25}{12} = 2.08$$

$$\frac{2gD}{L} = 0.581$$

Inlet and outlet losses:

$$\text{Inlet} \quad \frac{0.5v^2}{2g}$$

$$\text{Outlet} \quad \frac{v^2}{2g}$$

$$\text{Total} \quad \frac{1.5 v^2}{2g}$$

Example 2:

In a reactor clad rods 1.00 in. diam are giving up heat to water flowing at 25 ft/sec (inlet) in a tube 1.250 in. ID. Inlet water temperature is 60°F and the exit water temperature 200°F. The heated section of the tube is 25 ft long with 8 ft inlet and outlet sections. What is the pressure drop over the tube? Assume that the tubes are drawn and that the clad rods have a roughness equivalent to regular iron pipe. Neglect gamma heating, assume no flattening, and neglect horizontal heat transfer, but take into consideration change of physical properties with temperature.

Solution:Values of Cosine at Various Points

$$\frac{\text{Distance}}{12.5} \times 90 = \text{angle}$$

since cosine at center must equal the maximum or 1.0 so use sine of angle.

L	Dist. in	Angle	Sine	Temperature Rise, ° F	Temperature, °F
8	0	0	0	0	60
10.5	2.5	18	0.309	3.4	63.4
13	5.0	36	0.588	13.4	73.4
15.5	7.5	54	0.809	28.9	88.9
18	10	72	0.951	48.4	108.4
20.5	12.5	90	1.000	70.0	130
23	15	72	0.951	91.6	151.6
25.5	17.5	54	0.809	111.1	171.1
28	20	36	0.588	126.6	186.6
30.5	22.5	18	0.309	136.5	196.5
33	25	0	0	140	200

Temperature rise = 200 - 60 = 140°F, or 70°F to center. Rate of temperature rise is proportional to the cosine or area under cosine curve.

Dist. in	Area, Avg. Sine	Temperature Rise in Unit Area
0 to 2.5	0.155	3.43
2.5 to 5	0.448	9.93
5 to 7.5	0.699	15.5
7.5 to 10	0.880	19.5
10 to 12.5	0.976	21.6
	3.158	70.0

$$\text{Temperature rise} = \frac{70}{\text{Total Area}} \times \text{unit areas}$$

Flow Areas

$$\text{Area of rods} = 0.785 \text{ in}^2 = 5.45 \times 10^{-3} \text{ ft.}^2$$

$$\text{Area of tube} = 1.227 \text{ in}^2 = 8.51 \times 10^{-3} \text{ ft.}^2$$

$$\text{Area of flow} = 3.06 \times 10^{-3} \text{ ft.}^2$$

Flow Per Second

$$\text{vol.} = 25 \times 3.06 \times 10^{-3} = 7.65 \times 10^{-2} \text{ ft.}^3/\text{sec.}$$

$$m = 7.65 \times 10^{-2} \times 62.4 = 4.77 \text{ lb/sec.}$$

PROPERTIES

<u>L, ft</u>	<u>Temp, °F</u>	<u>Avg. Temp., °F</u>	<u>ρ, lb/ft³</u>	<u>vol, ft³/sec</u>	<u>v, ft/sec</u>	<u>μ, Centi-poises</u>	<u>μ, lb/ft sec</u>	<u>$\frac{Dv\rho}{Re, \mu}$</u>
0	60							
8	60	60	62.4	7.65×10^{-2}	25	1.1	7.40×10^{-4}	44,200
10.5	63.4	61.7	62.4	7.65×10^{-2}	25	1.1	7.40×10^{-4}	44,200
13	73.4	68.4	62.4	7.65×10^{-2}	25	0.95	6.39×10^{-4}	51,300
15.5	88.9	81.2	62.3	7.66×10^{-2}	25	0.85	5.71×10^{-4}	57,300
18	108.4	98.7	62.1	7.68×10^{-2}	25.1	0.70	4.71×10^{-4}	69,500
20.5	130	119.2	61.9	7.71×10^{-2}	25.2	0.56	3.77×10^{-4}	86,900
23	151.6	140.8	61.4	7.78×10^{-2}	25.4	0.46	3.09×10^{-4}	106,000
25.5	171.1	161.4	61.0	7.82×10^{-2}	25.6	0.38	2.55×10^{-4}	129,000
28	186.6	178.9	60.8	7.85×10^{-2}	25.7	0.33	2.22×10^{-4}	148,000
30.5	196.5	191.5	60.4	7.90×10^{-2}	25.8	0.30	2.02×10^{-4}	162,000
33	200	198.2	60.2	7.92×10^{-2}	25.8	0.28	1.88×10^{-4}	174,000
41	200	200	60.2	7.92×10^{-2}	25.8	0.27	1.82×10^{-4}	179,000

Note the end pieces need not be filled with slugs.

Note also that the only property that changes appreciably is viscosity.

$$\mu : \frac{\text{lb}}{\text{ft. sec.}} = \text{centipoises} \times 0.000672$$

$$Re : \frac{Dv\rho}{\mu} = 2.1 \times 10^{-2} \frac{v\rho}{\mu}$$

$$D : \text{For a 1 ft. length, } V = 3.06 \times 10^{-3} \text{ ft.}^3$$

$$\begin{aligned}\text{Wetted area} &= \frac{\pi \times 1.00 + \pi \times 1.25}{12} \\ &= 0.59 \text{ ft.}^2\end{aligned}$$

$$D/4 = \frac{3.06 \times 10^{-3}}{0.59}$$

$$D = 2.1 \times 10^{-2} \text{ ft.}$$

$$\frac{\epsilon}{D}, = \text{relative roughness}$$

$$\text{Equivalent } D = 2.1 \times 10^{-2} \times 12 = 0.252 \text{ in.}$$

$$\frac{\epsilon}{D} \text{ for ID of tube} = 0.00025$$

$$\frac{\epsilon}{D} \text{ for rods} = 0.078$$

$$\frac{\epsilon}{D} \text{ avg.} = \frac{1 \times 0.0078 + 1.25 \times 0.00025}{2} = 0.0041$$

Note - This evaluation is not accurate, but there does not appear to be any better method of estimating this factor.

<u>L, ft</u>	<u>$f' = \frac{2gDf}{v^2 L}$</u>	<u>$v^2 L$</u>	<u>f, ft.</u>	<u>psi</u>
0	0.032	5000	118	51.1
8	0.032	1565	37.1	16.0
10.5	0.031	1565	35.9	15.5
13.0	0.0308	1565	35.7	15.4
15.5	0.0303	1580	35.5	15.3
18	0.0300	1600	35.5	15.3
20.5	0.0298	1612	35.5	15.3
23	0.0296	1640	36.0	15.4
25.5	0.0295	1655	36.2	15.3
28	0.0294	1660	36.2	15.3
30.5	0.0294	1660	36.2	15.2
33	0.0294	5310	105	44.3
41				

$$2 gD = 32.2 \times 2 \times 2.1 \times 10^{-2} = 1.35$$

$$\text{psi} = \frac{f}{144}$$

Total less ends 154 psi

ends 95

Total 249 psi

Inlet and outlet losses -

Assume inlet pipe infinite size:

$$F_c = \frac{Kv_2^2}{2g}$$

$$K = 0.5$$

Exit -

Assume: $\frac{v_2^2}{2g}$ since all kinetic energy is lost

$$\begin{aligned} \text{Losses for inlet and outlet} &= 0.5 \frac{v_2^2}{2g} + 1.0 \frac{v_2^2}{2g} \\ &= \frac{0.5 \times 25.0^2}{64.4} + \frac{1.0 \times 25.8^2}{64.4} \\ &= 15.2 \text{ ft. of head or } 6.3 \text{ psi} \end{aligned}$$

Total pressure drop over reactor as given by statement of problem

$$= 255 \text{ psi}$$

If ends are not filled with slugs, we get about

$$154 + 6 = 160 \text{ psi}$$

Two Phase Pressure Drop

In the Martinelli-Nelson method for calculating the pressure drop in two-phase isothermal flow such as boiling in a reactor, allowances are made for the increase in momentum of the liquid and vapor as more vapor is formed, and also for the slippage of the vapor past the liquid. There is always mixing of the liquid and vapor but at the exit the vapor always has a velocity much higher than the liquid.

Martinelli and Nelson calculated curves of $\frac{\Delta P_{TPF}}{\Delta P_0}$ versus pressure for various exit steam qualities. We will not go through the derivation of their equations here. $\Delta P_{TPF}/\Delta P_0$ is simply the ratio of friction drop in two phase flow to that in the liquid flow for the same system. Use of this ratio gives the increase in friction only.

We must also calculate the increase in the momentum of the liquid-gas system. Martinelli and Nelson have calculated factors for estimating the pressure drops due to the acceleration of the fluids during evaporation:

$$\Delta P_a = r_2 \frac{G^2}{g} \quad (9)$$

The nomenclature used by Martinelli and Nelson follows:

ΔP_{TPF} = Frictional pressure drop, psf

ΔP_0 = Pressure drop in flow circuit with saturated water at the same rate and pressure, psf

ΔP_a = Pressure drop due to acceleration of the fluid, psf

r_2 = Multiplier of $\frac{G^2}{g}$, ft^3/lb

G = Weight flow per unit area, lb/hr ft^2

g = Gravitational force per unit mass

$$= 4.17 \times 10^8 \frac{\text{lb}}{\text{lb hr}^2/\text{ft}}$$

The values of $\Delta P_{TPF}/\Delta P_0$ and r are obtained from the charts given in Martinelli and Nelson's paper, which are reproduced here as Figures 4 and 5.

Example 3:

A vertical forced circulation evaporator is operated at an exit pressure of 300 psig. The tubes are 1.00 in. ID and 10 ft long over the heated section. Heating is carried out by high pressure steam. Five per cent of the water is evaporated per pass. Inlet velocity is 10 ft/sec. Assume that the external pipe resistance is negligible. Calculate the circulating pump pressure.

Solution:

Calculation of pressure drop assuming no vaporization:

$$\frac{Dv\rho}{\mu}, D = \frac{1}{12} = 0.0833$$

$$v = 10$$

$$\rho = 52.6$$

$$\mu = 0.057 \text{ centipoises} = 3.83 \times 10^{-5}$$

$$\text{Re} = 1,140,000$$

Figure 4 - Ratio of Pressure Drop in Boiling to Non-Boiling

Figure 5 - Multipliers for Momentum Calculations

$$\text{Temp} = 422^{\circ}\text{F}$$

$$\frac{\epsilon}{D} = 0.000060, \text{ assume tubing}$$

$$f' = \frac{2gDf}{v^2L} = 0.0128$$

$$f = \frac{0.0128 \times 10^2 \times 10}{64.4 \times 0.0833}$$

$$= 2.39 \text{ ft of head}$$

$$\frac{\Delta P_{\text{TPF}}}{\Delta P_0} = 4.4, \text{ see Figure 4}$$

Total friction drop in tube = 10.5 ft of head = 3.84 psi

Pressure drop due to momentum increase:

$$\Delta P_a = r_2 \frac{G^2}{g}$$

$$G \text{ wt flow/unit area, lb/hr ft}^2 = \frac{W_T}{A_p}$$

$$W_T = \text{total wt flow, lb/hr}$$

$$A_p = \frac{\pi}{4} D^2 = \text{cross section area, ft}^2$$

$$g = \text{gravitational force per unit mass}$$

$$= \frac{4.17 \times 10^8 \text{ lb}}{\frac{\text{lb hr}^2}{\text{ft}}}$$

$$W_T = \rho_v \times 3600$$

$$= 52.6 \times 10 \times 3600$$

$$= 1.893 \times 10^6 \text{ lb/ft}^2 \text{ hr}$$

$$G = 1.893 \times 10^6$$

r_2 from

$$\text{Figure 5} = 0.039$$

$$\Delta P_a = r_2 \frac{G^2}{g}$$

$$= 0.039 \times \frac{(1.893 \times 10^6)^2}{4.17 \times 10^8}$$

$$= 335 \text{ lb/ft}^2$$

$$= 2.33 \text{ psi}$$

Inlet and outlet losses since ΔP_a is increase in momentum:

$$\Delta P \text{ cont. + exit} = \frac{1.5 V_2^2}{2g} = \frac{1.5 \times 100}{64.4} = 2.33 \text{ ft of head}$$

$$= 0.85 \text{ psi}$$

Total pressure drop over tube:

Inlet and exit:	0.85 psi
Friction:	3.84
Increase in momentum:	2.33
Total	7.0 psi

This assumes the pressure in the downcomer offsets the pressure in the vertical tubes.

Gases

Bernoulli's theorem over any system states

$$\int_1^2 \bar{V} dP + \frac{\Delta v^2}{2g} + \Delta z = -w - f \quad (10)$$

Substituting for f its value we have

$$\int_1^2 \bar{V} dP + \int_1^2 \frac{v dv}{g} + \Delta z = -w - \int_1^2 \frac{f v^2 dL}{2g}$$

$$(11)$$

where $\bar{V}$ refers to unit mass. In order to evaluate the integrals, we use the ideal gas law

$$\bar{V} = \frac{RT}{MP}$$

where M is the molecular weight, or for non-ideal gases

$$\bar{V} = \frac{zRT}{MP}$$

where z is the compressibility factor.

fv^2 must be known as a function of L . Since this is not known, resort must be made to a stepwise solution using short sections and average values of $\bar{V}$ and v . Or we may use the mass velocity, G ,

$$\int_1^2 \bar{V} dP + \int_1^2 \frac{G^2 \bar{V} d\bar{V}}{g} + \Delta z = -w - \int_1^2 \frac{G^2 \bar{V}^2 f dL}{2gD} \quad (12)$$

G is the mass velocity in $\frac{\text{lb mass}}{\text{ft}^2 \text{ sec}}$

For gas flowing through a reactor, Δz will be zero or small, and w over the reactor section only will be zero. Since G is constant, the Reynolds number will change only as μ changes. If μ is constant, the Re number will be constant, f will be constant and we may write the above equation (after dividing by $\bar{V}^2$) as:

$$\int_1^2 \frac{dP}{\bar{V}} = -\frac{fL}{2gD} \frac{G^2}{\bar{V}} - \int_1^2 \frac{G^2 d\bar{V}}{g\bar{V}} \quad (13)$$

and the last term may be dropped if velocity effects are negligible.

For isothermal gas flow, this equation may be integrated by substituting

$$\bar{V} = \frac{zRT}{PM}$$

in which

M = molecular weight of the gas,

or

$$\frac{M}{zRT} \frac{(P_2^2 - P_1^2)}{2} = -\frac{fLG^2}{2gD} - \int_1^2 \frac{G^2 d\bar{V}}{\bar{V}} \quad (14)$$

If velocity effects are small, the last term may be dropped.

For adiabatic conditions, the equation may be integrated by substituting from

$$\bar{V} = \frac{1}{\rho}$$

or

$$\rho \int_1^2 dP = \frac{fLG^2}{2gD} - \int_1^2 \frac{G^2 d\bar{V}}{g\rho} \quad (15)$$

Using an average ρ the equation can be readily solved.

Integrated Form

A slightly different form of the friction equation with f assumed constant for a gas in a channel is:

$$\Delta p = p_1 - p_2 = \frac{G^2 (v_2 - v_1)}{g_c} + \frac{G^2 f v_m L}{2 g r_m} \quad (16)$$

p	= absolute pressure	lb/ft ²
G	= mass velocity	lb/ft ² sec
v	= specific volume	ft ³ /lb
v_m	= mean specific volume	ft ³ /lb
g	= conversion factor 32.2	ft lb mass/sec ² lb force
f	= friction factor	-
L	= length	ft
r_m	= hydraulic radius $\left(= \frac{De}{2} \right)$	ft
T	= absolute temperature	°R or °K

$$v_m = \frac{1}{L} \int_0^L v \, dL \text{ and for a perfect gas } v_m = v_1 \frac{p_1 T_{av}}{p_{av} T_1}$$

$$v_m = \frac{p_1 v_1}{L T_1 p_{av} w c} \int_0^L \left[\int_0^L q \, dL \right] dL,$$

may be used graphically to obtain the value of v_m when q versus L is known.

$$v_m = \frac{v_2 - v_1}{\ln \frac{v_2}{v_1}} \text{ may also be used.}$$

This leads to

$$1 - \frac{p_2}{p_1} = \left(\frac{G^2 v_1}{g p_1} \right) \left[\frac{v_2}{v_1} - 1 \right] \left[1 + \frac{(fL/2r_m)}{\ln \frac{v_2}{v_1}} \right] \quad (17)$$

In systems being heated in turbulent flow, the data for computation of Re is correlated well by evaluating the physical properties at

$$t_{0.4} = 0.4 (t_0 - t_b) + t_b \quad (18)$$

in which

t_0	=	inside wall temperature °R or °K
t_b	=	bulk temperature of fluid °R or °K
v_b	=	bulb velocity ft/sec
μ	=	viscosity lb sec/ft ²
D	=	diameter ft
ρ	=	avg. density lb/ft ³

Equation 17 may be solved by use of Figure 6 which presents

$$\frac{D v_b \rho_{0.4}}{\mu_{0.4}} \text{ vs } f_{0.4}$$

Two-Phase Flow

One of the important applications of pressure drop calculations is that involved in a nuclear reactor in which water is boiling in the coolant channels. The mechanism of this phenomenon is discussed in some detail in Lecture 9.

Typical curves for the pressure drop for a given geometry, inlet subcooling, and pressure conditions but differing heat fluxes is presented in Figure 7.

The two phase region pressure drop can be calculated by the Martinelli-Nelson method. While the shape of the curve has been checked experimentally, the analytical methods do not check all points quantitatively.

The curve A is considered to be in a region of instability since increasing the pressure does not force the steam from the tube. The dotted line indicating the operating pressure crosses Curve A at two points. Once boiling starts, there is insufficient pressure to force the steam out of the channel and restore the full liquid conditions. In case B, there is stability and there is always potential pressure to drive steam from the system.

Returning to Curve A, the conditions that exist from points 1 to 2 will permit tube burnout. At rates of flow greater than indicated by point 1, the heat will be transferred by boiling and the channel surface temperatures will not be greatly in excess of saturation. When the inside of the channel becomes blanketed with steam, the heat transfer drops to a low value, and the surface temperatures may rise in a reactor to very high values. Melting of the fuel cladding would take place in a matter of a very few seconds.

Figure 6 - Friction Factor Correlation

Figure 7 - Typical Flow Curves for Heating Water

The points at which burnout can occur must be determined experimentally. As will be noted in the next lecture, several attempts have been made to correlate the data but extrapolation of the data available over wide ranges is uncertain. In vertical channels with L/D ratios greater than 50 and heat fluxes less than 200,000 Btu/ft² hr. the overheating occurs with steam qualities of 80 to 100%. The effect of tube annulus has not been investigated separately from the L/D ratio.

Parallel Channels

If we plot the pressure drop curves for two parallel channels, letting A represent a channel with average flow and B represent a channel with higher than average heat generation, we obtain Figure 8.

If the pressure drop across the tubes should fall below the minimum point on Curve B, this channel may burnout since the channel is one of instability and steam would continue to blanket the tube. This happens since the generation of the steam increases the pressure drop decreasing the flow even further.

It is to be noted that an obstruction in one of a number of parallel channels can cause burnout even though the obstruction was not serious in series flow.

Orifices

An orifice changes the characteristics of flow in parallel tubes by maintaining more nearly constant pressures over the individual tubes. For a reactor designed to operate in the non-boiling region, the orifice provides extra pressure for forcing steam out of a tube in a multiple channel system. This is illustrated in Figure 9. The header pressure DD may be about 30% above B in some reactors.

Sudden and small changes in flow levels will be leveled out by orifices. Hot spots also will be leveled out even though boiling occurs at these points.

Flow measurement and temperature checking also are capable of preventing burnout since they check the actual water flow out of the tubes along with the temperatures.

When possible, natural circulation by use of vertical tubes and upflow should be used.

Figure 8 - Pressure Drop in Parallel Flow

Figure 9 - Effect of Orifice on Parallel Flow

Let us turn to the multiple tube heat exchanger in which all the tubes are in parallel and none are orificed. The pressurized water-boiler, Na-NaK, and NaK-boiler are applications of such heat exchangers. In these heat exchangers the maximum temperature is obviously the temperature of the hot liquid inlet so that there is no danger of burnout. In case of an obstruction in one tube, there will be possible overheating with higher stresses, but no danger of burnout.

Friction Drops Across Tubes

Friction drops across banks of staggered and unstaggered tubes are given by the following equations.

Friction drop for isothermal turbulent flow across tube banks is given by the equation

$$\frac{\Delta P}{\rho} = \frac{f_1 N v_{\max}^2}{2g} \quad (19)$$

where

ΔP = pressure drop, lb/ft²

ρ = density, lb/ft³

f_1 = friction factor given by the following equations

N = number of rows of tubes

$v_{\max}$ = maximum velocity based on minimum free area, ft/sec

g = acceleration of gravity, 32.2 ft/sec²

Staggered tubes:

$$f_1 = 4 \left[0.23 + \frac{0.11}{(x_t - 1)^{1.08}} \right] \left[\frac{D_0 G_{\max}}{\mu_f} \right]^{-0.15} \quad (20)$$

Unstaggered tubes:

$$f_1 = 4 \left[0.044 + \frac{0.08x}{(x_t - 1)^n} \right] \left[\frac{D_0 G_{\max}}{\mu_f} \right]^{-0.15} \quad (21)$$

x_t = ratio of the transverse pitch to the tube diameter

$n = 0.43 + \frac{1.13}{x_1}$

x_1 = ratio of longitudinal pitch to tube diameter; pitch is measured from center to center of the tubes.

D_0 = diam (outside) of pipe, ft

$G_{\max}$ = maximum mass velocity based on the minimum free area for flow through tube banks

μ = viscosity, lb/ft sec

Vibration

Vibration of fuel assemblies due to water flow is caused by change in the pressures exerted on the two sides of a fuel plate due to variations in pressure caused in turn by variations in velocity and to turbulence. The variations in velocity are not usually considered important since the pressure change associated with the velocity variations are small. However, the velocity and the pressure may differ considerably between the outside and inside of a fuel assembly.

The variations in pressure due to the horizontal components of the kinetic energy due to local turbulence may cause vibration of the fuel plates or fuel slugs.

These effects are always small as long as the vibrational frequency of the fuel plate and flow turbulence do not coincide. When they do coincide, there is danger of building up a vibration which could become harmful. The probability of this happening is very small in reactor systems.

The effects of turbulence at the exit ends of fuel plates is more drastic because it is possible for conditions to exist in which the vortices on the opposite sides of a fuel plate become additive.

Den Hartog presents a discussion of "Karman Vortices." The wake behind a cylindrical obstacle in a fluid stream is not regular, but contains vortices shed alternately from the two sides of the obstacle in a regular manner. This is to be expected from the nature of turbulent flow. The data can be represented by the dimensionless relationship

$$\frac{fD}{v} = 0.22 \quad (22)$$

where

f is the frequency of shedding of the vortices

D is the diameter

v is the velocity

Experimental results on a reactor fuel plate gave the equation

$$f = 40 v \quad (23)$$

Den Hartog's equation gives

$$f = \frac{0.22 v}{0.064/12}$$

since the reactor fuel plate is 0.064 in. thick - or

$$f = 41 v \quad (24)$$

While the two equations were derived for different systems they do indicate that the shedding of vortices from plate ends may cause vibration whose frequency is proportional to the coolant velocity.

Formulas for vibrations of plates and assemblies are presented in various texts like Den Hartog's. For example, the fuel assembly may be considered a "clamped-hinged" beam for which

$$f = \frac{a_n}{2} \sqrt{\frac{E I g}{w l^4}} \quad (25)$$

has been derived.

- f = frequency of mode n , cycles/sec
- a_n = constant depending on type of restraint and mode of vibration and has the values here of 15.4, 50, 104
- E = modulus of elasticity, psi = 10,000,000
- I = moment of inertia of beam section (in)⁴
 - $I_x = 3.45$ (in)⁴; (x axis parallel to plates)
 - $I_y = 3.89$ (in)⁴; (y axis $\perp$ to plates)
- g = $32.2 \times 12 = 386$ in/sec²
- w = weight/unit length = 0.400 lb/in.
- l = length of beam in., = 41 in.
- f_x = 266, 864, 1800 cycles/sec
- f_y = 282, 917, 1910 cycles/sec

For the plates as "clamped-clamped" beams

$$f = 1815, 5030 \dots\dots\dots \text{cycles/sec.}$$

Measured values of the frequency of vibration had increased components at 24 - 30 and 35 - 37 ft/sec with frequencies of 900 - 1200 and 1400. At 30 ft/sec the maximum measured vibration was 0.7 mil, see Figure 10.

Vibrations can only be serious in fairly large pipes that have long unsupported spans.

Figure 10 - Typical Vibrographs of an Experimental Fuel Assembly Measurement Made on Fuel Plate Near Inlet at Various Water Velocities

Cavitation

Cavitation is a much more serious problem because it is possible to obtain high forces for long periods. Cavitation occurs wherever an obstruction in the path of the liquid causes a vortice which has a velocity component perpendicular to the line of flow with greater force than the amount the absolute pressure exceeds the vapor pressure. Thus, cavitation increases with height of obstructions, increases with temperature, and decreases with pressure.

Cavitation may occur at the tips of propeller blades, etc., but there it may not be harmful. If the cavitation occurs near the wall such that there are shocks on collapse of the bubbles, the forces may be great enough to crack the oxide film on Al tubes. If this happens, corrosion rapidly eats through the wall.

High speed motion pictures have indicated that bubbles collapse when the internal pressure is considerably higher than the vapor pressure, with velocities as high as 800 ft/sec and estimated internal pressures of 50,000 psi. Such high pressures could occur over only microscopically small areas. The force at collapse would become very large per unit area as the surface tension increases inversely as the radius of the bubble. The presence of fixed gases would decrease these forces greatly.

LECTURE 9

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Introduction

In reactor design the bulk of the heat is generated in the fuel plates, rods, or tubes, and we have to consider the transfer of the heat through these plates, rods, or tubes to the outside (or to the inside) and the film coefficients through the coolant films. In homogeneous reactors, the entire mass being heated is transferred out of the reactor so that we have no heat transfer to consider within the reactor. Radiant energy does not have to be considered in most reactors as a factor because, even in reactors gas-cooled with He or air, the temperature is never high enough that the He or air has appreciable radiant heat absorption. There is, of course, a small amount of radiant heat transfer between surfaces in gas-cooled reactors.

On a microscopic scale the heat is available at a uniform temperature which is as high as the materials of construction can stand. This differs from the boiler plant in which the hot gases are cooled rapidly by the boiler tubes and the heat from the gases has to be recovered over a range of temperatures. At some low temperature the gases have to be rejected from the boiler to the economiser and finally from the economiser at a still lower temperature to the stack. From an entropy viewpoint, the heat in a reactor is available at a constant high entropy while the heat in a boiler is available at ever decreasing entropy values.

In reactors, the heat production rate will not be constant over the length of the coolant passage, since the rate of heat production is proportional to the neutron flux. The neutron flux usually drops off towards the end of the coolant passages due to neutron leakage. In addition, the central tubes will be in a zone of the highest neutron flux due to side leakage. This is discussed in more detail in a subsequent lecture.

The only new concept which we must introduce is a calculation of heat transfer through solid materials in which heat development is taking place through the solid. In actual thermal reactors if the fuel plates, rods, or tubes are of thin cross section, the heat development will be relatively

uniform. But in thick rods more heat will be developed on the outside than the inside due to the lowering of the neutron flux through the rods as the thermal neutrons are absorbed. In fast reactors heat development will be quite uniform.

Conduction

Conduction is defined as the flow of heat through a stationary medium. The basic equation for steady state conduction is

$$q = -kA \frac{\Delta t}{\Delta x} \quad (1)$$

or, in differential form,

$$q = -kA \frac{dt}{dx} \text{ (Fourier's Law)}$$

where q = quantity of heat flowing through material, BTU/hr

k = thermal conductivity, BTU ft/hr ft² °F

A = area normal to direction of heat flow, ft²

Δt = temperature difference, °F

Δx = thickness, ft

The thermal conductivity k in the equation for heat conduction varies widely for different materials. For porous materials, such as are used for insulation, the material is in reality a discontinuous phase of fluid maintained stagnant by supporting porous solids, and k represents a combined effect of fluid and solid heat transmission. Gaseous fluids, such as air, are low in thermal conductivity, although they may transfer heat readily by convection; but the minute film of air in the interstices of a porous insulating material exhibits a minimum of convection current. Therefore, the heat transfer through such insulating material takes on the characteristics of heat conduction through the fluid phase.

Since the density of the insulating material will reflect the proportion of the solid to fluid, the conductivity of porous material varies directly with the density and approaches the low value for the conductivity of the fluid as the solid path for heat flow becomes smaller, so long as the gas spaces are small enough to minimize convection.

The range of values of the thermal conductivity for four general types of materials are listed in Table I.

Table I

THERMAL CONDUCTIVITIES

<u>Materials</u>	<u>k (BTU ft/hr ft² °F)</u>
Metals	10 to 200
Insulation	0.04 to 1
Liquids - other than metals	0.04 to 0.4
Gases	0.004 to 0.04

Fourier's Law may be applied to a number of resistances in series, each of which transmits the same quantity of heat under steady state conditions. Consider, for example, three slabs of materials with different coefficients of thermal conductivity.

For each slab we can write:

$$q = -k_1 A_1 \frac{\Delta t_1}{\Delta x_1} = -k_2 A_2 \frac{\Delta t_2}{\Delta x_2} = -k_3 A_3 \frac{\Delta t_3}{\Delta x_3}$$

from which the individual temperature differences can be expressed as follows:

$$\Delta t_1 = \frac{-q \Delta x_1}{k_1 A_1}; \quad \Delta t_2 = \frac{-q \Delta x_2}{k_2 A_2}; \quad \Delta t_3 = \frac{-q \Delta x_3}{k_3 A_3}$$

The total temperature difference is

$$\Delta t = \Delta t_1 + \Delta t_2 + \Delta t_3 = -q \left(\frac{\Delta x_1}{k_1 A_1} + \frac{\Delta x_2}{k_2 A_2} + \frac{\Delta x_3}{k_3 A_3} \right) \quad (2)$$

The heat transfer through the resistances in series is

$$q = - \frac{\Delta t}{\frac{\Delta x_1}{k_1 A_1} + \frac{\Delta x_2}{k_2 A_2} + \frac{\Delta x_3}{k_3 A_3}} \quad (3)$$

For a series of flat plates $A = A_1 = A_2 = A_3 = \dots$
and

$$q = \frac{-A \Delta t}{\frac{\Delta x_1}{k_1} + \frac{\Delta x_2}{k_2} + \frac{\Delta x_3}{k_3} + \dots} = -UA \Delta t \quad (4)$$

where the over-all heat transmission coefficient is

$$U = \frac{1}{\frac{\Delta x_1}{k_1} + \frac{\Delta x_2}{k_2} + \frac{\Delta x_3}{k_3} + \dots} \quad (5)$$

In the case of thermal resistance of scale, the thickness usually is not known explicitly. By rearranging the basic heat transfer equation, we obtain

$$q = -kA \frac{\Delta t}{\Delta x} = -\frac{k}{\Delta x} A \Delta t = -h_{\text{scale}} A \Delta t \quad (6)$$

For scales and tarnishes h_{scale} is about 10,000 - 50,000 BTU/hr ft² °F as determined from heat transfer tests with distilled water in a stainless steel system.

In the case of a reactor with a maximum heat flux of $q/A = 500,000$ BTU/hr ft², and a scale with an $h_{\text{scale}} = 10,000$ BTU/hr ft² °F

$$\Delta t \text{ across scale} = \frac{5 \times 10^5 \text{ BTU/hr ft}^2}{10^4 \text{ BTU/hr ft}^2 \text{ } ^\circ\text{F}} = 50^\circ\text{F}$$

The scales or deposits which would result in this Δt would have appreciable thickness. Most scales on the inside of the central station boiler tubes are made up of complex deposits of CaCO₃, CaSO₄, and Ca₃(PO₄)₂ in various hydrated forms. Corrosion products, various oils, and other insoluble materials, possibly including boiler additives, may be present. In general, the deposits will be porous. In water-cooled reactors, the deposits will consist mainly of oxides, possibly in hydrated forms.

Data obtained by Pridgeon and Badger by measuring over-all heat transfer coefficients from condensing steam to water boiling at 167°F in a horizontal submerged tube evaporator are presented in Table II.

Table II

OVER-ALL HEAT TRANSFER COEFFICIENTS
 ΔU_0 FROM CONDENSING STEAM TO BOILING WATER

Δt_0 , over-all Δt , °F	<u>18</u>	<u>27</u>	<u>36</u>	<u>45</u>	<u>54</u>
U_0 (rusty iron)	280	300	325	350	375
U_0 (clean iron)	300	385	460	535	610
U_0 (slightly dirty copper)	580	780	950	1120	
U_0 (polished copper)	820	1110	1470	1810	2120

After acid cleaning the copper tubing gave U_0 as high as 3070 with $\Delta t = 38^\circ\text{F}$. It was found incredibly difficult to reproduce the high values of U_0 due to the thin collection of deposits on the tubes. Calculated scale thicknesses of 0.0053 to 0.0079 were required to give the loss in heat transfer. Thus, it is apparent that there is need for complete cleanliness in high heat flux heat transfer equipment.

In the design of the MTR, a scale coefficient of 25,000 was used. In the MTR, SR, NRX, and STR highly purified water is used and the scale coefficients are low. This is discussed further in the lecture on corrosion and water cleanup. In the Hanford reactors, filtered and treated Columbia River water is used. Here scale resistances can become high and artificial cleaning is required.

If the resistance is a thick-walled cylinder, it can be treated as a series of thin-walled cylinders, each of thickness " dr " (see Figure 1). For each cylinder of length " L ,"

$$q = -kA \frac{dt}{dx} = -k2\pi rL \frac{dt}{dr} \quad (7)$$

This equation may be rearranged for integration. Assume outward heat transmission; then,

$$q \int_{r_i}^{r_o} \frac{dr}{r} = -2\pi kL \int_{t_i}^{t_o} dt$$

Integrating we obtain

$$q = -2\pi kL \left[\ln \frac{r_o}{r_i} \right]^{-1} (t_o - t_i) = 2\pi kL \left[\ln \frac{D_o}{D_i} \right]^{-1} (t_i - t_o) \quad (8)$$

in which

$$\frac{2\pi L}{\ln(D_o/D_i)} = \frac{A_m}{\Delta x}$$

is the ratio of the logarithmic mean area to the wall thickness.

As an approximation the arithmetic mean area can be substituted if the ratio of D_o/D_i is not greater than 1.5.

The error introduced by using the arithmetic mean is as follows for three ratios of (D_o/D_i) :

Figure 1 - Resistance in a Thick Walled Cylinder

<u>Ratio (D₀/D_i)</u>	<u>ln $\frac{D_0}{D_i}$</u>	<u>Error, Per cent</u>
e	1	8.15
2	0.6935	3.99
1.5	0.4055	1.32

Since the ratio (D₀/D_i) for heat exchanger tubes usually is less than 1.5, we can write

$$q = k\pi L \frac{D_0 + D_i}{2\Delta x} (-\Delta t_w)$$

for either inward or outward heat transfer.

Reactor Fuel Elements Heat Transfer

Heat generation and heat transfer occur simultaneously in reactor fuel elements, materials subject to radiation, and in electrically heated sections. For example, consider a flat plate with an adiabatic insulating blanket against one face. All generated heat is transmitted through the uninsulated face. Let τ = the heat generation rate per unit volume. The heat generation rate in a slab of thickness dx is $dq = \tau A dx$ in the plate between the insulation and the slab.

$$q_x = \tau Ax = -kA \frac{dt}{dx}, \quad (9)$$

and in the entire plate

$$q = \tau Aw$$

see Figure 2.

The q_x equation, which also equates the generation to the transmission at the slab, may be arranged for integration

$$\tau \int_0^w x \, dx = -k \int_{t_i}^{t_0} dt$$

Therefore, the temperature drop through the plate is

$$t_i - t_0 = \frac{\tau w^2}{2k} = \frac{qw}{2Ak} \quad (10)$$

Figure 2 - Fuel Element Section

If the insulation is removed, heat transfer occurs outward from the center plane to both faces and the upper limit of integration across the thickness is $w/2$. The resulting temperature drop is:

$$(t_i - t_o) = \frac{\tau w^2}{8k} = \frac{qw}{8Ak} \quad (11)$$

As a second example, consider uniform heat generation per unit volume in a thick-walled cylinder of inside radius r_i and outside radius r_o with outward heat transmission. The heat generation rate in a thin-walled tube of unit length and volume $2\pi r dr$ is

$$dq = \tau 2\pi r dr,$$

the total rate within the cylindrical section of radius r_o is

$$q = \pi \tau (r_o^2 - r_i^2),$$

and the rate of generation out to dx is,

$$q = \pi \tau (r^2 - r_i^2)$$

which must be transferred through the tube.

$$\pi \tau (r^2 - r_i^2) = -k 2\pi r (dt/dr) \quad (12)$$

Integrating,

$$\tau \left[\frac{r_o^2 - r_i^2}{2} - r_i^2 \ln \frac{r_o}{r_i} \right] = -2k(t_o - t_i) = 2k(t_i - t_o) \quad (13)$$

or, since

$$\tau = \frac{q}{\pi(r_o^2 - r_i^2)},$$

$$(t_i - t_o) = \frac{\tau}{2k} \left[\frac{r_o^2 - r_i^2}{2} - r_i^2 \ln \frac{r_o}{r_i} \right] = \frac{q}{2\pi k} \left[\frac{1}{2} - \frac{r_i^2}{(r_o^2 - r_i^2)} \ln \frac{r_o}{r_i} \right] \quad (14)$$

which reduces to

$$(t_i - t_o) = \frac{q}{4\pi k} \text{ per unit length.} \quad (14a)$$

for a solid rod as

$$\lim_{r_i \rightarrow 0} \frac{r_i^2 \ln(r_o/r_i)}{r_o^2 - r_i^2} = \lim_{r_i \rightarrow 0} \frac{r_i^2}{\ln r_i} = \frac{2r_i dr_i}{dr_i/r_i} = 2r_i^2 = 0$$

For inward heat transmission

$$t_o - t_i = \frac{q}{2\pi k} \left[\frac{r_o^2}{r_o^2 - r_i^2} \ln \frac{r_o}{r_i} - \frac{1}{2} \right] \quad (15)$$

Heat Transfer Coefficients

Dimensionless analysis has been utilized to develop equations that express heat transfer coefficients in a reasonably useable form. The most used dimensionless coefficients are:

Nu, Nusselt	hD/k
Re, Reynolds	$Dv\rho/\mu$
Pr, Prandtl	$C_p\mu/k$
Pe, Peclet	$Dv\rho C_p/k = \text{Re} \times \text{Pr}$
St, Stanton	$h/C_p G$

Fluid in Pipes

It has been determined experimentally that the film coefficient h for fluids (except liquid metals) which are being heated inside conduits may be computed from the equation:

$$\frac{h}{C_p G} = 0.023 \left(\frac{\mu}{DG} \right)^{0.2} \left(\frac{k}{\mu C_p} \right)^{0.6} \quad (16)$$

For water in turbulent flow through heated pipes, non-circular ducts, and annuli, the equation is given in the Reactor Handbook as:

$$\frac{hD_e}{k} = 0.023 \left(\frac{D_e G}{\mu} \right)^{0.8} \left(\frac{C_p \mu}{k} \right)^{0.4} \quad (17)$$

These equations are equivalent. They have been experimentally verified for water flowing in heated tubes up to 2500 psi, 600°F water temperature with heating rates up to 1,000,000 BTU/hr ft² for Re of 50,000 to 700,000. The Reactor Handbook gives plotted results for water at 0.25 in. equivalent diameter tube and correction factors for other equivalent diameters.

For water we may use

$$h = 170(1 + 0.01t - 0.00001t^2) \frac{V^{0.8}}{D^{0.2}}$$

up to 600°F.

Heat transfer to air at high temperatures on short stage fuel elements from 1.0 to 3.5 in. long is correlated by the Dittus Boelter equation except for the effects of the entrances. In the fully developed flow regions

$$\frac{hD}{k_f} = 0.024 \left(\frac{DG}{\mu_f} \right)^{0.8} \left(\frac{T_b}{T_f} \right)^{0.8} \left(\frac{C_p \mu}{k} \right)_f^{0.4} \quad (18)$$

holds for $Re > 30,000$. In this equation T_b = temp. bulk coolant; °R, μ_f , T_f , $\left(\frac{C_p \mu}{k} \right)_f$ are to be taken at film temperature where

$$T_f = T_b + \left[(T_s - T_b) / 2 \right]$$

and T_s is the temperature of the "stage" or wall.

Flow Across Pipes

The following equations are suggested to be used for calculating the heat transfer coefficient for the flow of water normal to banks of staggered tubes, not baffled, and for values of $\frac{D_0 G_{\max}}{\mu_f}$ greater than 10,000.

$$\frac{h_m D_0}{k_f} = 0.33 \left[\frac{D_0 G_{\max}}{\mu_f} \right]^{0.6} \left[\frac{c_p \mu_f}{k_f} \right]^{1/3} \quad (19)$$

The subscript f in the above equation indicates that the physical properties of the water should be evaluated at the film temperature and h_m is the mean value of heat transfer coefficient over the entire surface of the tubes, and $G_{\max}$ means G is to be measured over the minimum free cross sectional area. For unstaggered tubes with values of $\frac{D_0 G_{\max}}{\mu_f}$ greater than 10,000 the equation suggested is:

$$\frac{h_m D_0}{k_f} = 0.26 \left[\frac{D_0 G_{\max}}{\mu_f} \right]^{0.6} \left[\frac{c_p \mu_f}{k_f} \right]^{1/3} \quad (20)$$

These equations apply only to ten or more rows of tubes. The coefficient h_m is the mean or average coefficient for all of the tubes. Table III gives the ratio of h_m for a tube bank N rows deep to that ten rows deep.

Table III

EFFECT OF NUMBER OF ROWS OF TUBES ON THE
AVERAGE HEAT TRANSFER COEFFICIENT (h_m)*

Number of Rows	1	2	3	4	5	6	7	8	9	10
Ratio for Staggered Tubes		0.70	0.82	0.87	0.92	0.94	0.96	0.97	0.99	1.0
Ratio for In-line Tubes	0.64	0.76	0.83	0.87	0.92	0.94	0.96	0.97	0.99	1.0

* McAdams

The average heat transfer coefficient for the flow of water normal to a single tube is given by:

$$\frac{h_m D_0}{k_f} = 0.26 \left[\frac{D_0 G}{\mu_f} \right]^{0.6} \left[\frac{c_p \mu_f}{k_f} \right]^{0.3} \quad (21)$$

for $D_0 G / \mu_f$ from 1000 to 50,000. The coefficient will vary around the cylinder according to its angular position. The magnitude of this variation is estimated to be approximately 30 per cent for water.

The above formulas, correlated with data from gases at lower Reynolds' numbers, have been used generally as the basis for obtaining heat transfer coefficients for water flowing across tube banks. It is recognized that the existing correlations must be extrapolated to much higher Reynolds numbers for use with water.

Recent work has been published by Brookhaven, BNL-203, on film heat transfer coefficients for water in cross flow through tube banks for the following conditions: tube, 0.810 in. OD; length, 12 in.; spacing 1-9/32 in. equilateral pitch center to center; 10 tubes wide x 20 tubes long; water 360°F; number of active elements 3; and maximum temperature rise of water 0.4°F/pass. The results were correlated by

$$h = a Re^b \quad (22)$$

in which $a = 0.755$ and $b = 0.65$ for the front row and $a = 0.18$ and $b = 0.8$ for all subsequent rows.

McAdams on pages 211 to 215 presents some excellent photographs of fluids flowing past pipes.

In the range of Re of 10^5 to 10^6 , a has a value of about 0.19 and b about 0.8. While these values are presumably dependent upon the tube arrangement, this was not investigated. In the tests there was an increase in coefficients up to 15% as the sidewalls were approached.

Randomly Packed Spheres

The heat transfer coefficient for the flow of water through beds of randomly packed spheres of equal diameters may be calculated according to the equation

$$\frac{h_m D_s}{k} = 0.58 \left[\frac{G_s D_s}{\mu} \right]^{0.70} \left[\frac{c_p \mu}{k} \right]^{0.30} \quad (23)$$

h_m = Mean value of h over entire sphere.

D_s = Diameter of sphere

G_s = Mass velocity/unit area of a bed of spheres.

The correlation was based on experiments with air. The properties should be evaluated at the film temperature when large temperature differences exist between the sphere surface and the bulk of the fluid. No data are available on the local heat transfer coefficient or the variation in the coefficient over the surface of a sphere.

Condensing Vapors

For saturated vapors condensing on vertical surfaces, Brown recommends:

$$h = 1.18 \left[\frac{k_f^3 \rho_f^2 g \pi D}{\mu_f W} \right]^{1/3} \quad (24)$$

or

$$h = 1.13 \left[\frac{k_f^3 \rho_f^2 g (\Delta H)}{L \mu_f (T_{sv} - T_s)} \right]^{1/4}$$

in which k_f , ρ_f , and μ_f are to be evaluated at

$$T_f = \left[T_{sv} - \frac{3}{4}(T_{sv} - T_s) \right]$$

T_{sv} = Temperature of saturated vapor, °F

T_s = Temperature of surface, °F

H = Latent heat of condensation, BTU/lb

L = Length of tube, ft.

g = Acceleration of gravity, ft/hr²

D = Outside diameter, ft

W = lb of condensate/hr

For saturated vapors condensing on horizontal tubes:

$$h = 0.725 \left[\frac{k_f^3 \rho_f^2 g (\Delta H)}{ND \mu_f (T_{sv} - T_s)} \right]^{1/4} \quad (25)$$

N = number of tubes in vertical row.

Boiling Coefficients

The heat transfer coefficients for boiling liquids have not been reduced to any reasonable equations. Different phenomenon occur depending on whether the temperature of the bulk of the liquid is at or below the boiling point and upon the quality of the mixture when the liquid is at its saturation temperature.

In the first case, with the temperature of the liquid below its boiling point, the temperature of the heated surface must be some 25°F above the saturation temperature before local boiling at the surface occurs when the liquid is at high Re . When boiling does occur with relatively cold bulk liquid temperatures, the bubbles which form on the hot surface are quickly broken off and condensed. This is known as nucleate boiling. When the temperature is raised still further, the bubbles of vapor may actually coalesce and blanket the tube - thus reducing the rate of heat transfer very greatly.

When the bulk water temperature is at the boiling point, nucleate boiling does not occur, but rather the vapor tends always to blanket the heated surface. Since the thermal conductivity of the vapor is low compared with that of the liquid, the rate of heat transfer is reduced drastically. Brown gives the chart in Figure 3 for coefficients of heat transfer to boiling liquids outside of horizontal tubes (natural circulation) as a function of the temperature drop between the solid surface and the boiling liquid.

In general, boiling coefficients increase with pressures. Jens and Lottes showed that the heat flux varied with pressure in accordance with

Figure 3 - Heat Transfer Coefficient for Boiling Liquids Outside Horizontal Tubes, $\text{BTU}/(\text{hr})(^\circ\text{F})(\text{ft}^2)$

$$\Delta t_{\text{sat}} = \frac{60 \left(\frac{q''}{10^6} \right)^{1/4}}{eP/600} \quad (26)$$

in which $\Delta t_{\text{sat}} = (t_w - t_{\text{sat}})$ = temperature difference between heated wall and saturated temperature, °F.

q'' = heat flux, BTU/hr ft²

P = pressure, lb/ft²

for water flowing inside 0.226 in., 0.143 in., and 0.18 in. ID 347 and 304 s.s. and Ni tubes. Thus the heat flux at 100 psia is indicated to be about 25% greater than that at 50 psia. The data covered pressures of 2500 to 85 psia, mass flow rates of 8000 to 7,650,000 lb/ft²hr, water temperatures of 300 to 600°F, and heat fluxes up to 2.5×10^6 BTU/ft²hr.

Kreith and Sommerfield using a 0.587 in. ID s.s. tube, found a 60% increase for the same pressures. The temperature of the heat transfer surface during boiling has been found to be independent of the water temperature and velocity and dependent only on the heat flux and water pressure. Dissolved gas has little effect on the temperature of the heat transfer surface.

Jens and Lottes, ANL-4627, found that the heat flux that caused burnout was correlated as a function of water temperature, mass rate of flow, and pressure by means of the following type of equation:

$$\left(\frac{q''}{10^6} \right)_{\text{B.O.}} = C \left(\frac{G}{10^6} \right)^M (t_{\text{sat}} - t_b)^{0.22} \quad (27)$$

in which

C and M are functions of the water pressure, see Table IV.

t_b = bulk water temperature, °F

G = mass flow rate, lb/hr ft²

$q''_{\text{B.O.}}$ = heat flux at burnout, BTU/hr ft²

Table IV

VALUES OF BOILING HEAT TRANSFER CONSTANTS

<u>Pres., psi</u>	<u>M</u>	<u>$\frac{C}{10^6}(0.226 \text{ in. Tube})$</u>	<u>$\frac{C}{10^6}(0.143 \text{ in. Tube})$</u>
500	0.16	0.817	
1000	0.275	0.626	0.915
2000	0.500	0.445	0.545
3000	0.725		0.300

Recent work at Argonne has been correlated by Mumm by the equation

$$\frac{hD_e}{k} = \left[4.3 + 5.0 \times 10^{-4} \left(\frac{V_{fg}}{V_f} \right)^{1.64} x \right] \left[\frac{q''}{Gh_{fg}} \right]^{0.464} \left[\frac{GD_e}{\mu_f} \right]^{0.808} \quad (28)$$

in which

h = Coefficient of heat transfer based on the temperature difference between the wall and saturation temperature which may or may not be the temperature of the bulk fluid, BTU/hr ft² °F

D_e = equivalent diam, ft

k = thermal conductivity,
BTU/hr ft² °F/ft

V_{fg} = specific volume change from liquid to vapor phase,
ft³/lb

V_f = specific volume of liquid, ft³/lb

x = fraction of flow evaporated

q'' = heat flux, BTU/ft²hr

G = mass flow rate/unit area, lb/ft²hr

h_{fg} = latent heat of vaporization, BTU/lb

μ_f = viscosity of the liquid, lb/ft²hr

The equation was based on work carried out in a horizontal, electrically heated s.s. tube, 0.465 in. ID by 7 ft long. Up to 0.60 weight fraction was evaporated. The pressures varied from 45 to 200 psia, heat fluxes from 50,000 to 250,000 and flow rates from 70 to 280 lb/ft² sec.

If we analyze what may happen in a long tube or coolant channel in a water-cooled reactor, we find that the following conditions hold:

At the inlet the water will be below its boiling or saturation point. Heat will be transferred to the liquid in accordance with normal heat transfer as long as the temperature of the fuel element does not rise above some 25°F above the saturation temperature. When the heated surface rises much above the 25°F limit, nucleate boiling will start. The nucleate boiling will greatly increase the rate of heat transfer. As further increases in temperature occur, the fuel elements may become blanketed with steam and the heat transfer rate will drop. Finally, if the temperature becomes exceedingly high, heat transfer by radiation will begin to be important and the total heat transfer rate again increases. In general, the heat transfer surface will burn out or melt at the point at which the tube becomes blanketed with steam. Figure 3 shows the drop off in heat transfer coefficients for liquids boiling outside of horizontal pipes.

The pressure drop over a given tube will not change appreciably when nucleate boiling occurs. However, as soon as the fuel assemblies become blanketed with steam, the pressure drop will increase. For a single tube this would not be serious, since the incoming water would force the steam from the tube, but for a tube which is fed from a common header with many other tubes, the increase will reduce the flow in the first tube causing boiling and resulting in more steam production. It is normal to operate reactors well below the burnout region and most reactors have been run well below the nucleate boiling range. Operation in the nucleate boiling range would, qualitatively, permit higher power levels.

When a tube becomes covered by steam, there will be some wetting of the surface due to turbulence in the liquid-vapor mixture. This liquid will be rapidly evaporated causing local cooled spots. The net effect will be a much lowered rate of heat transfer. As noted above, as the quality increases the pressure will increase with a decrease in rate of flow. This analysis has led to the concept of boiling disease or the fear that burnout conditions will be aggravated by steam formation. In order to avoid these conditions, orifices may be placed in the tube inlets to cause definite pressure drops and thus maintain the rate of flow fairly constant and independent of fluctuations in pressure due to localized overheating with steam formation.

Let us consider a long, single tube cooled by water with an inlet velocity of 25 ft/sec and heated uniformly over its length at such a rate that the exit end would burn out. Under these conditions, we would expect to find

nucleate boiling begin in a region at which the temperature of the tube wall exceeds the bulk water temperature by about 25°F. At the exit of the tube the average steam temperature might exceed the average liquid temperature by 50°F or more. The conditions are illustrated qualitatively in Figure 4.

Argonne has run a very large number of burnout tests using vertical Ni tubes, 0.306 ID, 23-1/4 in. long and an exit pressure from 575 to 2000 psia. In these tests, the flow rate, total power, and system pressure were fixed at predetermined values and the heat exchanger partially or totally cut out of the flow circuit. Under these conditions, the inlet temperature at the test section increased slowly with time and the quality of the steam increased until burnout occurred. The inlet water temperature was read at burnout and the quality of the steam calculated by a heat balance. A few of the results are presented in Table V.

Table V

BURNOUT TEST DATA

Exit Pres., psia	Inlet Subcooling, °F	Inlet Velocity, ft/sec	Inlet Velocity, 10 ⁶ lb/ft ² hr	Heat Flux, 10 ⁶ BTU/ft ² hr	Exit Steam Quality, %
600	0	2.58	0.46	0.23	20.5
600	24	3.10	0.57	0.28	16.7
600	200	3.52	0.73	1.04	29.3
600	94	10.2	1.70	0.82	3.6
575	31	7.78	1.44	1.35	27.9
585	64	17.1	3.25	1.79	9.6
600	31	19.0	3.30	1.32	10.0
600	36	24.0	4.20	1.34	5.9
600	137	15.0	2.88	1.93	1.8
600	77	33.1	6.34	2.16	0.35
1000	4	10.3	1.72	0.87	19.4
1000	134	15.0	2.88	1.90	2.6
2000	159	4.24	0.76	0.66	13.4
2000	55	20.9	3.22	1.15	2.0
2000	0	24.6	3.45	0.75	12.1

Figure 4 - Qualitative Conditions in a Long Tube with Boiling

Slip ratio, $\frac{V_v}{V_\ell}$, was found to be 1.2 for 600 psia and 1.09 for 2000 psia in forced circulation boiling density tests. In this equation V_v = velocity of the vapor and V_ℓ = velocity of the liquid.

Usually, the heated surface temperature is very close to saturation with boiling steam and water mixtures. A number of equations have been developed to calculate the difference in temperature between the hot surface and the coolant; see the paper by Lottes, Monson, and Silvers. The range of superheat varies from about 60°F at atmospheric pressure to about 5°F at 2000 psia. The actual value at high pressure operation is of little interest since boiling coefficients are high and the limiting flux will usually be set by the thermal conductivity of the fuel element. However, there are critical values of steam quality at which the heat transfer coefficient drops and tube burnouts result. The critical points at which the coefficients drop seem to be a function of mass velocity as well as pressure, heat flux, and length. Burnout occurs at 80% quality and velocities of 1 ft/sec with the quality decreasing with velocities as indicated in the table previously given.

Liquid Metals in Pipes

Lyon gives the equation for heat transfer to liquid metals as

$$Nu = 7 + 0.025(Pe)^{0.8} \quad (29)$$

where $Pe = (Re)(Pr) = \frac{Dv\rho C_p}{k}$

This equation has been the subject of much argument and investigation as to whether heat transfer to liquid metals is dependent on viscosity. This equation indicates no dependence.

Apparently there are other factors, such as wetting the surface, that are important. For instance, Quittenton (Thesis, University of Toronto) believes that gas entrainment is important. At present no one knows. Quittenton reviews the available data and suggests that the above equation is of value as an upper limit. Most data fall below Lyon's equation, as indicated in Figure 5.

Johnson, Glabaugh, and Hartnett carried out extensive tests on Hg. They used Hg in turbulent flow with Re numbers of 14,000 to 400,000. Tests with He and argon indicated that gas entrainment between the circulated Hg and the steel was not responsible for the low heat transfer rates. Their results were about 38% under the Lyon equation.

The equation

$$\frac{hD_e}{k} = 5.8 + 0.02(Pe)^{0.5} \quad (30)$$

Figure 5 - Liquid Metal Heat Transfer Data

has been suggested as an improved version. If the liquid does not wet the surface, it has been suggested that the value of $1/2$ to $1/3$ the above be used. Actually, one has difficulty in understanding what wetting has to do with it. Cleanliness of surfaces and liquid metal and scales or films are most important.

The Liquid Metals Handbook gives

$$Nu = 4.4 \quad (31)$$

for viscous flow.

Brookhaven has run several series of tests of heat transfer coefficients of liquid metals flowing across tube banks. Using tubes 1.500 in. OD on an equilateral triangle with $11/16$ in. pitch, the coefficients varied with Re and angle of approach. The frontal coefficient was about twice that of the rear, as illustrated in Figure 6.

Example 1

Calculate the heat transfer coefficients for the materials listed below at a point in a reactor having 0.100 in. x 1.000 in. cooling channels with the various cooling media at 400°F and under the velocities and pressures indicated:

<u>Material</u>	<u>Velocity, ft/sec</u>	<u>Pres., psia</u>
He	80	200
CO ₂	80	200
H ₂ O	30	
Diphenyl	30	
Na	30	

Solution

Calculation of D per unit length

$$\text{Wetted surface/ft} = 0.183 \text{ ft.}$$

$$\text{Area} = \frac{0.100 \times 1.00}{144} = 0.000694 \text{ cu ft/ft}$$

$$D/4 = 0.00379 \text{ ft}$$

$$D = 0.01516 \text{ ft}$$

Figure 6 - Heat Transfer Coefficient Versus Angle Cross Flow

Need to check CO₂ only for variation from ideal gas laws:

$$\text{CO}_2: t_c = 31.1^\circ\text{C}; p_c = 73.0 \text{ atm.}$$

$$400^\circ\text{F} = 202^\circ\text{C}$$

$$t_r = \frac{273 + 31}{202} = 1.50$$

$$p_r = \frac{200/14.7}{73} = 0.187$$

$$z = 1.00$$

Data

Volume/mol. of CO₂ and He =

$$359 \times \frac{860}{460} \times \frac{14.7}{200} = 48.3 \text{ ft}^3$$

	$\frac{\rho}{\text{lb/ft}^3}$		μ		k	
		Ctpois.	$\frac{\text{lb}}{\text{ft sec}}$	$\frac{\text{lb}}{\text{ft hr}}$	BTU/ft ² hr °F/ft	C _p
He	0.0829	0.0265	0.0000181	0.0651	0.119	1.24
CO ₂	0.911	0.0222	0.0000150	0.0540	0.0184	0.265
H ₂ O	53.7	0.060	0.000041	0.148	0.380	1.000
Diphenyl	55.2*	0.374	0.000252	0.905	0.101*	0.508**
Na	53.2	0.381	0.000260	0.935	47.1	0.320

* Dowtherm A

** McAdams, pg. 401

For He, CO₂, H₂O, and Diphenyl use

$$\frac{h}{C_p G} = 0.023 \left(\frac{\mu}{DG} \right)^{0.2} \left(\frac{k}{\mu C_p} \right)^{0.6}$$

$$\text{He: } \frac{h}{1.24 \times 80 \times 0.0829 \times 3600} =$$

$$0.023 \left(\frac{0.0000181}{0.01516 \times 80 \times 0.0829} \right)^{0.2} \left(\frac{0.119}{0.0651 \times 1.24} \right)^{0.6}$$

$$h = 153$$

$$\text{CO}_2: \frac{h}{0.265 \times 80 \times 0.911 \times 3600} =$$

$$0.023 \left(\frac{0.0000150}{0.01516 \times 80 \times 0.911} \right)^{0.2} \left(\frac{0.0184}{0.0540 \times 0.265} \right)^{0.6}$$

$$h = 284$$

$$\text{H}_2\text{O}: \frac{h}{1 \times 30 \times 53.7 \times 3600} =$$

$$0.023 \left(\frac{0.000041}{0.01516 \times 30 \times 53.7} \right)^{0.2} \left(\frac{0.380}{0.148 \times 1} \right)^{0.6}$$

$$h = 16,400$$

$$\text{Diphenyl: } \frac{h}{0.508 \times 30 \times 55.2 \times 3600} =$$

$$0.023 \left(\frac{0.000252}{0.01516 \times 30 \times 55.2} \right)^{0.2} \left(\frac{0.101}{0.905 \times 0.508} \right)^{0.6}$$

$$h = 2910$$

$$\text{Na: } \frac{hD}{k} = 7 + 0.025 \left(\frac{Dv\rho C_p}{k} \right)^{0.8}$$

$$\frac{h \times 0.01516}{47.1} = 7 + 0.025 \left(\frac{0.01516 \times 30 \times 53.2 \times 0.320 \times 3600}{47.1} \right)^{0.8}$$

$$h = 34,600$$

Example 2

A tubular uranium fuel element is being considered for use in a power reactor. The slug dimensions are:

U I.D.	1.00 in.
U O.D.	1.75 in.
U Length	12.0 in.
Sheath, s.s.	0.010 in. thick
Bond, Na	0.001 in. Na

It is desired the maximum temperature of the U will be 1050°F. Assuming Na coolant flowing at 25 ft/sec, determine the maximum heat production per element and plot the temperatures across the center line of a fuel element at its vertical center assuming a bulk Na temperature of 600°F with center cooling only.

For the purposes of this problem assume that the heat production is uniform through the U and determine this heat production and the heat transfer coefficients for the maximum rates possible under the conditions stated. Neglect gamma and neutron heating in the Na, s.s., and surroundings.

Solution

See Figure 7.

$$k \text{ for U} = 0.0772 \text{ cal/cm}^2 \text{ sec}^2 \text{ C/cm}$$

$$= 0.0772 \times \frac{1}{4.133 \times 10^{-3}}$$

$$= 18.7 \text{ BTU/ft}^2 \text{ hr } ^\circ\text{F/ft}$$

$$600^\circ\text{F} = 371^\circ\text{C}$$

For inward heat transmission

$$t_0 - t_i = \frac{q}{2\pi k} \left[\frac{r_0^2}{r_0^2 - r_i^2} \ln \frac{r_0}{r_i} - 1/2 \right]$$

in which

r_0 = radius of outside of tube

r_i = radius of inside of tube

τ = heat production/unit volume, BTU/in.³

$$q = \tau \pi (r_0^2 - r_i^2)$$

For inward heating only:

$$t_0 = t_{\max} = 1050^\circ\text{F}$$

$$r_0 = 0.875; r_i = 0.500$$

$$q = \tau \pi (0.875^2 - 0.500^2) \text{ per in. length}$$

$$= \tau \pi (0.765 - 0.250)$$

Figure 7 - Tubular Fuel Element, Na Bonded with Reference Dimensions in Inches from Reference Line

$$q = \tau \pi 0.515 = 1.616 \tau \text{ BTU/in. length}$$

$$= 19.40 \tau, \text{ BTU/ft length}$$

$$q = \frac{\Delta t_m}{\underbrace{\frac{x_1}{k_1 A_1}}_{\text{Na Bond}} + \underbrace{\frac{x_2}{k_2 A_2}}_{\text{S.S.}} + \underbrace{\frac{1}{h_4 A_4}}_{\text{Na Coolant}}}$$

$$t_o - t_i = \frac{q}{2\pi k} \left[\frac{r_o^2}{r_o^2 - r_i^2} \ln \frac{r_o}{r_i} - 1/2 \right]$$

$$q = \frac{(t_o - t_i) 2\pi k}{\left[\frac{r_o^2}{r_o^2 - r_i^2} \ln \frac{r_o}{r_i} - 1/2 \right]}$$

$$19.40\tau = \frac{\Delta t_u 2\pi k}{\left[\frac{r_o^2}{r_o^2 - r_i^2} \ln \frac{r_o}{r_i} - 1/2 \right]} = \frac{\Delta t_m}{\frac{x_1}{k_1 A_1} + \frac{x_2}{k_2 A_2} + \frac{1}{h_3 A_3}}$$

$$\Delta t_u + \Delta t_m = 450^\circ \text{F}$$

$$\Delta t_u = 450 - \Delta t_m$$

$$19.40\tau = \frac{(450 - \Delta t_m) 2\pi k_u}{\frac{0.875^2}{0.875^2 - 0.500^2} \ln \frac{0.875}{0.500} - 1/2} =$$

$$\frac{\Delta t_m}{\underbrace{\frac{x_1}{k_1 A_1}}_{\text{Na Bond}} + \underbrace{\frac{x_2}{k_2 A_2}}_{\text{S.S.}} + \underbrace{\frac{1}{h_3 A_3}}_{\text{Na Coolant}}} = a$$

$$k_u = 18.7 \text{ for avg. of } 800^\circ \text{F U.}$$

$$\frac{(450 - \Delta t_m) 117.4}{\frac{0.765}{0.515} \ln 1.75 - 1/2} = a = \frac{(450 - \Delta t_m) 117.4}{1.488 \times 0.560 - 1/2} = \frac{117.4(450 - \Delta t_m)}{.832 - .500}$$

$$= 354(450 - \Delta t_m) = 159,000 - 354 \Delta t_m$$

To evaluate a we must evaluate all quantities as follows: For Na bond:

$$x_1 = 0.001 \text{ in.} = 0.0000833 \text{ ft}$$

$$k_1 = 35.3$$

$$A_1 = 2\pi r L = \frac{2\pi \cdot 0.4995}{12} \times 1 = 0.262 \text{ ft}^2/\text{ft}$$

Rate of heat production

$$q = 1042 \Delta t_m = 1042 \times 114 = 119,000 \text{ BTU/hr/ft of length}$$

$$q = \frac{119,000}{0.256} = 465,000 \text{ BTU/hr ft}^2$$

Temperature at radius 0.750

$$t_o - t_i = 1050 - t_{in} = \frac{119,000}{2\pi \cdot 18.7} \left(\frac{0.875^2}{0.875^2 - 0.750^2} \ln \frac{0.875}{0.750} - 1/2 \right)$$

$$1050 - t_i = 1012 \left(\frac{0.765}{0.765 - 0.601} \times 0.120 - 1/2 \right)$$

$$= 70.9^\circ\text{F}$$

$$t_i = 979^\circ\text{F}$$

For stainless steel sheath

$$x_2 = 0.010 \text{ in.} = 0.000833 \text{ ft.}$$

$$k_2 = 16$$

$$A_2 = 2\pi \frac{0.494}{12} = 0.259 \text{ ft}^2/\text{ft}$$

For Na film; use 1/2 Lyon Equation

$$\frac{hD}{k} = 7 + 0.025 \frac{(Dv\rho C_p)^{0.8}}{(k)}$$

$$D = \frac{2 \times 0.489}{12} = 0.0815$$

$$k = 35.3$$

$$v = 25$$

$$\rho = 0.860 \times 62.4 = 53.7 \text{ lb/ft}^3$$

$$C_p = 0.308$$

$$h \times \frac{0.0815}{35.3} = 7 + 0.025 \frac{(0.0815 \times 25 \times 53.7 \times 0.308 \times 3600)^{0.8}}{(35.3)}$$

$$h \times 0.00231 = 7 + 0.025(3430)^{0.8}$$

$$= 23.8$$

$h_3 = 10,300$; let us use $0.5 h_3$ to correct for high value of Lyon Equation. This is an assumption.

$$A_3 = 2\pi \frac{0.489}{12} = 0.256$$

$$\frac{\Delta t_m}{35.3 \times 0.262} + \frac{\Delta t_m}{16 \times 0.259} + \frac{1}{5150 \times 0.256} =$$

$$\frac{\Delta t_m}{0.00000900 + 0.000201 + 0.000748} = \frac{\Delta t_m}{0.000958}$$

$$159,000 - 354 \Delta t_m = 1042 \Delta t_m$$

$$1396 \Delta t_m = 159,000$$

$$\Delta t_m = 114^\circ\text{F}$$

$$\Delta t_u = 336^\circ\text{F}$$

Temperatures: Outside of U: 1050°F , see Figure 8.

$$\text{U Inside} \quad 714^\circ\text{F}$$

$$\text{SS Outside} \quad 714 - \frac{0.0000090}{0.000958} \times 114 = 714 - 1.1 = 713^\circ\text{F}$$

$$\text{SS Inside} \quad 713 - \frac{0.000201}{0.000958} \times 114 = 713 - 24 = 689^\circ\text{F}$$

Heat Distribution along a Reactor Coolant Passage

Goodman gives the following derivation of heat distribution along a passage in which there is a rather large temperature rise of the water and the heat production follows a cosine curve. In actual power reactors, the power production should be made more uniform throughout the tube length

Figure 8 - Temperature Distribution Through Rod and Sheath

and radially by various means of flattening. Flattening may be carried out by poisoning the center, enriching the outside, or providing various reflectors.

Let us consider a reactor in the form of a cube with sinusoidal energy production in the three directions, x , y , z , and zero at the surface and with no reflector. Then the rate of energy production per unit volume will be:

$$\frac{dQ}{dV} = \left(\frac{\pi^3}{8}\right) \left(\frac{Q}{L^3}\right) \cos \frac{\pi x}{2x_0} \cos \frac{\pi y}{2y_0} \cos \frac{\pi z}{2z_0} \quad (32)$$

Q = power production of reactor, BTU/hr

V = volume, ft^3

L = length of cooling passages, ft

x , y , z = distances outwards of the point under consideration, ft.

x_0 , y_0 , z_0 = distances from center to outside along principal axes, ft, $= \frac{L}{2}$

$\left(\frac{\pi^3}{8}\right) \left(\frac{Q}{L^3}\right)$ is the maximum rate of energy production at the center of the reactor, see Figure 9.

Let us consider a reactor in which the heat transfer passages are of uniform diameter and spacing without any orificing. The pressure drop and velocity will be constant but the temperature rise and the average temperature difference will vary from passage to passage. The energy production at the center will be

$$\frac{\pi^2}{4} x \text{ average} = 2.46 \text{ avg.}$$

Now let us orifice the passages so that the pressure drop and temperature rise is the same in all passages. The center passage will have no orifice. Then

$$\left(\frac{dQ}{dV_h}\right)_{\max} = 2.46 \left(\frac{dQ}{dV_h}\right)_{\text{avg}} \quad (33)$$

V_h = volume of heat transfer passages, ft^3

Q for a reactor may be evaluated as

Figure 9 - Reference Ordinates for Reactor Cube

$$Q = wC_p \Delta T = V_c C_p \rho \Delta T = hn \pi DL \Delta t$$

w = weight of coolant, lb/hr

ΔT = temperature rise of coolant, °F

V_c = volume of coolant, ft³/hr

ρ = density, lb/ft³

h = heat transfer coefficient, BTU/ft²hr °F

n = number of passages

D = diameter of passages, ft

L = length of passages, ft

Δt = average temperature difference between wall and bulk fluid, °F

For a single tube or passage

$$\Delta t_{\max} = \frac{\pi}{2} \Delta t_{\text{avg}}$$

It is possible to obtain an equation for the temperature of the tube wall as a function of the initial temperature of the coolant, over-all temperature rise of the coolant, the average temperature difference between the tube wall and the coolant, and the position along the tube, see Figure 10.

Thus, for a cubical reactor, we are interested in the heat flux in the central tube for the conditions of h and ρ at the center. For a reactor with $z = 0$ at the center we have, in terms of tube length L and variable position z ,

$$\Delta T_c(z) = C \int_0^z \cos \frac{\pi z}{L} \quad (34)$$

$$\Delta T_c = C \left[\left(\sin \frac{\pi z}{L} \right) + C' \right]$$

$$\text{At } z = 0; \Delta T_c(z) = \frac{\Delta T_c}{2}$$

$$C' = \frac{\Delta T_c}{2}$$

Figure 10 - Reference Index for Coolant Tube

$$\text{At } z = -\frac{L}{2}; \Delta T_c(z) = 0$$

$$0 = C \left(-\sin \frac{\pi z}{2} \right) + \frac{\Delta T_c}{2}$$

$$C = \frac{\Delta T_c}{2}$$

$$\text{At } z = \frac{L}{2}; \Delta T_c(z) = \Delta T_c$$

$$\Delta T_c = \frac{\Delta T}{2} \sin \frac{\pi z}{2} + \frac{\Delta T_c}{2} = \Delta T_c$$

$$\Delta T_c(z) = \frac{\Delta T_c}{2} \left(1 + \sin \frac{\pi z}{L} \right) \quad (35)$$

or the temperature of the bulk of the coolant equals $T_0 + \Delta T(z)$.

The temperature of the wall in terms of the bulk coolant temperature can be determined in the same way:

$$t_w - T_{\text{Bulk}} = \Delta T_{\text{max}} \cdot \frac{\text{heat generation at } z}{\text{max. heat generation}}$$

$$t_w - T_{\text{Bulk}} = \Delta T_{\text{max}} \cos \frac{\pi z}{L}$$

$$\text{At } z = 0; t_w - T_{\text{Bulk}} = \Delta T_{\text{max}}$$

$$\text{since } \cos 0 = 1$$

$$\text{At } z = -\frac{L}{2} \text{ and } +\frac{L}{2}, \cos = 0$$

Combining the two equations for ΔT_c and $t_w - T_{\text{Bulk}}$ we obtain

$$t_w = T_0 + \frac{\Delta T}{2} \left(1 + \sin \frac{\pi z}{L} \right) + \Delta T_{\text{max}} \cos \frac{\pi z}{L} \quad (36)$$

This equation may be differentiated to give the value of $t_{w\text{max}}$ and its location:

$$\frac{dt_w}{dz} = \frac{\pi}{L} \frac{\Delta T}{2} \cos \frac{\pi z}{L} - \frac{\pi}{L} \Delta T_{\text{max}} \sin \frac{\pi z}{L} \quad (37)$$

$$\text{At the max } \frac{dt_w}{dz} = 0$$

$$\frac{\Delta T}{2} \cos \frac{\pi z}{L} = \Delta T_{\max} \sin \frac{\pi z}{L} \quad (38)$$

From which

$$\begin{aligned} \tan \frac{\pi z}{L} &= \frac{\Delta T}{2\Delta T_{\max}} \\ \sin \frac{\pi z}{L} &= \left(\frac{\Delta T^2}{\Delta T^2 + 4\Delta T_{\max}^2} \right)^{1/2} \\ \cos \frac{\pi z}{L} &= \left(\frac{4\Delta T_{\max}^2}{\Delta T^2 + 4\Delta T_{\max}^2} \right)^{1/2} \end{aligned}$$

Substituting in (36) we obtain

$$\begin{aligned} t_{w\max} &= T_0 + \frac{\Delta T}{2} + \frac{\Delta T}{2} \left(\frac{\Delta T^2}{\Delta T^2 + 4\Delta T_{\max}^2} \right)^{1/2} \\ &\quad + \Delta T_{\max} \left(\frac{4\Delta T_{\max}^2}{\Delta T^2 + 4\Delta T_{\max}^2} \right)^{1/2} \end{aligned} \quad (39)$$

Simplifying:

$$t_{w\max} = T_0 + \frac{\Delta T}{2} + \frac{1}{2} \left(\Delta T^2 + 4\Delta T_{\max}^2 \right)^{1/2} \quad (40)$$

The location of the maximum will be

$$\sin \frac{\pi z}{L} = \left(\frac{\Delta T^2}{\Delta T^2 + 4\Delta T_{\max}^2} \right)^{1/2} \quad (41)$$

$$z = \frac{L}{\pi} \sin^{-1} \left(\frac{\Delta T^2}{\Delta T^2 + 4\Delta T_{\max}^2} \right)^{1/2} \quad (42)$$

see Figure 11.

In those cases in which $t_{w\max}$ is controlling, as it is in almost all actual reactors, an effort is made to improve this condition by use of reflectors to flatten the horizontal flux and some end reflectors to further flatten the reactor flux.

Example 3

Calculate the heat transfer surface, temperature and gpm of cooling water for a research reactor which is to be light water-cooled and moderated with a total heat production of 10,000 kw. It may be assumed that the following conditions hold:

Figure 11 - Empirical Curves for Tube Wall and Coolant Temperatures

Cosine flux over length of fuel elements 90% of total heat to be removed by cooling of the fuel elements;

Water velocity of 20 ft/sec;

Maximum fuel element surface temperature 200°F;

Water inlet temperature 120°F;

Fuel element length 30 in.;

Water openings 0.100 x 1.00 in., sides only heated;

Side reflector results in uniform cross sectional flux.

Solution

Heat to be removed per hour:

$$= 10,000 \times 0.90 \times 0.948 \times 3600$$

$$= 3.06 \times 10^7 \text{ BTU/hr}$$

General Data:

$$L = 30 \text{ in.} = 2.5 \text{ ft}$$

D: Wetted surface/ft (Perimeter)

$$= \frac{0.10 \times 2 + 1.00 \times 2}{12} = 0.183 \text{ ft}$$

$$\text{Area} = \frac{0.100 \times 1.00}{144} = 0.000694 \text{ ft}^2 \text{ or vol/ft}$$

$$D/4 = \frac{0.000694}{0.183} = 0.00379$$

$$D = 0.1516 \text{ ft (Equivalent)}$$

$v = 20$, neglect velocity changes

μ = varies with temperature although it should be sufficiently accurate to use average μ for any assumed exit temperature.

$$\rho = 61.8 \text{ lb/ft}^3$$

$$Re = \frac{DG}{\mu} = \frac{0.01516 \times 20 \times 61.8}{\mu} = \frac{18.75}{\mu}$$

$$C_p \cong 1.00$$

$$k = \text{variable}$$

$$G = 20 \times 61.8 \times 3600 = 4.45 \times 10^6 \text{ lb/ft}^2 \text{ hr}$$

Assume temperature rise of water = 30°F

$$t_{w\max} = T_0 + \frac{\Delta T}{2} + \frac{1}{2}(\Delta T^2 + 4\Delta t_{\max}^2)^{1/2}$$

$$200 = 120 + \frac{30}{2} + \frac{1}{2}(30^2 + 4\Delta t_{\max}^2)^{1/2}$$

$$65 = 1/2(900 + 4\Delta t_{\max}^2)^{1/2}$$

$$16,900 = 900 + 4\Delta t_{\max}^2$$

$$\Delta t_{\max}^2 = 4000$$

$$\Delta t_{\max} = 63.25^\circ\text{F}$$

$$\frac{h}{C_p G} = 0.023 \left(\frac{DG}{\mu} \right)^{-0.2} \left(\frac{\mu C_p}{k} \right)^{-0.6}$$

$$\mu = 0.489 \text{ centipoises at } 120 + 30/2 = 135^\circ\text{F}$$

$$k = 0.374 \text{ BTU/ft hr } ^\circ\text{F/ft}$$

$$\frac{h}{1.00 \times 4.45 \times 10^6} =$$

$$0.023 \left(\frac{18.75}{0.489 \times 0.000672} \right)^{-0.2} \left(\frac{0.489 \times 2.42 \times 1.00}{0.374} \right)^{-0.6}$$

$$h = 1.022 \times 10^5 \left(\frac{1}{57,000} \right)^{0.2} \left(\frac{1}{3.16} \right)^{0.6}$$

$$= 5730$$

$$q = hA \Delta t_{\max} \frac{2}{\pi}$$

$$3.06 \times 10^7 = 5730 \times A \times 63.25 \times \frac{2}{\pi}$$

$$A = 133 \text{ ft}^2$$

$$\text{No. of channels} = 133/0.416 = 318$$

$$\text{Vol/sec} = 0.000694 \times 318 \times 20 = 4.41 \text{ ft}^3$$

$$\text{Temp. rise} = \frac{3.06 \times 10^7}{4.41 \times 3600 \times 61.8} = 31.2^\circ\text{F}$$

Although we assumed 30°F , had we assumed that Δt_{max} was equal to $200 - 120 + 15$, or a straight line for the water heating with the maximum Al surface temperature at $L = 15$, the answer would not have differed greatly from the one obtained by use of Δt_{max} ; $\Delta t_{\text{max}} = 63.25$ in place of 65°F .

Temperature rise	31.2°F
Area	133 ft^2
GPM	1980 gal

Heat Exchanger Relations

$$\Delta t_m = \frac{\Delta t_1 - \Delta t_2}{\ln \frac{\Delta t_1}{\Delta t_2}} \quad (43)$$

This equation holds for parallel or countercurrent flow, see Figure 12.

The heat transfer performance of a shell-and-tube type of heat exchanger can be computed by application of the relation for three resistances in series; one, a cylindrical solid wall, and the other two, fluid films. The heat transfer equation for three resistances in series can be modified with the substitutions:

$$h_i = \frac{k_1}{x_1}, \quad h_o = \frac{k_3}{x_3}, \quad - \Delta t = \Delta t_m$$

$$A_i = A_1 = \pi L D_0 (D_i/D_0),$$

$$A_2 = \pi L (D_0 + D_i)/2 = \pi L D_0 \frac{D_0 + D_i}{2D_0}, \text{ and}$$

$$A_3 = \pi L D_0$$

Figure 12 - Mean Temperature Distribution

$$\begin{aligned}
 q &= \frac{-\Delta t}{\frac{x_1}{k_1 A_1} + \frac{x_2}{k_2 A_2} + \frac{x_3}{k_3 A_3}} = \frac{\Delta t_m}{\pi L D_0 \left[\frac{1}{h_i} \frac{D_0}{D_i} + \frac{2 D_0}{D_0 + D_i} \frac{x_2}{k_2} + \frac{1}{h_0} \right]} \\
 &= \frac{A_1 \Delta t_m}{\frac{1}{h_i} \frac{D_0}{D_i} + \frac{2 D_0}{D_0 + D_i} \frac{x_2}{k_2} + \frac{1}{h_0}} \quad (44)
 \end{aligned}$$

Method Used to Calculate Maximum Temperatures Inside a Reactor

The approximate neutron or heat flux in a small power reactor is shown in Figure 13.

In general a design should be such as to have as flat a flux distribution as possible. This prevents local hot spots and allows the maximum temperature rise for every coolant channel. In some cases other considerations enter, preventing the design of a flat flux reactor.

Hot Channel Factors

Hot channel factors are used as design factors in determining temperatures of the water and fuel plate for ease in computation and clarity. They are applied to water temperature rises, film temperature drops and heat fluxes to account for uncertainties and manufacturing tolerances. For example the hot channel factor for water temperature rise ($F_{\Delta t}$) is defined as the ratio of the maximum water temperature rise in a channel to the normal water temperature rise in that channel. Table VI lists the factors which should be considered as approximate values for each of these factors. The final factor is the product of all individual factors.

Table VI

HOT CHANNEL FACTORS

	$F_{\Delta t}$	F_{θ}	$F_{q''}$
Inability to predict flux	1.20	1.20	1.20
Variation in U^{235} concentration	1.10	1.20	1.20
Core eccentricity and variation in channel diameter	1.40	1.25	1.10
Effect of flow distribution	1.02	1.02	---
Effect of warping	1.04	1.03	---
Total Product	1.95	1.90	1.60

Figure 13 - Approximate Neutron or Heat Flux in a Small Power Reactor

$$\text{Average heat flux} = \frac{\text{Reactor Power}}{\text{Heat Transfer Area}} = q''_{\text{avg}} \quad (45)$$

$$q''_{\text{max}} = q''_{\text{avg}} (F_{q''}) \frac{\phi_{\text{radial}}}{\phi_{\text{avg}}} \cdot \frac{\phi_{\text{axial}}}{\phi_{\text{avg}}} \quad (46)$$

$$\text{Average Temperature rise} = \frac{\text{Reactor Power}}{C_p \times \text{flow rate}} = \Delta t_{\text{avg}} \quad (47)$$

If δ is defined as the ratio of the accumulated heat input into the channel up to the point at which the maximum heat flux occurs divided by the total heat input into the hot channel, then

$$(\Delta t) \text{ at } q''_{\text{max}} = (\Delta t_{\text{avg}}) \frac{\phi_{\text{radial}}}{\phi_{\text{avg}}} (F_{\Delta t})(\delta)$$

$$\text{Average film temperature difference} = \frac{q''_{\text{avg}}}{h} = \theta_{\text{avg}}$$

$$\theta_{\text{max}} = \theta_{\text{avg}} \frac{\phi_{\text{radial}}}{\phi_{\text{avg}}} \cdot \frac{\phi_{\text{axial}}}{\phi_{\text{avg}}} F_{\theta}$$

If the fuel plates have a very thin core, that is, it can be assumed that all the heat is generated at the center plane, the heat transmitted to each surface is $q/2$, the length of the resistance path through the cladding is $w/2$, and

$$\frac{q/2}{A} = -k \frac{\Delta t}{w/2}$$

$$\Delta t = \frac{-qw}{4Ak} = \frac{-q''_{\text{max}} w}{4k} \quad (48)$$

Then the maximum internal metal temperature is

$$T_{\text{metal}}(\text{internal}) = T_{\text{surface}} + \frac{q''_{\text{max}} w}{4k}$$

$$T_{\text{surface}} = T_{\text{saturation}} - C = T_{\text{water}} + \theta_{\text{max}}$$

$$T_{\text{water}} = T_{\text{inlet water}} + \Delta t_{\text{max at } q''_{\text{max}}}$$

where

w = thickness of fuel plate, ft

k = thermal conductivity, BTU/hr sq ft F

C = design constant to prevent local boiling. In the case of one actual reactor this constant is equal to 96°F. This is equivalent to designing so that local boiling starts at double the design power.

The above hot channel factors are admittedly very conservative. A reduction in these factors would permit a proportionate increase in the power of the reactor. It is this type of conservative design that is one cause of the high cost of estimated nuclear power. Experience in operating full scale power reactors will permit an increase in power and lowered costs.

Heat Transfer by Radiation

Heat transfer by radiation is proportional to the blackness of the bodies involved and the fourth power of the absolute temperature. The energy emitted per unit area is

$$E = q/A = e \sigma T^4 \quad (49)$$

in which e = the relative emissivity as compared with a black body.

σ is a constant depending on the units and called the Stefan-Boltzmann constant = 1.714×10^{-9} in BTU/ft²hr °R.

All bodies absorb and radiate energy in proportion to their temperatures and emissivities. A good absorber is a good emitter.

Heat being transferred between two plates, for example, is dependent on the temperature of both plates since heat will flow in both directions even though there is a large difference in temperature. The net exchange will be the difference between the heat flow by radiation in the two directions. For example, we may consider Figure 14.

Figure 14

If the plates are assumed large, so the end losses may be neglected, the heat emitted from plate 1 will be:

$$q/A = e_1 \sigma T_1^4$$

and the heat absorbed by plate 2 will be proportional to its absorptivity. Likewise the heat emitted from plate 2 will be:

$$q/A = e_2 \sigma T_2^4$$

and the proportion absorbed by plate 1 will depend on its absorptivity. Since the emissivity and absorptivity constants for materials are equal, we may write the equation for the net heat transfer from plate 1 to plate 2:

$$q_{1,2} = A_1 \sigma [e_1 T_1^4 - a_{1 \leftarrow 2} T_2^4] \quad (50)$$

The constant a depends on the various wave lengths of the emitted energy. A body that absorbs all wave lengths equally is called a "gray body." Generally, over-all results are about the same for solids and liquids (but not for gases) whether or not gray body conditions are assumed.

$$q/A = e_1 \sigma T_1^4 - a_{1 \leftarrow 2} T_2^4 \sigma$$

This equation may be written as

$$\frac{q_{1,2}}{A_1} = 1714 \quad e_1 \left[\left(\frac{T_1}{1000} \right)^4 - a_{1 \leftarrow 2} \left(\frac{T_2}{1000} \right)^4 \right], \text{ BTU/ft}^2 \text{ hr} \quad (51)$$

If gray body conditions are assumed, which is equivalent to stating that the emissivity of the lower temperature body is that of the higher temperature body, the equation becomes

$$q = 1714 A e_1 \left[\left(\frac{T_1}{1000} \right)^4 - \left(\frac{T_2}{1000} \right)^4 \right] \quad (52)$$

Emissivities vary from 0.9 to 0.95 for most materials. For polished metals and molten metals they vary from 0.1 to 0.30. Some values are indicated in Table VII.

Table VII
Total Emissivities of Surfaces

Surface	Temperature, °F	Emissivity
Highly polished Al	440 - 1070	0.039 - 0.057
Al oxidized at 110°F	390 - 1110	0.11 - 0.19
Highly polished Cu	242	0.023
Cu plate heated at 1110°F	390 - 1110	0.57
Fe oxidized at 1100°F	390 - 1110	0.64 - 0.78
Steel oxidized at 1100°F	390 - 1110	0.79
Steel plate, rough	100 - 700	0.94 - 0.97
Brick, red rough	70	0.93
Glass, smooth	72	0.937
Paints, except Al	100 - 212	0.80 - 0.96
Al paint, 10 to 26% Al	212	0.30 - 0.52
Refractories	1110 - 1830	0.65 - 0.90
Water	32 - 212	0.95 - 0.963

For most applications of radiant energy we must introduce geometric factors as the ratio of interchange per unit area to that which take place between infinite parallel planes or concentric spheres. The various references give some commonly used shape factors. The subject is quite complicated but we may assume all surfaces are gray as an engineering simplification. Further, if we assume that the heat lost through refractory nonactive walls is about equal to the heat transferred to these walls by conduction, the heat radiated to these walls is either reflected or reradiated. With these very great simplifications we may treat general furnace design problems readily. These problems include the heating of boilers, pipe stills, reheat furnaces, soaking pits, coke ovens, etc. We now introduce a shape factor, $\bar{F}$, into the equation as follows:

$$q = 0.171 A_1 \bar{F}_{1,2} \left[\left(\frac{T_1}{100} \right)^4 - \left(\frac{T_2}{100} \right)^4 \right] = 0.171 A_2 \bar{F}_{2,1} \left[\left(\frac{T_1}{100} \right)^4 - \left(\frac{T_2}{100} \right)^4 \right] \quad (53)$$

$\bar{F}$ may be evaluated for furnaces in which there are only one source and one sink with the rest of the area reflecting or reradiating by the equation

$$\bar{F}_{1,2} = \frac{A_2 - A_1 F_{1,2}^2}{A_1 + A_2 - 2A_1 F_{1,2}} \quad (54)$$

in which $F_{1,2}$ is the computed direct geometrical area factor. For a hollow cube heated on one side and radiating to the opposite side, $F_{1,2}$ is 0.20 and $\bar{F}_{1,2}$ is 0.523. For $\bar{\bar{F}}$, which is the factor to include the emissivities, the equation

$$\bar{\bar{F}}_{1,2} = \frac{1}{\frac{1}{\bar{F}_{1,2}} + \left(\frac{1}{e_1} - 1 \right) + \frac{A_1}{A_2} \left(\frac{1}{e_2} - 1 \right)} \quad (55)$$

has been derived. Similar equations have been derived for other conditions, but the above equations may be used in most cases by simple modification.

Irradiation from luminous and nonluminous flames is a particularly difficult problem. The luminous flame, such as produced by burning powdered coal, can usually be treated reasonably well since its size can be estimated. For semi-luminous flames, such as oil combustion, the size of the flame is difficult to estimate. For nonluminous flames the problem is still more difficult. Only CO_2 and H_2O of the normal combustion gases have sufficient emission bands that they need be considered. CO , SO_2 , and complex gases also radiate but are not usually present in combustion gases to an appreciable amount.

For CO_2 and H_2O the emissivities are dependent on the temperature, partial pressure of the gases, and the thickness of the flames. Charts with correction factors are given in all the standard texts. In use the walls of the combustion chamber reradiate to the gas, and back and forth. To evaluate the actual absorption, the emissivity of the absorption surface is taken halfway between its actual value and only the first impingement considered; thus

$$q_{G,s} = A 1714 \left(\frac{e_s + 1}{2} \right) \left[e_G \left(\frac{T_G}{1000} \right)^4 - a_G \left(\frac{T_s}{1000} \right)^4 \right] \quad (56)$$

Radiation Intercepted by a Shield

One of the important considerations is the effectiveness of radiation shields. Usually such shields are made of Al foil, are very thin, and assume an intermediate temperature between the radiating surfaces. Since the heat must be transferred in series:

$$Q = \frac{\sigma A}{\frac{1}{e_1} + \frac{1}{e_{\text{shield}}} - 1} (T_1^4 - T_{\text{shield}}^4) = \frac{\sigma A}{\frac{1}{e_{\text{shield}}} + \frac{1}{e_3}} (T_{\text{shield}}^4 - T_2^4) \quad (57)$$

But $e_1 = e_{\text{shield}} \neq e_2$

$$T_{\text{shield}}^4 = \frac{1}{2} (T_1^4 + T_2^4)$$

$$Q_n = \frac{1}{n + 1} Q \quad (58)$$

for multiple shields.

Effect of Solar Radiation on a Body at High Altitudes

The heat absorbed from the sun on an object high above the earth at which all the heat is transferred by radiation will be

$$q_s = 480 A e_B \cos \theta_B, \text{ BTU/hr} \quad (58)$$

in which

A = area of body

e_B = emissivity (absorptivity) of the body

θ_B = angle of plate with the sun. For odd-shaped articles, the projected area is used.

The radiation between a flat plate in space and interplanetary space, on the assumption that one side is perfectly insulated and the interplanetary space is at absolute zero, is

$$q_{sp} = -A 1714 e_{plate} \left(\frac{T_{plate}}{1000} \right)^4 \quad (60)$$

Equations for Temperature Gradient and Thermal Stresses for Various Shapes

The stress equations for some simple shapes are tabulated below. The equations are based on the theory of elastic deformation and do not take into account plastic flow. They are estimates only:

1. Flat Plate, Heat Generated at Center

Figure 15

$$\Delta T = \frac{\tau w^2}{4k} \quad (61)$$

$$S_z = \frac{\Delta T}{2} \frac{E\alpha}{1-\mu} = \frac{\tau w^2}{8k} \frac{E\alpha}{1-\mu} \quad (62)$$

in which

τ = equivalent heat generated per unit volume

α = coefficient of thermal expansion

μ = Poisson's ratio

2. Flat Plate, Uniform Heat Generation

Figure 16

$$\Delta T = \frac{\tau w^2}{8k} \quad (11)$$

$$S_z = \frac{2}{3} \Delta T \frac{E\alpha}{1-\mu} = \frac{2}{3} \frac{\tau w^2}{8k} \cdot \frac{E\alpha}{1-\mu} \quad (63)$$

3. Square Bar, Uniform Heat Generation

Figure 17

$$\Delta T = \frac{\tau w^2}{12k} \quad (64)$$

$$S_z = \frac{5}{8} \Delta T \frac{E\alpha}{1-\mu} = \frac{5}{8} \cdot \frac{\tau w^2}{12k} \cdot \frac{E\alpha}{1-\mu} \quad (65)$$

4. Rod, Uniform Heat Generation

Figure 18

$$\Delta T = \frac{\tau r^2}{4k} \quad (66)$$

$$S_z = \frac{\Delta T}{2} \cdot \frac{E\alpha}{1-\mu} = \frac{\tau r^2}{8k} \cdot \frac{E\alpha}{1-\mu}$$

5. Cylinder with Heat Generated at One Surface

Figure 19

$$\Delta T = \frac{q}{2.74k} \log \frac{D_2}{D_1} = 1.15 \tau \frac{(r_2^2 - r_1^2)}{k} \log \frac{D_2}{D_1} \quad (68)$$

$$S_z = \Delta T \frac{E\alpha}{1-\mu} f_1 = \frac{q}{k} \cdot \frac{E\alpha}{1-\mu} \cdot f_2 \quad (69)$$

Table VIII

D_1/D_2	Heat Flow In		Heat Flow Out	
	f_1	f_2	f_1	f_2
0.2	.730	.0933	.270	.0345
0.3	.686	.0670	.317	.0303
0.4	.645	.0470	.358	.0261
0.5	.610	.0337	.390	.0216
0.6	.575	.0234	.420	.0171
0.7	.555	.0158	.442	.0125
0.8	.535	.0095	.465	.0082
0.9	.515	.0045	.485	.0042
1.0	.500	---	.500	---

6. Cylinder with Uniform Heat Generation and Both Surfaces Cooled

Figure 20

$$\Delta T = \frac{q}{k} f_1 \quad (70)$$

$$S_z = \frac{q}{k} \left(\frac{E\alpha}{1-\mu} \right) f_2 \quad (71)$$

where q = heat generation per unit length, BTU/ft
 D = diameter of highest temperature

Table IX

D_1/D_2	f_1	f_2	D_1/D
1.00	0.000	---	---
.90	.004	.00138	.948
.80	.009	.00285	.891
.70	.015	.00460	.834
.60	.023	.00655	.761
.50	.035	.00885	.680
.40	.050	.01155	.590
.30	.070	.0145	.488

LECTURE 10

MATERIALSReferences

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Introduction

In Lecture 1, we discussed briefly the materials that can be used in reactors. It is the purpose of this lecture to discuss the selection of materials and some special subjects like toxic materials in some detail.

Materials are required in power reactors for a number of purposes: fuel, fuel diluent and cladding, fertile material to form new fuel, moderators in thermal reactors, coolants, control elements, structural materials, and neutron and temperature-sensing devices. The selection of many of these materials depends upon the normal industrial requirements plus a consideration of neutron properties. It should be remembered that the central problem is heat removal.

Selection Criteria

The criteria on which materials are selected might include the following items:

1. Cost
2. Availability
3. Neutron absorption and scattering cross sections
4. Corrosion resistance
5. Heat Transfer
6. Resistance to radioactive damage and to dimensional changes
7. Reactivity buildup
8. Toxicity
9. Physical properties such as ductility, tensile strength, and creep resistance at high temperatures
10. Mechanical properties, workability, ease of welding
11. Pyrophoric nature
12. Chemical properties

The neutron properties limit the practical materials, regardless of cost, to only a few elements. Some of these are presented in Table III of Lecture 1.

The Atomic Energy Commission through its contractors and national laboratories has had to develop Zr as a cladding or structural material and Be and BeO as moderator materials. Considerable development effort has been spent on Al, stainless steels, and graphite in addition to the work on U, Th, and the metals just mentioned.

The number of neutrons emitted per fission and the net numbers of neutrons available for use after the nonfission absorption is considered are important. These are illustrated in Table I of Lecture 2.

A study of these two tables along with the classified fission and absorption cross sections of Th in the thermal range, and of Pu and the U isotopes in the fast range will permit a selection of materials. For example, H is excellent in respect to moderating efficiency but has a high capture cross section. D is good as a moderator and excellent in respect to fission capture. Since O has a very low cross section, it is obvious that D₂O should be considered as a moderator.

Fuel

The choice of fuel to use in a reactor is dependent upon the type of reactor and its size. Small reactors with power levels up to a few thousand kw of heat are most cheaply fueled with fully enriched U^{235} since the fuel cost is small compared to other costs. Special research reactors even at higher powers will be U^{235} -fueled because of the high excess reactivity possible with relatively low inventories.

For power reactors, the fuel will be chosen in combination with the fertile material in order that as much new fuel will be produced as possible. There are several reasons for this: (1) the fuel more nearly maintains its original reactivity, (2) reloading is less frequent, and (3) the cost of the fuel is reduced drastically.

The U^{235} in natural U makes this an excellent fuel from a cost standpoint. Its drawback is that slight enrichment is required for use in H_2O and Na-moderated thermal reactors. When fuel grade Pu (high Pu^{240} content) becomes available from operation of a reactor, this can be used for part or all of the slight enrichment required. This places a considerable premium on natural U, as the U^{235} present in natural U is the cheapest fissionable material. However, only a part of this U^{235} can be used. The use of U^{233} and Th may prove to be economical because it may prove possible to obtain a higher conversion ratio in a thermal reactor than with U^{235} . The U^{233} formed in Th and the Pu formed in U can be completely separated by chemical processes.

For fast reactors Pu and U^{238} will be used because of their high conversion ratio of 1.5 or greater.

Very little can be said with certainty about the burnup of fuel elements that can be obtained in power reactors. The formation of the fission products and slowing down of the neutrons along with the temperature changes cause irradiation damage to the fuel. The fission products absorb neutrons and reduce the reactivity. Experiments on small samples have given indications that fuel elements can be made to sustain 2 atoms per hundred of burnup but this has not been demonstrated on a large scale.

The form in which the fuel is used will depend on the reactor type. Metallic U and Th have been used in most reactors since the metal has fair thermal conductivity, $k = 14.5$ for U and 23.6 to 24.4 for Th at 0 to 1000°F in Btu/hr ft² °F/ft. The main difficulty with U is that it is an anisotropic metal and tends to alter its shape due to temperature changes. In homogeneous reactors, a solution or slurry is used.

In the future, we should expect increased emphasis on slurries, especially of the oxides. Such slurries have the advantage of ease of preparation as well as providing no heat transfer or radiation damage problems. But they do have the disadvantages of danger of settling and erosion of the equipment.

Cladding Materials

The choice of a cladding material is dependent upon its corrosion resistance to the coolant and its ability to cover or protect the fuel. Other requirements are reasonable cost, high thermal conductivity, good tensile strength and good creep resistance, high ductility, ease of forming by rolling, extrusion, etc., and the ability to be easily removed or dissolved in the chemical separations plant.

The cladding must either bond to the fuel and change dimensionally with the fuel and prevent contact between the fuel and coolant or it must contain a thermal bond such as liquid Na between the fuel and clad. These things are essential to prevent mixing the fission products and fuel with the coolant and yet maintain high heat transfer rates. The high heat transfer rates are essential to low power costs.

Table I presents a few of the properties of interest in selecting a cladding material. While several materials are listed, the only practical ones are Al, Zr, and stainless steel. None of these three materials are desirable in all respects. The actual material used will depend upon the choice of coolant. If H_2O or D_2O are to be used, either Al or Zr would be used. Due to the high vapor pressure of water, the high pressures possible to be obtained with Zr and stainless steel clad fuel elements, would not add much in the way of efficiency. Al can be used up to $230^{\circ}C$ or higher, and Zr can be used up to $350^{\circ}C$ or so. Stainless steel could be used up to $650^{\circ}C$ or so except for the high pressures in the reactor and the high absorption of neutrons. Al must be especially prepared and protected to be useful above about $200^{\circ}C$. This is discussed later under Corrosion. Zr has the disadvantage of high costs, especially at the present state of development of its technology. Under certain conditions it may be pyrophoric in the chemical plant. Both Al and Zr are high in the electromotive series and potentially might react with water in case of a very severe power transient.

Al may be bonded to U by an alloy of AlSi. Other bonding procedures show promise. Zr readily bonds to U. In respect to heat transfer, Zr is about equal to stainless steel. Zr maintains its strength reasonably well at elevated temperatures, dropping from about 35,000 psi at room temperature

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Table I

PROPERTIES OF MATERIALS

Material	Purpose	Density, gm/cm ³	Approx. Cost, \$/lb	Melting Point, °C	Sp. Heat, cal/gm°C	Thermal Expansion, °C x 10 ⁶	Thermal Conductivity, Btu/lb ft ² °F/ft	Modulus of Elasticity, psi x 10 ⁶	Poisson's Ratio	Hardness, EHN	Tensile Strength, psi x 10 ³		Elongation in 2%		Thermal σ _B , Barns
											Annealed	Cold Worked	Annealed	Hardened	
Al	Clad Structural	2.7	0.30 to 0.60	660	0.22	24	118	10	0.3	20-25	13	24	35	5	0.22
Be	Moderator	1.85	80	1315	0.5	12	80	42		110	45		1.2		0.010
Ta	Structural Fast	16.6	65	2996	0.036	6.5	32	27		75-125	50	125	40	1	21
Ti	Structural Fast	4.5	6.5	1725	0.129	8.5	---	16.8		200	80	122	24	7.5	5.8
Zr	Clad Structural	6.5	15-40	1845	0.175	4.96	14.2	12	0.33		35	85	31	18	0.18
18-8 Stainless Steel	Clad Structural	7.9	0.50	1400±	0.12	16.7	16.2	29	0.28	160	90		45		2.9
Mg	Clad	1.74	0.25	651	0.25		93	6.5	0.35	50	32-46	32-50	16-12	8-3	0.06

to about 15,000 psi at 350°C. It is readily formed by rolling, extrusion, and welding by shielded arc. Since the oxide is soluble in the metal, welds are very sound.

For use with Na coolant, Zr or stainless steel might be used since the high temperatures available with the stainless steel and liquid Na system would lead to high thermal efficiencies. In this respect the North American Aviation group designing the SGR have considered both materials. The graphite will be clad with Zr and quite possibly stainless steel will be used to clad the fuel rods.

The problem in fast reactors is entirely different since all materials have low absorption cross sections for neutrons. Stainless steel and Na coolant are natural choices and lead to high temperatures and high thermal efficiencies. Here the fuel temperature is apt to be the limiting factor. Both Ta and Ti have been considered as cladding materials for use in fast heterogeneous reactors. They may well be used in the future.

Moderators

H₂O, D₂O, Be, BeO, and C are all usable moderators. Of these, H₂O has many advantages as listed in Table I of Lecture 3, but the disadvantages make it questionable. D₂O, Be, and BeO are all expensive, and C requires large volumes. BeO is cheaper than Be and also has a higher density of Be atoms. Be will probably be used only for special purposes, such as research reactors like the MTR. The decision between H₂O, D₂O, and C must be on a complete cost analysis of the various reactor types. Data that will become available as a result of construction of the five reactors as a part of the Five Year Plan will help answer this question.

In homogeneous reactors, D₂O is the only practical coolant-moderator for a UO₂SO₄ solution, but H₂O or Na might be used in slurry reactors. Be or BeO might be used in block form.

Coolants

The choice of coolant is also of primary importance and is tied in with the decision as to which type of reactor system is chosen. Most of the advantages and disadvantages of the various coolants have been listed in previous lectures. D₂O has the disadvantage of high cost. Except in boiling reactors, a large percentage of the moderator is outside of the reactor in

the coolant system. Nevertheless, if the cost of production can be reduced to a reasonable figure, D_2O will probably be extensively used not only in boiling but in pressurized power reactors.

Corrosionwise, both H_2O and D_2O are poor, as both are oxidants, reductants, acids, and bases under different operating conditions and are, therefore, very active chemically. Na is much less active but does have the disadvantage of high affinity for O_2 and H_2O ; it must be used with care.

Both H_2O and D_2O may be cleaned up readily and maintained in a very pure condition with 1 ppm or less of total solids present. Under these conditions Al and Zr corrode very slowly compared with ordinary water conditions. The use of mixed bed ion exchange systems to clean a small bypass stream is all that is required. The high purity water will corrode the stainless steel in the external system slowly and carry the corrosion products into the reactor. As these corrosion products contain Co and Mn, they become radioactive and it is necessary to remove them by ion exchange and their oxides by filtration.

Both H_2O and D_2O become intensely radioactive due to the (n,p) reaction with oxygen-16 forming nitrogen-16. The nitrogen-16 decay process includes the emission of very hard gamma rays, 7.5 and 6.1 Mev, but the half-life of the decay process is very short, 7.5 sec. This means that the nitrogen-16 will decay out in a minute or so. However, all piping and pumps must be heavily shielded to protect personnel and electrical instruments during operation.

Na has the advantage of high temperatures at low pressures. However, it has appreciable neutron absorption ($0.49 \times 10^{-24} \text{ cm}^2$) and freezes at 97°C . It is quite inert in respect to solution of steel, Zr, or even Al. Means have been developed for handling Na, and it can be readily cleaned. It becomes very radioactive with a half life of 15 hours. This means that several days are required before Na becomes reasonably low in activity and the system has to be heavily shielded at all times.

Na is the only practical coolant for use in fast heterogeneous reactors. In fast homogeneous reactors, the coolant must be Na, Bi, Pb or fused salts. Na could, of course, be used to transport a slurry. Bi and Pb both have low heat capacity on a weight basis and Bi forms the α -emitter Po.

Organic materials, such as diphenyl, diphenyl oxide, or the eutectic of the two have been proposed for coolants. While these materials have low pressures at high temperature, are good moderators, are reasonably thermally stable, and are not corrosive, they have the serious objections of high decomposition rates due to neutron damage, low heat transfer coefficients, and are hard to hold in a system. Their decomposition is so high that there appears little reason to consider them.

Structural Materials

In general, Al, Zr, and stainless steel structural materials will be used in a reactor when these same materials are used as the fuel cladding materials. In homogeneous reactors, stainless steel, Zr, Ti, or Ta will be used for most solutions. For liquid metals special stainless steel or alloys will be required.

For a large homogeneous power reactor, we could use a solution of UO_2SO_4 (natural U) in D_2O as fuel, moderator and coolant with stainless steel as the material of construction. The use of enriched fuel with Zr separating the reactor core from the blanket may be developed to give separate core and breeding blankets.

The external systems in most reactors have been built of stainless steel since the first requirement has been continuous trouble-free operation. In the future, more low alloy steels will be used as their use becomes demonstrated and the emphasis on the design is placed more on economy.

Shielding Materials

Shields are usually broken up into two parts. First, there is the so-called "thermal" shield. The purpose of the thermal shield is to reduce the neutron level as well as the gamma-ray level to levels about 10^8 per cm^2 per second. Since many fast neutrons reach the shield without being moderated or slowed down, the shield should contain a large amount of light hydrogen to slow down these neutrons. The shield should also absorb thermal neutrons rapidly without emitting additional gamma rays. At the same time, it is desired to absorb as many gamma rays as possible. Therefore, a dense shield is desired. And above all, the shield should be made of cheap materials.

The materials which have been used are hydrogenous materials such as wood and water combined with boron and iron to absorb the thermal neutrons. Iron absorbs neutrons satisfactorily, but at the same time gammas are emitted. Boron strongly absorbs neutrons without the emission of gamma rays. So the thermal shield consists of alternate layers of water and steel with a thin layer of boron carbide, held in aluminum, placed over the inner steel layer, or boron steel may be used. When energy of the neutron and gamma rays is absorbed, the thermal shield is heated so that it must be cooled. This is simple for a water-iron shield as cooling water can be pumped through the shield.

Outside of the thermal shield is the biological shield which reduces the neutron and gamma rays to a tolerable level for humans. As mass and neutron absorption only are needed, ordinary concrete or concrete with high specific

gravity aggregate such as barytes or limonite may be used. Thirteen inches of ordinary concrete reduces the gamma-ray intensity roughly by a factor of 10, so 8 ft or so of concrete is adequate.

Control Elements

For thermal reactors, B, Cd, or Hf are used and the cladding materials are the same as that used on the fuel.

Summary

A list of the materials that appear of practical use today in large central power stations is presented in Table II.

Table II

MATERIALS FOR POWER REACTORS

	<u>Heterogeneous</u>		<u>Homogeneous</u>	
	<u>Thermal</u>	<u>Fast</u>	<u>Thermal</u>	<u>Fast</u>
Fuel	U ²³⁵ , U ²³³	Pu ²³⁹	U ²³⁵ , U ²³³	Pu ²³⁹
Fertile	U ²³⁸ , Th	U ²³⁸	U ²³⁸ , Th	U ²³⁸
Moderator	H ₂ O, D ₂ O, C	None	D ₂ O	None
Coolant	H ₂ O, D ₂ O, Na	Na	D ₂ O	Bi, Pb, Fused Salts
Cladding and Structural	Al, Zr, S.S.	S.S.	S.S., Zr	S.S.
Control	B, Cd, Hf	None	B, None	None

Toxicity

Many reactor materials are toxic as are radium, etc., since they give off more or less active α particles. Table III gives the half-lives of various isotopes of interest.

The Bureau of Standards Handbook presents data on permissible concentrations. The danger due to the various isotopes depends on their location in the body, and the half-life, and the energy produced. Ra, Pu,

and Sr are deposited in critical body tissue where the rate of turnover is low or the biological half-life is very long. P^{32} , Na^{24} , and S^{35} have biological half-lives of a few days or weeks and are, on an energy basis, comparatively less harmful.

Table III

HALF LIFE AND ENERGIES

<u>Isotope</u>	<u>Decay</u>	<u>Half Life</u>	<u>Spontaneous Fission Half Life</u>
Th^{232}	α	$1.39 \times 10^{10}y$	$1.4 \times 10^{18}y$
U^{233}	α	1.62×10^5y	
U^{234}	α	2.48×10^5y	$2 \times 10^{16}y$
U^{235}	α	7.13×10^8y	$1.9 \times 10^{17}y$
U^{236}	-	2.39×10^7y	
U^{237}	β^-	6.75d	
U^{238}	α	4.49×10^9y	$8.0 \times 10^{15}y$
Np^{239}	β^-	2.33d	
Pu^{239}	α	24,400y	$5.5 \times 10^{15}y$
Pu^{240}	α	6,580y	
Pu^{241}	β^-	14y	

Natural U contains U^{238} , U^{235} , and U^{234} with the controlling half-life of U^{238} of 4.49×10^9y . One curie of natural U requires 1.5×10^6g so that the maximum permissible amount of the mixture in the body is $0.02 \mu c$, or $0.03 gm$ which is an unlikely dose and probably would be a chemical hazard. On the other hand, U^{233} only requires 100 gm/curie and Sr^{90} with a half life of 25 years, which requires only $6.3 \times 10^{-3} g/curie$, are much greater hazards. In fact Sr^{90} is probably the most hazardous fission fragment. Very short lived radioisotopes such as N^{16} with a half life of 7.35 sec, $10^{-11} gm/curie$, cannot be very dangerous. Those materials retained in the body which have half lives of 5 to 50 years are the most dangerous.

Actually it is the energy of the particles produced that does the harm. Thus, the energy of the α particles from Ra^{226} plus half the energy of Rn^{222} and its α emitting daughters are 14.5 Mev. The energy of Pu^{239} is 5.16 Mev, so one would expect Ra^{226} to be roughly 3 times as hazardous on this basis. However, the Pu^{239} is concentrated in the radiosensitive portion of the bone and may be more hazardous than the Ra^{226} .

The permissible amounts of some isotopes of interest are given in Table IV.

In addition to the harmful effects of the radioactive isotopes Be is chemically a dangerous poison, particularly in the oxide or sulfate form. The maximum permissible amount of powdered Be in each laboratory module for use in experimental work is 7 mg. This same figure applies also to Cd and chromic acid, while 11 mg of As, Pb, and U may be present in powdered form.

Berylliosis is selective in its attack as only one out of eight exposures contract the disease. The disease has many outward similarities to pneumonia. All work on machining must be done on especially enclosed and ventilated machines. Air samples are taken during all operations.

Limits on permissible amounts of radioactive materials and gaseous poisons would be more significant if tabulated as dissociations/minute/meter³ or as gm/m³ for the reason that a man breathes about 10 m³ per working day and 10 m³ during the rest of his day. A few materials are tabulated in Table V in this way and body burdens are given in Table VI.

Disintegrations x energy x distribution factor (in critical organ) gives maximum permissible critical amounts. The inhaling of fission products or irradiated fissionable materials is particularly dangerous.

Tritium is a bad actor. In the form of HTO or DTO in the atmosphere, the lungs will retain 97% of that taken in by breathing and will absorb an equal weight through the skin. Wearing a sealed mask will not give adequate protection. At NTP conditions T₂ has 2.5 curies/cm³.

Plutonium and perhaps U²³³ form aerosols, probably by breakup of the oxide by the emitted α particles. This makes it exceedingly difficult to process these materials. A very considerable amount of work has been done at Los Alamos on the corrosion of Pu. At low relative humidity, the rate of oxidation appears to be very low, but at high relative humidity, the rate is high. Pu may be handled in high purity He or in very dry air. U²³³ and Pu are about 10⁶ and 10⁷ as hazardous as our most dangerous cancer-forming chemicals.

The following discussion was prepared by Donald P. O'Neil at Argonne:

"The principle avenues of absorption for toxic materials are threefold: ingestion, inhalation, and absorption through the skin. Systemic industrial poisonings, i.e., total body poisonings, usually result from inhalation of the toxic agent. This is fairly obvious when gases like carbon monoxide and dusts such as silica are concerned. With volatile liquids, however, there is the additional possibility of absorption through the skin as is the case also with some gases such as hydrogen cyanide. Generally, however, skin

Table IV

MAXIMUM PERMISSIBLE AMOUNT OF RADIOISOTOPE IN TOTAL BODY AND MAXIMUM
PERMISSIBLE CONCENTRATION IN AIR AND WATER FOR CONTINUOUS EXPOSURE

<u>Isotope</u>	<u>Particle</u>	<u>Organ, gm</u>	<u>Microcuries in Total Body</u>	<u>Microcuries per ml of Water</u>	<u>Microcuries per ml of Air</u>	<u>Grams per m³ of Air</u>
Po ²¹⁰ Sol.	α	Spleen, 150	0.02	3×10^{-5}	2×10^{-10}	
Po ²¹⁰ Insol.	α	Lungs, 10 ³	7×10^{-3}		7×10^{-11}	
U Nat. Sol.	α	Kidneys, 300	0.2	7×10^{-5}	1.7×10^{-11}	
U Nat. Insol.	α	Lungs, 10 ³	0.009		1.7×10^{-11}	
U ²³³ Sol.	α	Bone, 7×10^3	0.04	1.5×10^{-4}	1×10^{-10}	1×10^{-8}
U ²³³ Insol.	α	Lungs, 10 ³	0.008		1.6×10^{-11}	1.6×10^{-9}
Pu ²³⁹ Sol.	α	Bone, 7×10^3	0.04	1.5×10^{-6}	2×10^{-12}	3.0×10^{-11}
Pu ²³⁹ Insol.	α	Lungs, 10 ³	0.008		2×10^{-12}	3.0×10^{-11}
H ³	β^-	Total Body, 7×10^4	10^4	0.2	2×10^{-5}	
Be ⁷	None	Bone, 7×10^3	670	1	4×10^{-6}	

Table V

MAXIMUM PERMISSIBLE AMOUNTS OF ISOTOPES IN AIR

<u>Isotope</u>	<u>Dis./min/m³</u>	<u>Remarks</u>
Sr ⁸⁹	4.4 x 10 ⁴	
Sr ⁹⁰ + Yttrium ⁹⁰	444	
Ra ²²⁶	18	
U ²³³	222	Soluble
	36	Insoluble
Pu ²³⁹	4.4	Soluble, bone
	4.4	Insoluble, bone
Po ²¹⁰	444	Soluble
	156	Insoluble

Table VI

MAXIMUM PERMISSIBLE TOTAL BODY BURDENS

<u>Isotope</u>	<u>Micrograms</u>
Nat. U	13 Insoluble
U ²³³	0.84 Insoluble
Pu ²³⁹	{ 0.65 Soluble
	{ 0.13 Insoluble

absorption is of secondary importance to absorption by inhalation when systemic effects are concerned. Local effects such as dermatitis due to direct contact with a liquid or a solid are, of course, common.

"With solid substances inhalation and ingestion are the most important avenues of intake and toxic solids are most dangerous when dispersed in the air as dusts, smokes, or fumes. Ingestion of toxic substances along with food, where workers are careless about washing, no doubt does occur although the classic cases of industrial poisoning via ingestion were those of the radium dial painters who absorbed radium mainly from the practice of using their lips to point the brushes wet with radium-bearing paint. This, of course, is one of a comparatively few isolated instances of exceptional practice. In the majority of cases the toxic substance entering the body is suspended in the air breathed by the worker.

"In entering the body, the air passes to the lung where the oxygen in the air passes through the thin wall into the blood stream.

"If the air inhaled contains other gases or vapors, they ordinarily reach the blood in a similar fashion, although, if the gas is water soluble, much of it may be deposited on the upper moist walls of the respiratory system where it finds its way into the blood stream, although less rapidly than through the alveoli.

"With dust, smoke, or mist the particle size is the governing factor in determining whether it reaches the alveoli and is absorbed into the body or whether it impinges upon a mucous surface and is brought back to an oral opening by ciliary action. It has been determined by extensive investigation that very few particles greater than 5-7 microns in diameter reach the alveoli. As a general rule, the smaller the particle size, the greater the proportion which reaches the alveoli and the faster and more severe the action. This is an important point in estimating the extent of a hazard when comparing two operations. Not only must the concentration and toxicity be considered, but also the particle size of the dust or fume generated.

"Once the solid particle gets into the lung it passes into the blood stream if soluble or remains in the lung where it is absorbed slowly. This points up another factor in the over-all toxicity - the solubility. For example, lead monoxide is much more toxic than lead carbonate or lead chromate because of its greater solubility in blood serum.

"Once materials are absorbed in the blood they are transported throughout the body and are deposited to some extent in appropriate tissues, according to the fundamental laws of physical chemistry.

"The removal of toxic materials from the body takes place mainly through the respiratory tract, the intestinal tract, and the urinary tract. Gases such as carbon monoxide and radon are removed mainly through the

lungs, while vapors of lower volatility and many metals notably Hg and As may be eliminated through the urine. However, many other metals such as lead, manganese, and radium are excreted mostly through the intestines. Many of the poisonous elements, such as metals with chemical properties similar to those of calcium (radium, lead) which are deposited in the bones, can be stored for long periods of time in the body. Fluorine is also stored in the bones, while mercury may be stored for long periods in other parts of the body.

"The effect of a toxic material on the body can be acute or chronic. Three types of acute effects which may result from the inhalation of a toxic fume, but particularly from gases and vapors, are asphyxiation, irritation of the respiratory organs and narcosis. In addition, there are other effects which may be acute but which are more typically chronic. These include damage to blood, nervous system, liver, kidneys, bones, lungs, etc. Some pathological changes may not become evident for as long as five years after the last exposure, as has been the case with a number of beryllium poisonings.

"In estimating or evaluating a potential hazard, the following points should be considered:

1. Toxicity of substance handled or present
2. Physical properties of such substances
3. Possibilities of absorption by workers
4. Quantities involved
5. Continuity of exposure
6. Complaints or evidence of injury

"If from the preliminary investigation a possibility of contamination and subsequent injury exists, a thorough investigation of the entire project should be undertaken. People are available to evaluate potentially hazardous conditions or operations wherever they may occur and to test and evaluate atmospheric concentrations of all known toxic materials as well as for analyzing biological specimens such as blood and urine to determine the extent of past exposures.

"Once a hazard is recognized as to causative agents and point of origin, the problem of control is then resolved by applying one of the basic methods of correction - the one, that is, that will control the hazard and will also satisfy those doing the work. They are elimination of the operation causing the hazard, isolation to an area where it will cause no harm, substitution of a less hazardous material, local ventilation, general ventilation, or use of respiratory equipment or a combination of these.

"Table VII is a list of some reactor materials with a few pertinent facts regarding their toxicity."

TABLE VII
TOXICITY OF REACTOR MATERIALS

Element	MAC	Fire Hazard	Pertinent Facts
Aluminum	None	Severe as a finely dispersed powder	
Beryllium	2.0 gm/M ³	Be(NO ₃) ₂	Stringent regulations have been set up to control dissemination of Be and its compounds. Be nitrate may explode in contact with organic matter.
Bismuth	2 mg/M ³		Large amounts can cause kidney damage. Typical heavy metal poison.
Boron			Not considered an industrial poison although some compounds are dangerous if ingested in large amounts.
Cadmium	0.1 mg/M ³		Very toxic as freshly generated fume from welding or other high temperature work.
Graphite	Nuisance		
Hafnium	No information		Believed to be of low order of toxicity.
Lead	0.15 mg/M ³		Cumulative poison affecting blood producing organisms.
Potassium		Reacts violently with H ₂ O	Contact with skin can cause severe burns. Fumes from fire are a severe respiratory irritant.
Sodium		Reacts violently with H ₂ O and will burn in air if heated	Contact with skin can cause severe burns. Fumes from fire are a severe respiratory irritant.
Stainless Steel	30 mg/M ³	None	Steel itself is only a nuisance. Chromium in stainless and fluoride flux on welding rods is, however, hazardous.
Tantalum	None	None	Not hazardous in itself. Necessity of using CCl ₄ as coolant and lubricant when machining, however, does create a hazard.
Thorium	0.5 mg/M ³	Dangerous fire hazard as a powder	Primarily a radiation hazard.
Titanium		Explosive as a dust	Chiefly a nuisance as Ti. However, as TiCl ₄ it is irritating and corrosive and forms HCl in the body.
Uranium	0.05 mg/M ³ soluble compounds 0.25 mg/M ³ insoluble compounds	Burns readily, violently in finely divided form	Causes acute kidney damage.
Zirconium		Fire and explosion hazard	Number of recent explosions of U-Zr alloys after nitric acid washing.

LECTURE 11

PREPARATION OF PURE MATERIALSReferences:

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"Introduction to Reactor Engineering Principles," Samuel Glasstone, to be published.

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Introduction

Because of the high cross section of impurities such as B and Hf, high purity has been a premium in all reactor materials. In general, the special metals used in nuclear reactors are high in the electrochemical series and are difficult to reduce and to purify. High purity has been obtained at the cost of intricate and expensive processes involving non-familiar techniques.

The ores of such materials as U, Th, and Zr, which are generally of very low concentration and include many varieties of impurities, are concentrated by the normal methods of ore concentration, such as gravity separation and flotation. Then the concentrates are purified, in most cases before reduction to the metals. This differs from the normal industrial practice with such metals as Fe in which the purification is carried out by oxidation and slagging of the molten metal. But the procedure is similar to that used in the manufacture of Al in which the ore is purified before reduction. Al also is high in the electrochemical series.

High purification of U ore is carried out by use of liquid-liquid extraction by extracting the U as a nitrate from the impurities present by an organic solvent, such as diethyl ether.

The metals U, Th, Be, and Zr are all reduced from the fluorides or chlorides by Mg or Ca. All four metals are handled in very small batches. In the case of U and Th, the reactions are carried out in bombs; Th is co-reduced with Zn to form a lower melting alloy in order that the slag will separate from the metal.

Work on preparation of pure U by electrolysis has given some promise of success. Similar work on the other metals may eventually permit electrolysis from the fused chlorides or fluorides.

Alloys of U and Zr are desired. Coreduction of the elements to form the alloys directly by solution in the liquid state, and powdered metallurgy have been used. When we look closely at the alloys, we find that their method of preparation is in a very primitive state of development.

The work on the purification of U by metallurgical processes such as liquation, slagging, and zone melting may teach us how to purify the reduced metals more cheaply than we now purify the ores prior to reduction. Rapid changes in technology of these materials is to be expected over the next few years.

U Ores

Natural U occurs very widely in nature and in a great variety of compounds. It is present to the extent of 4 ppm of the earth's surface. Unfortunately, there are only a few known deposits of high grade ores such as pitchblende and these occur in eastern Europe, Belgian Congo, and Canada. The ore is 1 to 4% U. Carnotite, $K_2O \cdot 2UO_3 \cdot V_2O_5 \cdot xH_2O$, occurs widely over the Canadian shield that extends into the Colorado Plateau. Most of these ores contain 0.1 to 0.5% U. Uraninite, UO_2 , and autunite, $CaO \cdot 2UO_3 \cdot P_2O_5 \cdot xH_2O$, occur with carnotite. The deposits near Marysville, Utah, are mainly of these types.

The African gold residues contain about 0.02% U. In addition, the phosphate rocks, leached zones, and oil shale deposits contain about 0.01% U. Monazite sand contains both Th and U, in the proportion of 100 to 3. Some rare earth deposits do not contain Th and U in appreciable amounts while others do contain these elements.

The cost of mining some U ores is relatively expensive because the concentration of the ores requires several steps. Other ores are less expensive to handle. In the case of the South African gold ores, they have already been mined and crushed for their gold, so the U has to stand only a part of the cost.

Pitchblende is normally upgraded to about 50% U_3O_8 at the mine by crushing, screening, washing, flotation, and gravity separation - all normal ore concentration procedures. Low grade ores are further concentrated by leaching with H_2SO_4 and precipitation by MgO of magnesium uranate, known as MgX .

Most of the Colorado plateau ores contain V and the U is concentrated and separated from the V by the acid or carbonate process - the choice of process is made primarily on the basis of the amounts of $CaCO_3$ and $MgCO_3$ in the ore. The flow sheet for the acid process is shown in Figure 1.

The following details of the process are taken from AECD-3241:

"(1) The ore is crushed (dry) to less than 14 mesh, mixed with 8-10% salt ($NaCl$) and roasted in a multiple-hearth furnace. (The roast temperature is 800-850°C.)

"(2) Calcines from the roaster are first leached (percolation) with water which removes 70-75% of the vanadium as water soluble sodium vanadate.

"(3) The water-leached tailings are then leached in place (percolation method) with dilute HCl solution which has been obtained by scrubbing out the HCl produced in the salt-roasting step. This operation removes over 90% of the uranium and also dissolves approximately 15% additional vanadium.

"(4) The water leach is treated for vanadium recovery by precipitation as "red-cake." The acid leach is treated to obtain a concentrated precipitate of uranium and also a precipitate of iron-vanadate which is recycled to the roasters."

The South African ores are separated by adding H_2SO_4 and leaching out the UO_2SO_4 . The acid cost is low since the ores are almost pure quartzite, and the acid is produced from the pyrite in the ore. U is also recovered from phosphate residues. The so-called "leached zone" materials which overlie the Florida phosphate pebble beds contain 0.01 to 0.015% U and methods are being worked out to recover this material.

The world's known reserves of U and Th ores have been studied to determine if they are adequate to support a large scale power industry. Large scale exploration for U began in 1948. Since then many discoveries have been made. For example, the Colorado Plateau has become very important and the production is over \$100,000,000 per year and doubling every 18 months. Canada, in addition to the Great Bear Lake area, has become an important producer. The South African gold mines promise to be one of the largest producers.

Figure 1 - U Separation by Acid Process Carnotite Ore

The known deposits of high grade ores are limited and will be exhausted in a relatively short time. The low grade ores are still being discovered and it is probable that the discoveries will keep ahead of the mining. In fact, it is doubtful if the large future production areas have been located.

More practical and immediate questions are those relating to cost of recovery and the rate of supply of U and Th. If we assume that new ore bodies will be found as rapidly as the known mines are exhausted, there will be a sufficient supply at \$10 per pound or so to support a very large scale power industry over the entire world. An expansion of 20,000,000 electrical kw per year is reasonable in a decade or more. Both the rate and amount of supply of U appear to be adequate.

An expansion of 20,000,000 kw will require 10,000 to 15,000 tons of U and Th as inventory. The amount burned up will depend on whether breeding becomes practical or not. With breeding the U. S. would need the energy from fissioning only about 500 tons of U but without breeding perhaps 5000 tons of U would be required yearly in addition to the inventory requirements.

If we assume \$300 a pound as the top limit for U and Th, there appears to be large amounts of U and Th that could be mined profitably. With complete breeding, the heat value of one pound of U or Th would be equal to some \$8000 worth of coal.

Separation of U

U is separated from the concentrates by solvent extraction in packed columns using diethyl ether or TBP. A flow sheet for this second process is outlined in Figure 2.

The use of diethyl ether as a solvent extraction reagent is described in the literature as early as 1912. This is illustrated by the flowsheet.

Preparation of Metal and Hexafluoride

The uranyl nitrate is heated to 450°C to form UO_3 (mainly). This is reduced to UO_2 by H_2 at 800°C. The oxide is converted to the fluoride by HF at 600°C:

To make metal, the tetrafluoride is reduced with Mg in a refractory lined bomb at 650°C. The reaction takes place at 515°C:

Figure 2 - U Separation from Concentrates by Solvent Extraction in Packed Columns using TBP.

The MgF_2 and impurities act as slag and may be broken off the surface on cooling. The metal is etched with acid to remove surface impurities, vacuum melted in graphite crucibles at 1300°C and poured into ingots for rolling into rods. During the melting various impurities are volatilized and others are concentrated in the top of the ingot which is cropped.

The impurities include nitrides, oxides, and carbides. Since the pours are relatively small, about 300 lbs, the use of liquation and slagging to purify the metal is relatively unimportant.

In order to prepare the hexafluoride, F_2 is passed over the tetrafluoride in trays at 360°C . The trays are tipped a bit and the volatile hexafluoride flows off into liquid CO_2 -cooled traps.

Th

While Th is about 3 times as abundant as U, there are few known deposits containing significant amounts, so the known deposits are less than those of U. On the other hand, the search for Th has not had the interest and pressure that U has had. Much of the readily available ores occur as monazite sands in Brazil and India. These sands are concentrated by ore dressing after which the concentrates contain 0.15 to 0.25% U, 5 to 8% Th, and some 50% of rare earth elements mostly in the form of phosphates.

The monazite sands are processed by alkaline or by acid processes. The acid process in brief is shown in Figure 3.

The U and R.E. are worked up by solution in nitric acid and TBP extraction of the U.

The metal is prepared in a method similar to that used in preparing U. The oxalate is heated to 650°C in air:

Then HF is passed over the ThO_2 at 550°C :

B and Si are volatilized, if present.

The ground fluoride is mixed with metallic Ca and 10 to 12% ZnCl_2 . The mixture is charged into a refractory lined bomb and heated to 670°C :

Figure 3 - Acid Process for Recovery of Th from Monazite Sands

The ZnCl_2 provides additional heat which is necessary to liquify the Th, alloy with the Th to lower the melting point, and allow separation of the slag.

After cooling, the biscuit is cleaned and the Zn distilled off. The Th is vacuum melted in a BeO crucible and poured in graphite molds at 1800°C .

Graphite

Graphite is prepared by commercial methods except that care is given to selection of raw materials. Petroleum coke is ground, mixed with pitch, gas baked, reground, etc., and given a second gas bake. It is then placed in an electric furnace and heated for several weeks at 2700 to 3000°C . The impurities are largely vaporized and the purest material is in the center of the furnace.

Be

Be is processed from beryl, $3\text{BeO}\cdot\text{Al}_2\text{O}_3\cdot 6\text{SiO}_2$. Beryl is found widely, but most comes from Brazil and South Africa. The demand is such that "high grading" mining methods only have been followed.

The metal is prepared by the flow sheet in Figure 4.

Zr

Zr is also difficult to win from its ores. Since Hf occurs with it and has the same chemical properties, separation must be carried out. The process is shown in Figure 5.

D_2O

Selak and Finke present the only summary of D_2O production in the U.S. and Canada available. Their list of the properties of D_2O is presented in Table I.

During the war, distillation and electrolytic separation plants were built. In addition, development work was carried out on exchange systems.

The distillation of water is straightforward. The difficulty with the process is the small difference in volatility between H_2O and D_2O . Very large vacuum columns and a very large number of stages with high steam consumptions are required.

Figure 4 - Preparation of Beryllium

Figure 5 - Zirconium Preparation

Table I

PROPERTIES OF D₂O

Density, maximum	1.106 at 11.2°C
Melting point	3.81°C
Boiling point	101.43°C
Relative viscosity to H ₂ O	1.25 at 20°C
Per Cent Abundance in Normal Water	0.0146% (1 part in 6,850)

The electrolysis was carried out by electrolyzing a K₂CO₃ solution. It was used essentially in the finishing units.

In the exchange process, the reactions:

are used with the reaction being carried out over a platinum catalyst. The equilibrium constant

$$k = \frac{(\text{HDO})(\text{H}_2)}{(\text{H}_2\text{O})(\text{HD})}$$

varies from 3 at 20°C to 2 at 100°C. If an H₂ and HD gas mixture with excess H₂O vapor is passed over a catalyst, HDO is formed and this may be scrubbed from the vapor phase by water - thus enriching the water. This process was incorporated into the first section of the Trail Plant for production of 2.3% D₂O water. This water is then fully enriched by electrolysis.

Properties of the Pure Materials

We have noted in a previous lecture the radioactivity, toxicity, and pyrophoric nature of some of the reactor materials. In general, we may say that reactor materials are far from ideal in their chemical, physical, and mechanical properties, in spite of their high cost, but we must use them because of their nuclear properties.

a. U: U melts at 1125°C and has a low vapor pressure so that it can be melted in vacuum. It forms three allotropic forms:

α : room to 662°C Orthorhombic
 β : 662 to 772°C Tetragonal
 γ : 772 - 1125°C Cubic

with a density at room temperature of 19.05.

The alpha phase is anisotropic and the coefficients of linear expansion in the principal crystallographic directions vary as indicated below:

<u>Direction</u>	<u>Coef. of exp./°C; 250 to 300°C</u>
a (100)	$(23 \pm 3) \times 10^{-6}$
b (010)	$(-3.5 \pm 2) \times 10^{-6}$
c (001)	$(17 \pm 2) \times 10^{-6}$

This anisotropy of the alpha phase causes extreme difficulties in use of U as a reactor material. In rolling or extruding U, particularly in the gamma phase and to a lesser extent in the alpha and beta phases, the crystals are highly oriented, the 001 plane being approximately parallel to the rod axis after working. The position of the other planes depends on the rolling temperature. As a net result the metal is subject to dimensional changes due to temperature changes. Repeated heating and cooling over the range of 50 to 500°C will result in large increases in length. In a reactor such fuel will undergo pimpling, growth, and warpage.

Orientation of the metal may be removed by heating into the beta range and quenching. This effectively randomizes and refines the crystals so that dimensional changes in thermal cycling are reduced.

If a bar is repeatedly heated into the beta range and quenched, it will crack in the center and finally will become hollow. This may be explained by the plastic flow of the outside during heating and the retention of this plastic flow due to quenching and subsequent plastic flow of the core of the bar before it cools.

U oxidizes slowly in air at room temperatures. The oxide is adherent and tends to protect the U. In boiling water it forms a resistant oxide but at higher temperatures the U reacts rapidly with water. When canned and used as a reactor fuel, water reacts with the U to form oxides and hydrides which may burst the container due to decrease in density.

Beta heat treated U has a yield strength of 25,000 to 40,000 psi and an ultimate strength up to 80,000 or more psi. It also has sufficient ductility for plastic deformation to correct for slight changes in dimensions necessary to relieve excessive temperature stresses.

U is processed by rolling and finish machining for use in reactors.

b. Th: Th is a respectable metal with a face centered cubic crystal structure. The melting point is high, 1690°C.

It can be rolled and handled in a normal fashion. It oxidizes readily in air at rolling temperature. Its rate of oxidation is about 0.1 that of U in water. It has a yield strength of 20,000 to 50,000 psi with high reductions in area of 70% or so. Even after cold working, the reduction in area is 30% or more.

c. Zr: Zr is of interest because of its high melting point, corrosion resistance, and low absorption of thermal neutrons. It exists as a hexagonal close-packed lattice metal in the alpha range up to 863°C and as a body-centered cubic lattice up to its melting point of 1845°C.

Zr can be worked readily. The oxide is soluble in the metal so welding is readily carried out. It bonds to U readily. Its yield strength is low - some 15,000 psi - but this is increased greatly by alloying. The metal is ductile with an elongation of 40%.

d. Be: Be has a melting point of 1315°C, yield strength in the range of 17,000 to 30,000 psi for hot pressed powders, and elongations of 10 to 20%. Extruded material has yield strengths of 25,000 to 80,000 psi, but with low ductility and elongations under 5%.

Physical and Chemical Properties

The Reactor Handbook and Liquid-Metals Handbook give considerable data on the physical and chemical properties of reactor materials. Neither of these books give the sources of the data so that, frequently, one has difficulty in evaluating the data. In most cases the data are believed reasonably accurate although the probable errors are hard to estimate. In the heat transfer data, it is not believed that they can be relied upon for a greater accuracy than $\pm 15\%$ and in the case of alloys, etc., an accuracy of $\pm 30\%$ only can be expected.

Argonne has set up recently equipment for measurement of thermal conductivities of irradiated metals. No data are available to date, but many measurements of changes in electrical conductivity have been made.

A few physical properties of some materials are included in Table II. The thermal conductivity of NaK eutectic is about $1/3$ that of Na.

Table II

PHYSICAL CONSTANTS OF SOLID REACTOR MATERIALS

Quantity	Units	Al	Be	Zr	U	Th	Vapor Pres.	
							Na	
							°C	mm
Cross sec., thermal	barns/atom	0.215 ± 0.008	0.0090 ± 0.0005	0.18 ± 0.02	--	--		
Density, room temp.	gm/cm ³	2.70	1.79 - 1.86	6.50	19.0	11.6	440	1
Melting pt.	°C	660.2	1315	1845	1133	1690	548	10
Boiling pt.	°C	2327	2970	--	3900	>3000	696	100
Vapor pres.	atm °C	10 ⁻⁶ 1017		--			752	200
	atm °C	10 ⁻⁴ 1272					815	400
	1mm °C	1537					883	760
	100mm °C	2081						
Sp. heat	0°C	0.208	0.42	0.069	0.027(27°C)			
	100°C	0.225	0.51	0.074(127)	0.032(227°C)	0.028		
	300°C	0.248		0.081(425)	0.038(427°C)	0.028		
	600°C	0.277	0.69	0.086(727)	0.045(627°C)			
	800°C		0.74					
Thermal conductivity, k, BTU/ft ² hr.°F/ft								
	Room	122	87	11.3(75°F)	14.5			
	100°C	122	82	11.2(200°F)	15.3	21.8		
	200°C	128	78	11.0(300°F)	17.2			
	400°C	132	68	10.8(400°F)	18.9			
	600°C		58	10.7(500°F)		26.2(650°C)		
Coef. linear exp.	20 to 100°C	23.8 x 10 ⁻⁶	11.54 x 10 ⁻⁶	*	**	11.5 x 10 ⁻⁶		

*C axis 6.15 x 10⁻⁶; a axis 5.69 x 10⁻⁶**(100) 23.3 x 10⁻⁶; (010) 3.5 x 10⁻⁶; (001) 17.0 x 10⁻⁶

LECTURE 12

PREPARATION OF ALLOYS - PHASE DIAGRAMSReference:

"Metallurgy of Uranium," H. A. Saller, F. A. Rough, Am. Inst. Chem. Eng. Mon., 11:63 (1954)

Introduction

Alloys are of interest to the reactor engineer since the properties of the alloys may be better in certain respects than those of pure metals. Pure metals are usually soft, ductile, and rather low in strength. Alloys can be varied because in many alloys compounds are present. Metallic compounds are usually hard, and have very low ductility and high strength. Alloys permit the use of the entire range of properties between these two extremes.

Many alloys have solid solutions with properties similar to the pure metals. When a compound in an alloy forms a solid solution at high temperatures, but the compound is less soluble at lower temperatures, the compound tends to precipitate out. Rapid cooling may prevent complete precipitation. This results in a very hard, high strength material. This process is used in hardening 24S Al alloys and in hardening steel.

Metals are frequently soluble in the molten state and not in the solid state. This results in lowering the melting points with the formation of mixed crystals known as eutectics. Some metals are not soluble in the molten or solid state.

The reactor engineer is interested in alloys for various reasons. Some of these are listed below:

a. Corrosion Resistance - While it is unsafe to generalize about alloys in respect to corrosion resistance, it is frequently true that solid solutions and pure elements are more corrosion resistant than systems containing compounds. For example, ordinary low carbon steel has less corrosion resistance than pure iron, but high-alloy steels are generally more corrosion resistant than pure iron.

b. Diluting Agents - Enriched fuels must be diluted with either source or non-absorbing materials to increase their volume and give properties similar to the cladding used with them.

c. Reduce Phase Transformation Points - Alloys are occasionally used to reduce transformation points. For example, in steel, high Ni permits the holding of the solid solution of γ iron at room temperature with the attendant high corrosion resistance of the solid solution.

d. Reduce Melting Points - Alloy additions to U have been tried to give alloys with sufficiently low melting points that the solutions can be pumped through reactor fuel elements.

It should be noted that the development of special alloys for use in nuclear reactors is in a very elementary state since the major effort within the Commission has been on the development of pure materials. This has been necessary since it was essential to have some materials for use even if they were not the best. The developments to date should be considered the first step in the development of adequate metals for reactor use with the expectation that much improved materials will become available in the future.

The alloys of interest developed to date may be grouped as follows:

a. Highly Enriched Fissionable Materials

Al-U
Zr-U
Pb-Bi-U
Fe-U
Cr-U
Al-Pu

These alloys are of interest for several reasons. The Al-U, Al-Pu, and Zr-U alloys are useful in fully enriched research and power reactors. The Al and Zr are primarily fuel diluents or extenders and permit the design of fuel elements with sufficient area to permit cooling at reasonable rates. Also, the alloy as used is dilute and has properties close to Al and Zr.

The Fe-U and Cr-U alloys could be used, because of their lower melting points, in place of U in fast reactors. Unfortunately, the depression in melting point of U is not high. For example, U-Cr has a eutectic at 20 a/o or 5 w/o Cr of about 840°C.

The Pb-Bi-U alloys are useful in liquid metal-fueled reactors. It is unfortunate that the melting point of these alloys is so high for reasonable solubility of U.

b. Natural or Slightly Enriched U Alloys

U-Zr (2%)
U-Nb (3-6%)
U-Si ($2\frac{1}{2}\%$)
U-Nb-Zr ($1\frac{1}{2}$ - 5%)
U-Mo

The alloys in this group have been studied primarily to obtain an U alloy with increased water corrosion resistance. U, in high temperature water, corrodes very rapidly. In a power reactor, a defective fuel element would be completely oxidized in a short time with the fission products, U, and Pu present distributed rapidly throughout the cooling system. When properly alloyed and heat treated, the U-Nb, U-Nb-Zr, U-Si, and U-Mo alloys show remarkably improved water corrosion resistance.

c. Coolants

NaK
Pb-Bi-U

Phase Diagrams

The metallurgist and physical chemist use phase diagrams to illustrate the changes that take place in metals and oxides when they are mixed. Many metals are soluble in one another in the molten state, but many of these are less soluble in the solid state. This also applies to oxide systems and to salt systems. Compounds of metals form at low temperatures, but may break up at higher temperatures. Further, when materials are soluble in the molten state and less soluble in the solid state, they tend to lower the melting point.

Usually phase diagrams are developed from cooling rate curves of different alloys combined with metallographic examination of the alloys.

Typical phase diagrams are presented in Figures 1 and 2.

Preparation of Alloys

Alloys may be prepared by a number of different means. These include:

- a. Additions to the melt
- b. Coreduction
- c. Powder metallurgy
- d. Diffusion

Figure 1 - U-Al Alloys

Figure 2 - U-Zr Constitutional Diagram

To date relatively little has been done on coreduction of the oxides or fluorides in respect to project materials. Monel metal is made by coreduction of Ni and Cu. Diffusion is not used to any extent today. However, all steel was made, until the 1850's, by diffusion of C into wrought iron.

Alloys, then, are most commonly made by dissolving the alloying elements together in the molten state or by mixing the various powders before pressing in powder metallurgy. In making alloys by mixing the melt, it is frequently only necessary to throw the alloying element in and depend on thermal mixing. This is usually done in steel practice as the alloys are simply shovelled into the open hearth or electric furnace. U may be alloyed with Zr and Nb by addition of the wire to the molten U while the U is held in an induction furnace.

In powder metallurgy, the ground powders of <40 micron size are mixed and pressed hot. Diffusion tends to produce relatively uniform materials.

LECTURE 13

PREPARATION OF FUEL ELEMENTSReferences:

- "Melting and Fabrication of Zirconium," J. W. Holladay, J. G. Kura, Amer. Inst. Chem. Eng. Mon., 11: 23 (1954)
- "Powder Metallurgy of Zirconium and Beryllium," H. H. Hausner, H. B. Michaelson, Ibid., 11: 11 (1954)
- "Preparation, Properties, and Uses of Beryllium," E. J. Boyle, J. L. Gregg, Ibid., 11: 53 (1954)
- "Thorium Metallurgy," J. R. Keeler, Ibid., 11: 57 (1954)
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- "Electrolytic Preparation of Uranium from a Fluoride Bath," Ibid., 12: 63 (1954)

Introduction

The development of fuel elements for heterogeneous reactors and of fuel solutions for homogeneous reactors has been and continues to be one of the most important problems in reactor development. This is so since the fuel element is the most important part of the heat removal system. Temperatures of operation are limited by the corrosion and stresses which the fuel element can withstand.

Fuel element design and development has occupied a considerable effort during the life of the nuclear project. While efforts to date have been fruitful, we still do not know how to build completely satisfactory fuel elements for most reactors. In a way this is to be expected, since the design requirements for fuel elements are raised as rapidly as they can be met.

Requirements for Fuel Elements

The requirements for fuel elements may be broken up into three groups depending on the use or type of reactor. These groups are the heterogeneous, homogeneous, and liquid fuel types. The liquid fuel reactor fuels may be subdivided into those which act as fuel only and those which act as fuel and coolant combined.

1. Heterogeneous Reactors

a. Subdivision

In respect to subdivision there are two opposing requirements for such fuels as natural or slightly enriched U and Th. First, it is desirable that the fuel be subdivided to give good thermal contact with the coolant. This requires high surface area and small fuel elements. On the other hand, both U and Th have high resonance capture and the fuel should be clumped in order to obtain high resonance escape probability. A higher degree of subdivision of U and Th can be used to achieve high specific power if additional enrichment is added to maintain criticality. The increased resonance capture in the U^{238} tends to offset the loss in thermal capture in U^{238} , thus keeping the conversion ratio about the same. This is approximately true for Th, also, as the resonance capture is somewhat less than for U^{238} .

For natural U reactors such as Hanford, clumping is carried out by use of single large rods (rows of slugs) or several rods grouped in a close assembly. CP-2 was actually built with clumps of fuel, but the use of rods does not decrease the resonance escape probability too much. The use of seven or nineteen rods in a group permits clumping with large heat transfer area. For the high heat rates needed for production of low cost power, the maximum rod size is not over about 1/4 in. diam. Plates will probably be much used in future heterogeneous reactors.

For enriched fuels the maximum subdivision needed from a heat transfer viewpoint is used. However, too much subdivision may increase the fuel cladding materials to an excessive amount.

b. Stability

The fuel and source materials must have sufficient stability to withstand the thermal stresses and not undergo undue warpage or growth as a result of the temperature changes and irradiation damage. Irradiation damage is caused by the slowing down of neutrons and fission fragments. In general, the fuel should not undergo phase changes.

U undergoes a phase change at 660°C. Ordinary rolling of U in the α range produces rods that have crystal orientation and will grow under thermal cycling in a longitudinal direction.

c. Corrosion Resistance

The corrosion of the fuel element cladding material usually is the limiting factor on temperature of operation of heterogeneous reactors. The cladding materials must protect the fuels from interaction with the coolant or the materials must be chosen so that the rate of reaction is low.

Both U and Th react rapidly with high temperature water. U forms oxides with water and, with a deficiency of water, hydrides. The oxides and hydrides have low density and, once a lead through the cladding occurs, they tend to break the fuel element cladding and release the oxide and hydride powders to the coolant system. Corrosion resistant alloys are being developed in an attempt to eliminate the rapid corrosion of U and permit continued operation in case of a clad failure until the next convenient shutdown. As noted elsewhere, some of these corrosion resistant U alloys lose part of their corrosion resistance due to irradiation.

d. Fission Fragments

Fission fragments travel the order of 0.1 mm. The clad must be thick enough to stop them, else they will make the coolant radioactive. Actually, this is no problem as all clad materials are thicker than this.

e. Size

The fuel elements should be of such size that they are readily handled and loaded and unloaded in reasonable time. The tendency is toward larger and larger assemblies that will provide the minimum number of assemblies to be handled.

f. Manufacture

The cost of manufacture of fuel elements has been high. In general, fuel elements used to date have been complex and many steps involved in their manufacture. Simplification must be accomplished if we are to have an economic power industry.

g. Recovery

The fuel element must be designed so that the U and Pu may be readily and cheaply recovered. Here again, costs to date have been excessive, although work to reduce the costs is underway at several laboratories.

2. Homogeneous Reactors

a. Subdivision

The fuel must form a stable emulsion or solution with the coolant. This means that the clumping of the fuel to increase the resonance escape probability is impossible, and either enrichment utilized or a larger reactor used to offset the loss in reactivity.

The use of a two-core reactor may effectively accomplish clumping. In this case the reactor fuel is enriched and the target material is placed outside of and separated from the active core by a Zr or s.s. wall. Two-core reactors, as they are called, have been designed, but none have been built to date.

b. Stability

The fuel solution must not form a second phase under the conditions of operation or a slurry must not settle at any point in the system. So far, only solutions have been used. Work with slurries or emulsions has not been too satisfactory.

$\text{UO}_2\text{SO}_4\text{-H}_2\text{O}$ solutions normally form two phases at temperatures above 285°C .

c. Corrosion Resistance

The corrosion of homogeneous reactor systems occurs mainly in the external parts rather than in the reactor itself. The problem is simplified to the extent that high cross section materials may be used, but the solutions are generally more corrosive than pure water or Na.

d. Recovery

The great advantage of homogeneous fuels is their ease of recovery; xenon and krypton can be removed as fast as formed - thus the xenon does not have to be burned out. Nor do the materials, in general, have to be dissolved, and they never have to be refabricated. Exchange reactions have indicated such promise that decontamination by removal of fission products may prove to be very cheap.

e. Decomposition

The fuel must not be such that it decomposes in the reactor producing precipitates of fissionable materials.

3. Liquid Fuel Reactors

In liquid fuel reactors, the moderator is fixed and separated from the fuel coolant. The requirements for the liquid fuel are about the same as for the homogeneous reactor.

Classification

We might classify fuel elements as follows:

a. Natural U or Slightly Enriched U

Operating Reactors

Hanford, X-10, Brookhaven, Chalk River, NRX, most European reactors.

Proposed Reactors

Monsanto, Commonwealth, NRU, nearly all overseas reactors.

b. Highly Enriched U, Highly Diluted

Operating Reactors

MTR, LITR, BSF, STR, CP-5

Proposed Reactors

Some aircraft, most research reactors, SIR

c. Breeder Fuels

Operating Reactors

EBR (Enriched U^{235} , natural U blanket)

Proposed Reactors

Detroit Edison

d. Homogeneous

Operating Reactors

HRE (Enriched U^{235})

Proposed Reactors

Aircraft, Power

Design of Fuel Elements

Fuel elements then must consist of a core of fissionable, and usually source materials or diluents, surrounded by an inert corrosion resistant clad. The cladding material serves to stop fission fragments, to eliminate corrosion of the fuel, to restrain the fuel to a certain extent from warping, and to cut down the danger of toxic materials getting into the coolants.

The fuel element should be bonded to the cladding material in order to give the maximum rate of heat transfer. The "bond" may be liquid or solid.

The fuel assembly should be designed to have an adequate life. The length of time a fuel assembly remains in a reactor will depend on the flux and permissible burnout. The permissible burnout is a function of the conversion ratio in power reactors.

Fuel elements may be made with high Zr or Al-enriched U, Th-U²³³ or U²³⁵, natural or slightly enriched U alloys. The percent of enrichment is based on a comparative cost analysis of enrichment versus coolant and other costs in the case of thermal reactors.

Fuel elements may be built in any suitable physical form and the metal, oxide, or other form may be used. Actually most fuel element designs, to date, have been based on using metallic fuel.

Enriched U Fuel Elements

Al-U fuel elements have been made by the following operations:

The U in the form of the oxide, UO₃, is reduced with excess Al in a crucible with cryolite as the fluxing agent. Reduction of the UO₃ and alloying of the U with the excess Al occurs at about 850°C. The melt is poured into a graphite crucible. After cleaning, the ingot is rolled nearly to proper thickness of the meat or alloy in the fuel element; the sheets are picture framed by cutting out a portion in a die; the sheets are cleaned; and the fuel plates are made up by rolling hot with the Al-U alloy surrounded by a punched side frame and top and bottom plates. From 10 to 18 of these plates are brazed to Al side plates. End fixtures are riveted to the extended side plates. Such fuel elements are used in the LITR, Swimming Pool Reactors, MTR, CP-5, etc. The alloy consists of 15 to 18% U by weight. Al-U alloy rods have been made up for use in CP-3' and other reactors. In this case the alloy is simply rolled to shape, cut to length, and canned in Al cans without bonding.

LECTURE 14

CORROSION OF REACTOR MATERIALSReferences:

"Reactor Handbook," A. H. Roebuck, Vol. II, p. 213

"Aqueous Solution of 2S-Al at Elevated Temperatures," J. E. Draley, W. E. Ruther, ANL-5001

"Corrosion of Zirconium in High Temperature Water," D. E. Thomas, Amer. Inst. Chem. Eng. Mon., 12: 16 (1954)

"Seventeenth Semiannual Report of the Atomic Energy Commission," Gov. Printing Off., p. 37, (1955)

"Corrosion and Impurities in the Heavy-Water System of the Dutch-Norwegian Reactor," T. J. Barendregt, K. H. Brakstad, Amer. Inst. Chem. Eng. Mon., 11: 183 (1954)

Reactor materials may be divided into source and fissionable, moderator, and structural (including cladding) materials as far as their resistance to corrosion is concerned. The materials of interest in these three groups are: (1) U, Th; (2) Be, BeO, C; and (3) Mg, Al, Zr, s.s. Ta and Ti may be considered for structural materials in the future. The alloys of U, Th, and Zr are particularly important.

Corrosion may be defined for our purposes as the transfer of a component of a solid to a fluid or as the transfer of a component of the fluid to the solid. Corrosion engineers always use the term corrosion to refer to the transformation of a metal from the free to the combined state such as an oxide or a sulfide.

The types of corrosion we are most interested in are those which result in oxidation by water and dissolution or reaction with liquid Na. In graphite moderated reactors, reaction of C with oxygen or gaseous oxides is of importance, but this can hardly be called corrosion.

Most of the materials which are used in reactor technology are very reactive, i.e., they have large $-\Delta F^\circ$ values when they are oxidized. This means that there is always a thermodynamic force towards oxidation and replacement of hydrogen in water, and the materials are used in non-equilibrium states. Unless the oxidation reactions are slowed by

mechanical means, Al, Zr, Ta, Ti, Be, U, and Th would all react with water or oxygen from the air quite rapidly. A list of heats of formation is presented in Lecture 27, Table V.

If the oxide layer has a volume greater than the metal it replaces, there will be tendency to form a continuous film which is under compression; if $M/D < \frac{m}{d}$ or $\frac{Md}{mD} < 1.0$, a discontinuous film may be formed.

M = mass of oxide

D = density of oxide

m = mass of metal in M oxide

d = density of metal

This ratio has values in the cases of Na = 0.57, K = 0.41, Mg = 0.79, and Ca = 0.64; so these metals do not form continuous films and tend to completely oxidize once rapid reaction starts. A deep oxidized film on Mg gives nominal protection at room temperatures, but a break in this film results in rapid corrosion. Other metals have values over 1.0 for this ratio and tend to form compressive films. For example, Al forms $\text{Al}_2\text{O}_3 \cdot 2\text{H}_2\text{O}$ at 90°C and $\alpha\text{-Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$ at 130°C . These compounds would be expected to occupy much more volume than the Al in the films. The orientation of the molecules and the actual crystal lattice would be factors involved in the formation of adherent continuous films under compression. To form such a film on Mg will presumably require the alloying of Mg with other metals that will increase the volume of the oxide film.

The fact that Al forms a film in compression may be demonstrated by dissolving the Al away from the oxide and noting the wrinkles formed in the oxide. Fluctuating temperatures and mechanical shock may injure the film and increase corrosion.

Iron has two hydrated oxides, $\text{Fe}(\text{OH})_2$ and $\text{Fe}_2\text{O}_3 \cdot \text{H}_2\text{O}$ or $\text{FeO}(\text{OH})$. These have different solubilities and this fact is believed to be effective in changing the film so that it is not stable. The $\text{Fe}(\text{OH})_2$ is much more soluble than $\text{FeO}(\text{OH})$, so it is moved outwards. With a deficiency of O_2 , Fe_3O_4 or the hydrated form may be precipitated. It is the precipitated Fe_3O_4 that causes the trouble in s.s. -Zr loop tests by precipitating on heated surfaces.

Most corrosion involves localized electrical currents for the transfer of the ionic charges. In an iron-iron oxide system, a current will flow with the oxide as cathode and the metal as anode. At the cathode, oxygen will react.

At the anode, the reaction $\text{Fe} = \text{Fe}^{++} + 2e$ will take place. The result is the precipitation of $\text{Fe}(\text{OH})_2$ followed by its oxidation in oxygen-containing water. The iron goes into solution at one place, the oxygen is taken up at a second point, and the hydroxide or oxide formed at a third point; thus, there is very little tendency to form a continuous film.

In high purity water, the oxidation is inhibited by the ability of the water to conduct the current.

Frequently, the oxide film formed at first is a metastable oxide and after a time the stable form crystallizes out. Once this stable form is present, it catalyzes the transformation from the metastable to the stable form with the result that the corrosion rate is changed from that of the formation of the metastable to the stable form. In the case of Zr, the time at which the stable form begins is measured in months and in stainless steel in years. The initial formation of the stable form is known as the "breakaway" point. The rate of corrosion of Zr after the breakaway may be similar to that for Al.

Stresses and strains set up in rolling tubes in boilers, welding, etc., may cause increased corrosion rates. Intergranular attack or blister corrosion may occur in Al at high temperatures.

Cavitation may cause erosion-corrosion by mechanically breaking up the oxide films. In this case, corrosion follows the cavitation at a rapid rate.

To be meaningful, corrosion tests have to be conducted with great care and the conditions of use have to be simulated in detail. Corrosion usually results in the build-up of films. Either the thickness of the film has to be measured (and an estimate made of its density) or the film has to be removed in order to obtain true corrosion rates. A slow build-up of corrosion products as indicated by small weight changes and ion pickup in the purification system will indicate low corrosion rates.

The effects of irradiation on corrosion have been studied extensively. All that can be said is that the effect must be low in most cases.

While not a part of corrosion in the strict sense of the word, the solution of metals by liquid metal coolants is of extreme importance to the reactor engineer. In general, Na does not attack Zr or the steels. Attack of Al and Mg is slight in the absence of O_2 . Solubility is small up to fairly high temperatures. They form two liquid layers.

Source and Fissionable Materials

U and Th react rapidly with air at elevated temperatures.

Zr, Nb, Si, and Mo form alloys with U which are quite corrosion resistant. In some cases heat treatment is necessary to give the best corrosion resistant properties.

Very little work has been done on the preparation of stainless Th alloys, but such work is needed and will be inaugurated. Be and C seem to increase the corrosion resistance of Th while Cr and Zr seem to be harmful. Both of these elements are insoluble in Th. Ni is soluble and might increase corrosion resistance.

Moderator Materials

Be has been used in the pure state to date. Little, if any, work on development of special alloys has been carried out. There have been considerable differences in corrosion rates depending on the history of the material and the impurities present. In general, extruded or pressed metal has been most satisfactory. Early metal contained more impurities and showed correspondingly higher corrosion rates.

Pressed and sintered Be at 525°F lost 1.0 to 144.5 mg/dm²/mo in distilled water.

NaK attacks Be slowly at temperatures above 800°F with erosion rates of 1 mil or so/mo. Na attack at 930°F, 10 ft/sec, is 0.3 to 0.4 mils/mo. Attack by Bi below 1300°F is slow.

BeO begins to react with H₂O vapor above 1250°C forming a volatile compound. On cooling, the compound decomposes and the BeO condenses. C reduces BeO at high temperatures. Fused alkalies and alkali carbonates readily dissolve BeO.

C reacts with O₂ at temperatures above 375°C but the rate is very slow up to 425°C or above. Reaction with Na is being studied.

Structural and Cladding Materials

In heterogeneous reactors corrosion must be limited to small amounts because of the danger of deposition of the corrosion products on the heat transfer surfaces, in dead spaces, and on moving shafts. In addition, corrosion products may cause the coolant to become excessively radioactive with isotopes that will not decay rapidly, thus presenting additional difficulties in maintenance of the reactor system.

The coolants of most interest in heterogeneous reactors are water and Na; use of Mg is being studied. In general, water is a very active material, and its corrosive rates are dependent to some extent on the dissolved gasses such as O_2 and H_2 present and on the pH and other impurities.

Soluble salts, particularly Cl^- and Cu^{++} , are particularly harmful. In deionized water these salts are always too low to be harmful.

Na, while a very active material in respect to reaction with O_2 and H_2O , does not react with other materials at a rapid rate.

Mg

Mg reacts with water rapidly. While it has a favorable cross section, it has not been used as a structural material because of its high water corrosion rate as indicated below:

<u>Temp. °F</u>	<u>Mils/month</u>
95	0
150	0.27
180	0.75
212	2.9

Mg is subject to galvanic attack with severe pitting when coupled to other metals. Attack is uniform in dynamic systems but erosion-corrosion becomes serious above 25 ft/sec. Relatively little has been done on development of special Mg alloys to withstand high purity water. Work on this project is being started at Argonne.

Al

Al corrodes rather rapidly with severe pitting when utilized in impure water. However, in high purity water the corrosion rate is low due to the formation of an adherent film. In neutral or slightly acid water acceptable corrosion rates occur up to 200 to 250°C. In the neighborhood of 250°C blister type corrosion sets in and becomes severe at 275°C. In fact, at 300°C or so the samples completely disintegrate into a powder in a few days exposure.

Initial corrosion rates, usually indicated by weight increases, are high, but the rate of weight change drops off rapidly during the first few days and becomes constant at low rates after about ten days.

Corrosion rates average 1 mil per mo or less even up to nearly 300°C, but, as noted, blister corrosion limits use above 230°C. As would be expected, corrosion rates become high at high or very low pH. However, the resistance to acid increases with temperature and optimum corrosion resistance occurs at low pH at high temperatures. The optimum pH is 6.5 at 50°C and 2 at 315°C. Static test corrosion data at 200°C are:

pH 7	1.4 mils/yr
pH 5	0.7
pH 3.5	0.3

Recently several AlNi samples prepared by Dr. Joseph Draley have given remarkable corrosion resistance. The addition of certain inorganic salts to the test water protected 2S-Al. In addition, AlNi alloys and Ni plated Al test specimens run for 1000 hr or longer at 316°C have developed the blister corrosion very slowly. Samples not properly protected have completely oxidized under the conditions of test, see Figure 1. The work indicates that Al alloys may be developed that may be used in reactors for extended periods at temperatures over 250°C. At present the upper limit with reasonable corrosion rates is about 230°C.

No pitting has occurred in any high temperature Al corrosion using highly deionized water in the Argonne tests. On the other hand, Barendregt and Brakstad report "mostly great pittings" in 212°C autoclave tests for 100 hr using 2S-Al. At lower temperatures in the Argonne tests, pitting occurs at low water velocities and no pitting at water velocities of 10 ft/sec or over has been noted. Pitting appears to be associated with localized changes in pH or other ions in the water.

Zr

The corrosion resistance of unalloyed Zr in high temperature high purity water depends mainly on the trace elements present. Alloying is believed to depress or eliminate the harmful effects of the trace elements without contributing to improving the corrosion resistance of absolutely pure Zr. Nevertheless, the alloys are a very important improvement.

In oxygenated water, Zr can be used to 316°C.

Corrosion of Zr in water results in a dark adherent film. However, after several months in high temperature water, the corrosion rate may rapidly increase with the formation of a white non-resistant film. This is called "breakaway" corrosion. The rate becomes much higher.

Thomas presents an excellent discussion of Zr corrosion and presents numerous test results.

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Figure 1 - Ni Plated Al Tested 439 hr at 316°C
Degassed, Deionized Water 20 ft/sec

LECTURE 15

RADIATION DAMAGEReferences:

"The Effect of Irradiation on Mechanical and Physical Properties of Structural Materials with Particular Reference to the Mark I Naval Reactor," D. O. Leeser, ANL-4792.

"Radiation and Thermal Effects," S. H. Paine, Reactor Handbook, Vol. III.

"Radiation Damage to Nonmetallic Materials," V. P. Calkins, Amer. Inst. Chem. Eng. Mon., 12: 28 (1954).

"Radiation Effects on Structural Materials," C. R. Sutton, D. O. Leeser, Ibid. . 12: 208 (1954).

Theory

While radiation damage can be produced by beta particles and gamma rays, damage in reactors is mainly produced by fast neutrons and fission fragments. The beta and gamma rays cause electronic excitation and ionization while fission fragments and neutrons displace large numbers of atoms. The fission products or nuclei also cause ionization.

There are four types of disturbances in the atomic lattice that affect structural materials: vacancies, interstitial atoms, thermal spikes, and impurity atoms. Fission fragment damage is limited to the fissionable material and immediate adjoining clad materials.

Vacancy

Whenever an atom is knocked out of its normal site in its equilibrium position in a solid and moved to a new position, it leaves a vacant position in the lattice.

Interstitial

An atom that has been knocked out of its normal lattice position comes to rest as an interstitial atom unless it happens to enter a vacancy in the lattice. Since an interstitial atom occupies a non-equilibrium site, it will cause strains in the neighboring atomic bonds.

Thermal Spike

An atom knocked out of its normal position may travel many atomic distances before it stops. This atom will interact with the atoms near its trail as discussed below. Many of these will be knocked out of their lattice positions and many will be ionized. The net result will be that a large amount of energy is dissipated in the lattice vibrations of the solid. There will be a momentary high temperature with a temperature of 1000 to 1500°K for a volume of 5000 to 10,000 atoms for 10^{-10} second. Very localized volumes consisting of a maximum of several hundred atoms may be heated to several thousand degrees. While the energy of the atoms is extremely high, at least high enough for atomic diffusion to take place, it is probable that the gas is so highly compressed that the atoms essentially continue to occupy their normal lattice positions. Cooling takes place so rapidly that the atoms do not rearrange or diffuse to any extent. In fact, the temperature would tend to randomize the material. The rapid rate of cooling may in itself produce lattice strains or imperfections. In a metal, such as Cu, in which diffusion can take place at room temperature, we would expect rapid healing of radiation damage and no permanent effects. This is actually the case.

The actual temperature in a thermal spike is meaningless since atoms displaced from their normal lattice positions must be considered to have been at energy levels equivalent to the gaseous state. Many other atoms must have had energy levels equivalent to the liquid state. The energy is distributed to the surrounding atoms so rapidly, probably during a few vibrations, that very little diffusion to equilibrium lattice positions is possible. However, each section of a fuel element must be heated as a portion of a thermal spike many times by the time one atom per hundred has fissioned.

Impurity Atoms

The (n, γ) and fission reactions will result in many impurity atoms in the lattice. Some of these will cause strains of the bonds.

Since the neutrons have an initial neutron energy of 2 Mev and only about 25 ev are required to move an atom from its position, it is obvious that a single neutron can cause a large number of vacancies. Since the neutron has relatively little effect on the electrons, a fast neutron will lose most of its energy by direct impact or knock-ons. These will displace atoms and many of the displaced atoms will have sufficient energy to displace other atoms. Those atoms which absorb a portion of the neutron's energy without being actually displaced will exhibit the energy as heat. The net result is that most of the energy of the neutron is converted to heat, but the creation of vacancies and the movement of atoms from their normal sites uses energy, so a portion of the energy will be stored in the free and distorted bonds. Whenever a neutron displaces an atom with sufficient energy

so that it travels any distance, it will result in a thermal spike as discussed above. Each neutron in slowing down will create about 2000 vacancies, interstitials, and thermal spikes. These will be located at considerable distances from one another due to the relatively large distances traveled by the neutron between collisions.

A fission fragment is a highly charged ionized particle that creates a tremendous disturbance among the atoms through which it travels. While it starts out with energy of perhaps 80 Mev, most of this energy is lost by ionization of the neighboring atoms. The fission fragment is travelling at such speed that the electrons of the surrounding atoms have no time to adjust and they are swept off or displaced with the absorption of energy. It has been postulated that the velocity of the fission fragment at which the electrons can rearrange themselves is that at which the two velocities are equal. It is only after the fission fragment has been slowed by these electronic effects to the point that the electrons can readjust slightly that the fragment begins to have knock-on collisions and causes atomic displacements in any number. Direct impacts which occur with full energy would cause the displacement of the struck atom with sufficient energy to cause partial electron removal and an ionizing particle. The net result would be similar. The fission fragments themselves are probably very highly, if not completely, ionized. As the fission fragment slows down, it will collect electrons and its effectiveness as an ionizing agent will decrease.

Finally, when the fission fragment has only 1 or 2 Mev, it will begin to have knock-on collisions. The displaced atoms will displace others, etc., until the fission product is thermalized. Perhaps 50,000 atoms are involved with perhaps the order of 5,000 to 8,000 vacancies, interstitials, and thermal spikes formed.

Radiation damage has been studied by bombarding materials with the cyclotron beam, etc., and by placing samples in reactors. Rates of damage are much higher and more uniform when cyclotrons are used since the beams can be directed at a small target and the beams are made up of monoenergetic particles.

Irradiation damage due to (n, γ) reactions in structural materials are small since the structural parts have low cross sections and only one impurity atom is formed per neutron compared with 2,000 vacancies created by the slowing down process, etc. Mg, Cu, Ni and Fe register no change in electrical conductivity on irradiation. At liquid air temperatures irradiation damage can be induced in these materials, but this largely anneals at room temperature.

Calkins presents a table concerning the mechanism of irradiation damage which is reproduced here as Table I.

Table I

MECHANISM OF RADIATION DAMAGE BY REACTOR RADIATION

Damage by fast neutrons:

Scattering: recoil ion, proton ejection

Displacement: neutralized recoil ion

Damage by gamma rays:

	Atomic No.	
	27 or less	82 or greater
Photoelectric	< 0.1 Mev	< 0.5 Mev
Compton effect	0.1-10 Mev	1.0-4 Mev
Pair production	>10 Mev	> 4 Mev

Damage by thermal neutrons:

 (n, γ) reactions, $\text{Cl}^{37}(n, \gamma)\text{Cl}^{38}$ (n, p) reactions, $\text{N}^{14}(n, p)\text{C}^{14}$ (n, α) reactions, $\text{B}^{10}(n, \alpha)\text{Li}^4$

Fission product damage:

Direct ionization

Displacement (neutralized fission ion)

Beta ray damage:

Direct ionization

Without going into details, it is evident that organic compounds will be rapidly damaged in a high neutron flux. It is, therefore, important that no lubricants or insulation materials be placed in reactors. This means that all neutron and temperature-sensing instruments must be built with inorganic insulators.

No organic materials can be permitted in contact with the coolant since they will be carried into the reactor and carbonized.

The initial damage to an organic liquid will be polymerization due to breaking of bonds and evolution of hydrogen. This will be evidenced by increased viscosity or cross linkages in the case of plastics. Liquids will solidify. With continued damage tars and finally chars are formed.

Normally in power reactor design sufficient shielding will be provided that no damage will occur outside of the shield. Radioactive coolants and especially homogeneous reactor solutions will cause trouble. Gaskets, seals, pump parts, lubrication materials, and electric insulation must be considered.

The damage to metals used in reactors as cladding and structural materials is not of great significance. Al and Zr self-anneal at room temperature and no damage is to be expected at operating temperatures in fuel elements. Since fuel elements are continually renewed, damage to structural materials is greater than to the fuel cladding but even here is not considered in design of components using these materials.

The damage to structural steel, however, is significant, in that irradiation damage affects steel somewhat like work hardening. For stainless steel fuel cladding and core supports, this is probably of little concern and it is not seriously considered by the reactor designer. In the design of the pressure vessel for water-cooled reactors, low factors of safety are used and irradiation damage is given serious consideration. Each steel is thoroughly tested by irradiation of samples in high flux in the MTR and tested for change of properties. Usually, irradiation is carried to the equivalent of 10 to 20 years of reactor operation. This is possible since the pressure vessel is outside the core of the reactor and the flux level is low. Neutron and gamma-ray heating of the pressure vessel with the resultant thermal stress is also considered.

The changes induced by irradiation of structural metals and alloys are indicated below:

- + Penetration hardness, saturates
- + Tensile and yield strength
- Ductility, marked change
- + Electrical resistivity
- Impact transition temperature for ductile to brittle fracture is raised

Thermal healing activated by elevated temperatures

Ferrite precipitated from austenitic steels

The mode of damage results in changes to small portions of single crystals while cold work affects many crystals. The increase in yield strength, precipitation from solutions, and the changes in yield strength are more analogous to precipitation hardening or low temperature age hardening. In general, structural materials have excellent dimensional stability.

Perhaps the most significant damage is the increase in impact transition temperature of boiler steels. Sutton and Leeser present the data in Table II.

Table II

EFFECT OF IRRADIATION ON IMPACT PROPERTIES OF
BOILER STEELS

<u>ASME Type</u>	<u>Flux, nvt</u>	<u>Temp. °F</u>	<u>Change in Transition Temp. °F</u>	<u>Change in Max. Energy, %</u>
SA-70	7.5 x 10 ¹⁹ slow	125	+ 125	- 41
SA-212	8 x 10 ¹⁹ fast	125	+ 115	- 23
	9 x 10 ¹⁹ slow		+ 170	- 29
	2 x 10 ¹⁸ fast	580	+ 45	- 5
	8 x 10 ¹⁸ slow	425	+ 35	- 7
Al-killed, 0.34 C Steel	2 x 10 ¹⁸ fast	425	+ 20	- 4
	8 x 10 ¹⁸ slow	390	+ 20	- 2
		350	+ 20	- 4

Creep

The results of creep tests on various metals under irradiation have not been consistent. Some experiments have shown increased rates, and others indicate no gross changes. The effects in any case have been shown to be small.

LECTURE 16

MAINTENANCE OF COOLANTSReferences:

- "Radiation Damage to Water," A. O. Allen, Amer. Inst. Chem. Eng. Mon., 12, 238 (1954)
- "Ten Years Operating Experience of the O.R.N.L. Graphite Moderator Normal-Uranium Reactor," M. E. Ramsey, C.D.Cagle, Amer. Inst. Chem. Eng. Mon., 11, 149 (1954)
- "The Theoretical Distribution of Airborne Pollution from Factory Chimneys," O. G. Sutton, Quart. J. Roy. Meteorological Soc., 73, 426 (1947)
- "Test of Fiberfrax' Ceramic Fibers for High Temperature Gas Filtration," M. W. First, J. B. Graham, G. M. Butler, C. B. Walworth, Buffalo Forge Co., 1955.
- "Gas Evolution for Dosimetry of High Gamma, Neutron Fluxes," E. J. Hart, S. Gordon, Nucleonics, 12: No. 4, 40 (1954).

Water Decomposition

Water decomposes in reactors due to the presence of irradiation. The rate of decomposition is small. Since explosive mixtures may be formed or valuable heavy hydrogen lost, the gases are generally reformed and returned to the reactor. The mechanism of decomposition is complicated. Above 200°C, H₂ and O₂ are the products, while below this temperature O₂, and H₂O₂ are formed

In a closed system part of the decomposed water will recombine. This is especially true at high temperatures and pressures. On the other hand, in a system such as the MTR, the gases will be removed as rapidly as formed and there will be no back reaction.

Water decomposition results from interaction with protons removed by neutrons and by gamma rays. The protons can ionize water directly due to their velocity and own ionization or charge. The gamma rays do not ionize water directly, but are absorbed within the water by photoelectric effect, Compton effect, or pair production, and the resultant high speed electrons may cause ionization.

Some of the reactions are:

these free radicals then react:

These are forward reactions which convert water to H_2 and H_2O_2 . At high temperatures, the H_2O_2 decomposes into H_2O and O_2 .

Other possible reactions are:

or the net result

These reactions are the back reactions.

Still other reactions are:

to yield the net result

The decomposition of water is dependent upon:

Nature and intensity of radiation

Temperature of the water

Nature and extent of ionic
impurities present

Concentration of the decomposition
products in the water

The following conclusions may be drawn from the data obtained to date:

- a. Under normal operating conditions the amount of decomposition is low.
- b. Increased temperatures result in decreased amounts of decomposition products and apparently increased temperature also decreases the rate of decomposition.
- c. The presence of impurities results in increased decomposition rates and increased equilibrium amounts of decomposition products.
- d. At high temperatures a concentration of hydrogen of 5 to 10 psi partial pressure reduces the decomposition rate of pure water to essentially zero.
- e. At low temperatures the presence of hydrogen to 5 to 10 psi reduces the formation of decomposition products to very low values.

Experience has been obtained on several reactor tests; one of the most pertinent is presented below:

CP-3'

Epithermal neutron flux, 4×10^{11} ; water temperature, 100° F; volume of water under irradiation, 5,000 liters.

Initially, this reactor was built with water filters, but these were not used at all times. Due to aluminum corrosion, the water impurities became very high and ammonium hydroxide was added for pH control. At one time, the water contained about 1200 ppm ND_4NO_3 , 100 ppm Al, 50 ppm lead ion, and appreciable amounts of copper and other metal ions, and also chloride ions. The water had a specific resistance of 10,000 ohm cm and a rate of formation of deuterium of 60,000 to 100,000 ml/hr and of oxygen of 30,000 to 50,000 ml/hr. A still was installed and later ion exchange was added to purify the water. Under the new conditions of a specific resistance of 1,000,000 ohm cm, the rate of formation of deuterium was reduced to 200 ml/hr and the rate of formation of oxygen was negligible. The rates of formation of gas include the effects of corrosion of aluminum. With the high purity water, aluminum corrosion under the temperature conditions should be very small.

Boiling Reactors

While no data are available, we would expect boiling reactors to give appreciable amounts of H_2 and O_2 since these gases would be removed as rapidly as formed without appreciable back reactions. The gases would collect in the fixed gases from the condensers and would be either recombined catalytically or disposed of by mixing with ventilation air and rejected to the stack.

Water Purification

Deionization of water is carried out in mixed bed deionization equipment of regular commercial design. The water purity specifications are somewhat as follows:

Total solids	<1.0 ppm
Na	<0.5
Cl	<0.2
Fe	<0.2
pH	6-7
Specific Resistance	1,000,000 ohms/cm

The deionization equipment is designed to deionize a sidestream of such size that the total water of the system is run through the deionization beds every few hours. The side stream is run through a heat exchanger and cooler to cool it nearly to room temperature, filtered, and is then passed through the ion bed. Finally, it is partially reheated by passing through the heat exchanger and returned to the system. A pump is located before or after the ion beds. The impurities retained by the ion exchange beds means that the beds become highly radioactive. The resins can be washed out into burial tanks for disposal. When heavy water is being deionized, it is necessary to replace the D_2O with H_2O and to reclaim the mixed D_2O - H_2O water formed in reclaiming the D_2O . It is not necessary to use special resins as the H atoms from the resins do not interchange with the D atoms of the water.

Purification of Liquid Sodium

Sodium is usually very pure as received. It contains small amounts of $CaCl_2$ (about 0.04%) and some oxides. In addition, traces of rare earths must also be present as some activity builds up during use which has a longer half-life than sodium. Du Pont sodium has the following impurities (average analysis):

Al	1.7	Ca	350	Cs	2	Li	67	S	35
B	2	Cd	<1	Cu	<1	Mn	<1	Si	21
Bi	<1	Cl	190	Fe	0.5	Ni	<1		
Ba	<2	Co	<1	Hg	2	P	11	Total rare earths <0.5	
C	40	Cr	<1	K	150	Pb	<1		

Heating to 800 to 900°F decomposes the NaH or NaOH with the removal of the H₂ in the gas stream. Na₂O is very insoluble in Na at low temperatures, but solubility increases with temperature as indicated by the following solubilities:

200° F	0.001 wt % oxygen
400	0.003
600	0.017
800	0.052
900	0.077

This suggests that oxide removal may be readily carried out by cooling a side stream and filtering. A suggested trap utilizes stainless-steel wool at 300°F.

Since uranium and thorium are essentially insoluble in sodium, there is little danger of contaminating the sodium with powdered uranium oxides such as are produced in water-cooled reactors when a clad failure occurs.

Gas Coolants

Two problems exist in use of gas coolants as far as activity is concerned. These are, one, the problem of activity build up in air, and, two, the problem of active particulate matter. Dust in the air, graphite dust, or uranium oxides formed in case of a slug failure can all cause active dust problems.

Induced Activity

Argon-40, which comprises about 1% of the air, undergoes reaction to form A-41, which is a beta emitter with a 110-minute half-life. The product is non-active K. The danger from A-41 is very slight except for the high beta concentration in an actual cloud.

It is usually necessary to break up the volume in a reactor into several volumes in calculating A-41 buildup, as the fluxes will vary. The Brookhaven reactor emits some 6,000 curies/day while the MTR gives about 1,500 curies/day.

The small amounts of C-14 formed are more dangerous than the A-41 due to long half-life and the use of CO₂ in the biological processes. However, this is usually not a serious problem.

Air Dispersal

Under ordinary conditions, air dispersal of radioactive A and CO₂ causes no difficulty. Danger is encountered when there is an inversion and the stack air drops down and spreads out at the base of the stack. Such conditions are not common, but do occur. Oak Ridge is very bad in this respect, although the normal inversion at Oak Ridge means that the stack gases do not leave the valley. They seldom drop to the ground.

The dispersal of particulate matter in case of a slug failure is serious unless the air is filtered. In addition, dusts which are carried into the reactor with the air stream become radioactive if they settle out and later become airborne. Pieces of graphite in the X10 reactor are frequently airborne.

FG25 backed by FG50 glass-wool filters give excellent inlet and coarse stack filters. The stack filters can be backed by a separate set of CWS #6 or AEC #1 filters. The efficiency is 99.9 per cent or better for all particles above a fraction of a micron diameter. Below this size, Brownian movement and adsorption become important and most of these particles are removed. Buildup in the glass filters at Oak Ridge is about 2 in. H₂O per year pressure increase with a two-year life for the filters. The CWS filters buildup about 1.6 in. of H₂O pressure per year with a life of 2-1/2 years.

A Fiberfrax filter has been developed that has shown promise for use at temperatures up to 760°C.

Fall-Out

The main concern of the Reactor Hazards Committee has been the fall-out of particulate matter in case of a reactor failure. For example, the MTR builds up as much as 3.5×10^7 curies of activity before discharge. Perhaps 25% of this is gaseous at ordinary temperatures. In case of a major runaway and assuming that the reactor melted, all the gaseous products could be released in a cloud. In case the reactor core should somehow be vaporized, not only would there be nine million curies of gaseous products, but some 27,000,000 curies of particulate matter in the cloud.

The usual calculation of the spread of an air stream is made by use of the Sutton formula:

$$X(x,y,z) = Q \frac{e^{-\frac{y^2}{C_y x^{2-n}}}}{\pi u C_y C_z x^{2-n}} \left[e^{-\frac{(z-h)^2}{C_z x^{2-n}}} + e^{-\frac{(z+h)^2}{C_z x^{2-n}}} \right]$$

X = concentration, mg/m³

Q = rate of emission, mg/sec, or strength of source, curies/sec

u = wind speed, m/sec

n = a pure number

C_y = virtual crosswind diffusion coefficient, m^{n/2}

C_z = virtual vertical diffusion coefficient, m^{n/2}

h = stack height

The effluent plume is assumed to be a form lying between a cone and a paraboloid. The concentration along the X axis decreases as x^{2-n} where n varies between 0 and 1. The maximum ground level concentration is found to be inversely proportional to the square of the stack height although this does not apply to concentrations at fixed points.

C_z and C_y are dependent upon the amount of turbulence in the air. They are usually evaluated as 0.07 except for inversion conditions.

At ground level we may set $x = 0$ and $y = 0$; then

$$C_m = \frac{2Q}{\pi u C_y C_z x^{2-n}} e^{-\frac{h^2}{C_z x^{2-n}}}$$

This equation may be differentiated to give the maximum concentration at ground level down wind from a stack. The maximum concentration is

$$C_m = \frac{2Q}{\pi u e h^2} \frac{C_z}{C_y} \quad \text{curies/cm}^3$$

at a distance from the stack of

$$X_m = \left(\frac{h^2}{C_z} \right)^{\frac{1}{2-n}} \quad \text{cm} \quad .$$

Particulates continuously lose activity by fall-out to the ground. At the same time, there is an increase in concentration near the ground compared with the top of the cloud. The concentration at ground level may increase to a maximum and then decrease slowly as the particles decay.

The danger from a complete vaporization of a Hanford reactor is great since the activity might be carried for hundreds of miles.

These facts explain the isolation and enclosures which have been used with reactors.

LECTURE 17

XENON AND SAMARIUM: CHANGES IN REACTIVITYReference:

"The Elements of Nuclear Reactor Theory," S. Glasstone,
M. C. Edlund, Van Nostrand, 1952

Due to the formation of the very high cross section Xe^{135} by decay of I^{135} , a reactor which has been shut-down for an hour or so may have lost so much reactivity that it cannot be started until the Xe^{135} itself has decayed. The Xe^{135} is formed to the extent of 0.003 atom per fission, while Te^{135} is formed to the extent of 0.056 atom per fission of U^{235} . The decay is so rapid that we may consider I^{135} as formed at the rate of 0.056 atom per fission. The decay chain is:

The cross section of Xe^{135} is very high ($3.5 \times 10^6 \text{ b}$) due to resonance absorption.

The net result of the Xe^{135} chain is that in normal operation the Xe is partially burned out as fast as formed to give Xe^{136} which is stable and has low cross section. The remainder of the Xe^{135} decays to Cs^{135} . Thus, an equilibrium is set up so that the rate of production equals the rate of burnout and decay.

Difficulty is encountered after shutdown since the I^{135} continues to decay and the resultant Xe^{135} is not burned out as formed. It builds up and, due to its high cross section, increases the poison in the reactor to the point that the reactor cannot be made critical unless there is a large amount of excess reactivity built into it. Frequently, the amount of reactivity to override the xenon at all times after shutdown is either unsafe or impractical. Fortunately, after some 48 hours, both the I^{135} and Xe^{135} have decayed to a point which permits start-up.

A specific example of actual values may be taken from the operation of the MTR. The values of k are listed below for operation at 30 Mw:

$\Delta k/k$ for Xe poisoning, operation - 0.039

$\Delta k/k$ for Sm poisoning, operation - 0.010

The MTR was designed to permit 30-minute shutdowns. However, it can be reloaded so easily that it is practicable, if desired, to reload a portion or all the core. The fuel elements removed from the reactor may be reloaded after Xe decay.

An actual test was run on the MTR at 5 Mw in order to obtain a low enough value that the reactor could over-ride the xenon. This also avoided temperature corrections.

The experiment was conducted by reloading with fresh fuel. The reactor was operated at 5 Mw for 49 hours until the xenon equilibrium had been reached, the power was reduced to 30 kw, and the decay followed, see Figure 1.

The normal thermal flux in the MTR is 2×10^{14} at 30 Mw and 3.3×10^{13} at 5 Mw. The equilibrium burnout value of the Xe is indicated by Table I.

Table I

POISONING AT VARIOUS FLUXES DUE TO XENON

ϕ	P_0	ϕ	P_0
1×10^{12}	0.007	1×10^{14}	0.046
5×10^{12}		2×10^{14}	
1×10^{13}	0.03	1×10^{15}	0.048
5×10^{13}			0.05

Since Xe^{135} is removed by decay and by neutron absorption or burnout, the data would have to be calculated by taking both the decay and burnout into consideration.

Glasstone and Edlund and the Reactor Handbook give equations for the build up of xenon, for its equilibrium concentration, and decay.

Samarium

Promethium-149 has a half-life of 47 hours. Neodymium-149 has a half-life of 1.7 hours. Again, we may consider the longer life isotope as the one formed. The fractional yield of promethium then is 0.014. The cross section of Pr^{149} is very small so it may be neglected.

Figure 1

PRODUCTION AND DECAY OF XENON.
THE INITIAL POWER WAS 5 MEGAWATTS,
AND THE DECAY POWER WAS 30kw.

Samarium-149 is stable and not formed in fission. The equilibrium concentration for samarium is independent of the flux. The time to arrive at equilibrium is about 11 days.

Changes in Reactivity

In addition to the changes in reactivity caused by Xe^{135} and Sm^{149} , there are longer but very important changes in reactivity due to the changing concentration of different isotopes and to the general poisoning effect of the low cross-section fission products. Studies have been made of the various factors which enter into reactivity changes and the actual changes which occur.

Since it has been known that reactivity is lost for long irradiations, there are several obvious methods of operation which will work. These are (1) batch irradiation with excess activity added by control or shim rods as required; (2) batch irradiation with excess reactivity utilized to give additional production; and (3) excess reactivity in fresh fuel is utilized to make up reactivity losses by frequent changes of a small fraction of the fuel elements.

The excess reactivity can be obtained by use of enrichment or by recycling the Pu and U^{233} formed in the reactor.

As the fuel is fissioned, the following changes occur in the fuel:

1. The fission products absorb neutrons.
2. Fission and capture in the fissionable isotopes reduces the number of fissionable atoms and the reactivity is lowered.
3. Capture in fissionable isotopes produces new isotopes which are non-fissionable. Some of the new isotopes are fertile.
4. Capture in the new fertile isotopes produces new fissionable materials which increases the reactivity.

The long term trends in reactivity must consider the following items:

Fissionable nuclei	Fertile nuclei
Parasitic nuclei	Fission products
Higher chain nuclei	Neutron leakage
Fast fission effects	

The U-Pu system has been studied in much detail. The reactions studied are shown below:

The equations for isotopic abundance are presented below. In these equations:

σ_a = cross section for absorption

ϵ = fast fission effect

$\eta = \nu \cdot \frac{\sigma_a}{\sigma_f}$ = fast neutrons emitted per neutron absorbed in fuel

λ = decay constant

f = thermal utilization

$$\phi_t = \frac{K}{\sigma_f \text{U}^{235} + \sigma_f \text{Pu}^{239} + \sigma_f \text{Pu}^{241}}$$

Σ_a^{235} = neutrons absorbed in U^{235}

$\Sigma_a^{235} \eta \epsilon$ = number of fast neutrons per thermal neutron absorbed

$\Sigma_a^{235} \eta \epsilon (1 - p)$ = number of resonance neutrons captured per thermal neutron absorbed.

$\Sigma_a^{235}(p)e^{-B^2\tau}$ = number of fast neutrons remaining after leakage.

$$\text{ICR} = \frac{\Sigma_a^{238}}{\Sigma_a^{235}} + \eta \epsilon (1 - p) e^{-B^2\tau}$$

The equations of isotopic abundance which were used are:

$$\text{U}^{235}: \frac{d\text{U}^{235}}{dt} = -\phi_t \sigma_a \text{U}^{235}$$

$$\text{U}^{238}: \frac{d\text{U}^{238}}{dt} = -\phi_t \left\{ \sigma_a \text{U}^{238} + \epsilon (1 - p) e^{-B^2\tau} (\sigma_a \text{U}^{235} \eta + \sigma_a \text{Pu}^{239} \eta + \sigma_a \text{Pu}^{241} \eta) \right\}$$

$$\text{U}^{239}: \frac{d\text{U}^{239}}{dt} = \phi_t \left\{ \sigma_a \text{U}^{238} + \epsilon (1 - p) e^{-B^2\tau} (\sigma_a \text{U}^{235} \eta + \sigma_a \text{Pu}^{239} \eta + \sigma_a \text{Pu}^{241} \eta) - \lambda \text{U}^{239} \right\}$$

$$Np^{239}: \frac{dNp^{239}}{dt} = \lambda U^{239} - \lambda Np^{239}$$

$$Pu^{239}: \frac{dPu^{239}}{dt} = \lambda Np^{239} - \phi_t \sigma_a Pu^{239}$$

$$Pu^{240}: \frac{dPu^{240}}{dt} = \phi_t \left\{ \sigma_c Pu^{239} - \sigma_a Pu^{240} \right\}$$

$$Pu^{241}: \frac{dPu^{241}}{dt} = \phi_t \left\{ \sigma_a Pu^{240} - \sigma_a Pu^{241} \right\}$$

The cross sections of most fission products are not known accurately at present. Therefore, it was necessary to throw all of them together and assign the same value to them.

Calculations were carried out and the results plotted in various ways. The most significant plot is presented in Figure 2.

The following general conclusions may be drawn from the studies:

1. For high conversion ratio reactors, reactivity increases initially and reaches a maximum after which it decreases.
2. Initial conversion ratios as high as 1.2 are required for constant reactivity for irradiations as high as 20,000 MWD/ton.
3. With initial conversion ratios less than 1.0, loss of reactivity is rapid and becomes very serious after 10,000 MWD/ton.
4. The optimum method of operating power reactors from a reactivity view is to change a portion of the fuel elements at frequent intervals in order to utilize the initial gain of reactivity and to avoid excessive shim and control problems. However, economic balances will be required to determine what is the optimum method in a particular reactor plant since the reactor must be shut down to change fuel elements.

Figure 2

VARIATION OF REACTIVITY VS IRRADIATION TIME

Note:

1. These curves are based on a specific power of 19.27mw/ton. For any other specific power, vary time directly as the specific power.
2. Σ and ρ are assured constant.
3. $\Sigma_a^{\text{Mod} + \text{Structure} + \text{Xenon}} = 5\% \Sigma_a$
4. σ_a fission fragments = 50 barns / fission.

LECTURE 18

REACTOR CONTROLReference:

"The Elements of Nuclear Reactor Theory," S. Glasstone, M. C. Edlund, Van Nostrand, (1952).

Basically, the control of a nuclear reactor depends on the utilization of the delayed neutrons. In the fissioning of U^{235} most of the neutrons are emitted in a very short time after fissioning takes place. However, a fraction, about 0.0075, of the neutrons are delayed so that the average time lapse for all neutrons emitted is of the order of 0.1 second. The neutrons emitted at the time of fission are called prompt neutrons.

For a thermal reactor the life of a neutron after emission is of the order of 0.001 second. The increase of neutrons with time is given by

$$\frac{dn}{dt} = \frac{n(k-1)}{\ell} = \frac{nk_{ex}}{\ell}$$

in which

n = number of neutrons

$k-1$ = increase of neutrons per generation

ℓ = average life of neutrons in seconds.

This integrates into

$$n = n_0 e^{(k_{ex}/\ell)t}$$

where n_0 is the number of neutrons of the first generation.

Suppose $k = 1.005$ and $k_{ex} = 0.005$; if $\ell = 0.001$ (without delayed neutrons), then in 1 second, $n = n_0 e^5$, or there is a 150-fold increase. For $\ell = 0.1$, after 1 second, $n = n_0 e^{0.05}$ or 1.05.

The life and pertinent data concerning the most important groups of delayed neutrons are presented in Table I.

The control system for thermal reactors should be designed so that k cannot be increased over 0.5% in a very short time. This means that the design must be such that the operator, due to human error, cannot add more than 0.5% k at one time.

Table I

DELAYED NEUTRONS

<u>Half-Life,</u> <u>Sec</u>	<u>Mean-Life,</u> <u>Sec</u>	<u>Constant</u> <u>λ_i (sec⁻¹)</u>	<u>Fraction</u> <u>β_i</u>	<u>Energy,</u> <u>Mev</u>
0.43	0.62	1.16	0.00084	0.42
1.52	2.19	0.456	0.0024	0.62
4.51	6.50	0.151	0.0021	0.43
22.0	31.7	0.0315	0.0017	0.56
55.6	80.2	0.0124	0.00026	0.25

A second factor which is of importance in reactor control is the temperature effects. These effects are frequently very complex and difficult to analyze during design. The major effects which are obvious are the expansion of the core due to the heating and the general rise in average neutron energy with temperature. For power reactors which are designed to operate at high temperatures, these effects may be appreciable. Due to the density decrease, the diffusion length and age of the thermal neutrons will be increased. Leakage in general will be increased. All these factors indicate a higher critical mass. There will be complex changes in the cross sections. For example, the resonance absorption band in U^{238} will be widened and decreased in height. Examples of reactor temperature coefficients are given in Tables II and III.

Table II

TEMPERATURE COEFFICIENTS OF VARIOUS REACTORS

<u>Reactor</u>	<u>Temperature Coefficient</u>
HRE at 25°C	-3.8×10^{-4} in $k_{eff}/^{\circ}C$
at 215°C	-1.1×10^{-3} in $k_{eff}/^{\circ}C$
BNL fluid fuel	-2×10^{-4} in $k_{eff}/^{\circ}C$

Reactors are autocatalytic when their reactivity increases with temperature, and non-autocatalytic when their reactivity decreases with temperature.

Any reactor which is autocatalytic is potentially dangerous since a small excursion in reactivity may become magnified, leading to high power level excursions. If such high level excursions continue, they can lead to damage to the reactor or possibly to release of fission products to the surroundings.

Table III

TEMPERATURE COEFFICIENT IN MTR CRITICAL ASSEMBLIES

Radius, cm.	k	Temperature Coef., $10^5 \Delta k/k$ per °C			
		Expansion of Fuel Bearing Al Plates	Expansion of Water Between Fuel Plates	Change in Microscopic Cross Sections With Neutron Temp.	Total Coef, $10^5 \Delta k/k$ per °C
12.78	1.606	-1.6	-3.8	-15.7	-21
17.85	1.432	-1.4	-3.0	-10.5	-15
21.12	1.373	-1.3	-2.8	- 8.9	-13

In water-cooled reactors, a power excursion leads to a rapid increase in the temperature of the water with an expansion of the water and a decrease in the moderator. This lowers the reactivity slowing the reaction. The key question is whether the rate of water expansion will be great enough to over-ride the rate of reactivity increase.

Let us look at the problem from the excess reactivity point of view. Most reactor systems have several per cent of excess k or reactivity available in the control system. If the design is such that the operator can increase the excess k to the point that there are, say two per cent, excess k in the reactor, a power excursion will result since this is well over prompt critical. The two per cent has been considered about the top excess k that would be possible to add to a reactor under the most adverse conditions. A check was made of this in the Borax Reactor (see the lecture on Boiling Reactors).

Control systems are usually broken down into the three functions - shim, fine control, and safety. In power reactors with several critical masses present, a large amount of poison, or high cross section production material, must be added to counteract the excess reactivity present. This is known as shim control. For normal operations very small changes in reactivity must be made continually in order to hold k constant. This is the fine or running control. Then, finally, it is desired to add sufficient poison during loading and maintenance operations to make sure that there will be no power surge even though a shim control rod is removed for repairs. Also, it is always necessary that there be poison rods available that can shut down the reactor in a very short time. This is known as safety control. Frequently, two of these functions will be combined into one set of rods.

The introduction of control may be either positive or negative in that excess reactivity or poison may be added. As an example, we may operate a reactor such as the MTR by use of insufficient fuel for criticality and use U^{235} shim rods to make up the reactivity, or the reactor may be supercritical and controlled by addition of poison rods.

There are a number of methods of control as noted below:

- a. Insertion of poison rods into the core or between the core and reflector.
- b. Add or remove fuel from the core.
- c. Move portions of the core.
- d. Add fluid poison to the coolant.
- e. Add a burnable poison to the core in such amounts that it is burned at a rate which compensates for loss in reactivity.
- f. Remove moderator from the core by pumping, dumping, or boiling.
- g. Rotate sections of reflector material which are covered on one side by a neutron absorber.

The insertion of a poison rod is used in most thermal reactors as the means for fine control. Addition of fuel or burnout poison is used for shim purposes. Safety rods are usually poison. A poison rod which is inserted into a reactor provides a neutron sink and the flux pattern extrapolates to zero just inside the rod giving the effective radius, r_{eff} , of the rod. If we add a poison rod to the center of a cylindrical reactor and assume that the power remains constant, we displace the flux outwards from the center. If a small rod or a rod lightly poisoned is inserted in the center, this will depress the flux slightly. By means of many rods the flux pattern can be altered to the point that it is constant or flat nearly to the edges of the cylinder. Lengthwise of the coolant passages the cosine flux pattern can be flattened so that the heat flows from the fuel elements to the coolant and is maintained at such rates that the fuel element surface temperatures are held constant. This is known as roof topping. Flattening and roof topping permit a reactor to produce more power than cosine operations and not exceed the maximum allowable fuel plate temperature with excessive corrosion. Roof topping requires good reflectors.

Most reactors are designed so that there are at least two independent safety systems. In addition, design is usually carried out so that the reactors will be non-autocatalytic to the extent that little or no damage will result from a power surge. Recently, thought has been given to the design of a so-called third safety device as a further safety feature. The normal control and shim safety devices are under the control of the operator. The idea of the third safety device is that it would be completely independent of all other devices and not subject to control by the operator. Such a device would be initiated by a reactor fuze and would drop or force poison into the reactor. Ideas have been based on heating of a soldered U^{235} joint that trips a poison source. Such a device, in principle, would be similar to the low melting point metal fuze in automatic sprinkling systems for fire prevention. In use, the reactor fuze would function at perhaps 1.5 x normal power and

release a poison so rapidly that the reactor would be shut down before the power reached a dangerous level. While several such devices have been suggested, no completely satisfactory one has been developed.

The control devices used in a few of the various reactors built to date are discussed in the following paragraphs:

- CP-2:** Shim - 1 horizontal, steel supported Cd strip; Regulating - 2 horizontal, steel supported, Cd strips; Safety - 3 horizontal, steel supported, Cd strips; Reactivity change: 0.005.
- X-10:** Shim - four $1\frac{3}{4}$ in. x $1\frac{3}{4}$ in. x 19 ft horizontal boron-steel rods; Regulator - two $1\frac{3}{4}$ in. x $1\frac{3}{4}$ in. x 19 ft horizontal boron-steel rods; Safety - four $1\frac{1}{2}$ in. diam x 8 ft vertical boron steel rods; two $1\frac{3}{4}$ in. diam x 17 ft 2 in. vertical tubes for $\frac{3}{8}$ in. boron-steel shot; Reactivity change (late 1944 loading): 0.0389.
- Brookhaven:** Regulator - two 2 x 2 in. x $25\frac{1}{2}$ ft, horizontal $1\frac{3}{4}\%$ boron steel rods; Shim and safety - fourteen 2 x 2 in. x $25\frac{1}{2}$ ft horizontal $1\frac{3}{4}\%$ boron steel rods; two $3\frac{1}{2}$ in. diam diagonal shot wells in gap 2- $3\frac{1}{2}$ in. diam x 25 ft shot wells.
- MTR:** Shim - four combination Al-Cd box and fuel element rods, four combination Al-Cd box and Be rods; Regulator two $1\frac{1}{2}$ in. diam Al-Cd tubing; Safety - effected by all rods.
- CP-5:** Shim - four 1 x $5\frac{1}{2}$ x 60 in. Al-jacketed, Cd, signal-arm type rods; Regulator - one $1\frac{1}{2}$ in. OD, Al-jacketed Cd, vertical rod; Safety - effected by shim rods, quick drain of D₂O to core level; Reactivity change (by safety rods) - about 0.20.

We should note that the net effect of the temperature coefficients is to reduce reactivity. Such reactors as the HRE have so high a negative temperature coefficient that no control rods are required. In this case, fuel is added until the reactor becomes critical and the temperature is raised to the regular operating point. Further changes will be automatically adjusted to the power requirements on the electrical generator. Use of more power requires additional steam to the turbine; this reduces the temperature in the heat exchanger and, thus, the reactor. The reactivity changes in such a way that more heat is produced. This results from the slightly lowered reactor temperature and, thus, a denser fuel with more fuel in the reactor.

A power surge in the MTR would cause film or bulk boiling, a decrease in the moderator present, and shutdown of the reactor.

All reactor control systems are designed on the basis of fail safe. For example, rods are usually vertical and held to the drive by magnetic clutches so that they will drop into the reactor if there is a power failure. In the Brookhaven reactor horizontal rods are used, but the motor drives are operated constantly with heavy flywheels. These have sufficient energy that the rods will be driven in automatically in case of power failure. Heavy water reactors are usually provided with quick dump tanks for removal of the moderator. For very high flux reactors, dumping of the moderator could not be used as the decay heat will melt the fuel elements releasing the fission gases.

In case of failure of the control systems, the energy release in most reactors will shut them down without excessive damage to the surroundings. An energy excursion in CP-5 would probably blow steam out at the very worst, although the building is sealed and designed to withstand some pressure.

Let us look at the MTR as an example of a control system. Light water reactors are capable of very short periods, about 10^{-3} sec, minimum in the prompt critical range. Actually the control system will handle a period as low as 0.1 sec. The safety devices are designed to prevent a period of less than 1 sec from occurring.

The MTR has four shim-safety rods made up of Cd and enriched U^{235} plates so arranged that the Cd section is in the core during shutdown, and these rods are withdrawn in operation. Vertical withdrawal moves the Cd out of the core and pulls in whatever U^{235} is needed to sustain the reactivity. These rods may be dropped on scram signal by release of magnets. A period of 7 sec blocks withdrawal of the rods and a period of 5 sec drives them in and a period of 1 sec or less scrams the reactor releasing the magnets.

The control system is a combination of manual and operator control with the automatic safety system capable of over-riding the manual system if the operator manipulates the controls in an unsafe manner. The principal parts of the systems are:

- Sensing element, ion and fission chambers
- Regulating rods
- Shim rods
- Rod drive mechanisms
- Magnets
- Servos
- Period meters
- Interlocks
- Amplifiers

LECTURE 19

SHIELDING, GAMMA-RAY HEATINGReferences:

- "Introduction to Nuclear Shielding for Engineers," H. M. Glen, ORNL
 "Reactor Handbook," Vol. I

Definitions

There are a number of definitions of interest in shielding work. Some of these are:

Curie, c: The amount of radioactive material that gives 3.7×10^{10} disintegrations per second.

Roentgen, r: The roentgen is that amount of gamma radiation that will produce 1 electrostatic unit of ions, of one sign, in 1 cm^3 of dry air under standard conditions of temperature and pressure. The milliroentgen, mr, is usually used. The roentgen measures the quantity of radiation and not the intensity. Intensities are given per unit of time, as 1.0 r per hr.

Human Tolerance: The human body can tolerate indefinitely a weekly total body irradiation of 0.250 r of gammas, roentgen equivalents of neutrons or betas, or any combination. The life time total is 300 r or r equivalents. Design tolerances are 7.5 mr/hr or 50 mr/day.

Roentgen Equivalent Physical, rep: When dealing with radiations other than gammas, the rep is used. It is defined as that quantity of ionizing radiation that dissipates 93 ergs/gram of tissue.

Roentgen Equivalent Man, rem: Is that amount of radiation absorbed in tissue which has the relative biological equivalence in man of 1 r of gamma rays. The secondary ionization from recoil protons produced by fast neutrons (2 Mev) is about ten times as damaging as the same amount of energy absorbed from secondary electrons produced by gamma rays or for fast neutrons: 1 rem = 0.1 rep. We may add up different types of radiation as follows:

30 mr/hr gammas	= 30 mrem/hr
1.5 mrep/hr fast neutron	= 15 mrem/hr
3500 slow neutrons/cm ² sec	= 15 mrem/hr
<hr/>	
Total	= 60 mrem/hr

Maximum Permissible Exposure: This is usually 60 mrem/day or 300 mrem/wk. Examples are given below:

Gamma rays	1300 photons/cm ² sec of 1 Mev
Beta	17 betas/cm ² sec of 1 Mev max
Fast neutrons	22 fast neutrons/cm ² sec of 2 Mev
Thermal neutrons	600 thermal neutrons/cm ² sec of 0.02 ev
Alpha, internal only	0.003 alpha particles/cm ² sec of 5 Mev

Practical Limits

These theoretical limits are not of much avail directly. We usually wish to reduce the fast neutrons essentially to zero at the edge of a reactor shield and the gammas and thermal neutrons to 200/cm² sec each for usual human tolerance.

Attenuation of Gamma Rays:

Two measurements of the gamma energy emitted per fission are:

<u>Energy/fission</u>	<u>Energy/photon</u>
5.1	1.0
4.6	2.5

In order to be safe, we usually assume 5 gammas of 2.5 Mev.

Fission products release energy rapidly over the first few seconds. In the period 10 to 3×10^6 seconds we may use the gamma release rate in Mev/sec fission as

$$1.6 t^{-1.21}$$

where t is in seconds. The total or equilibrium delayed gamma energy per fission is 5.3 Mev.

In the attenuation of gamma rays the most important process is Compton scattering. In this process the gamma ray is considered as a particle; on collision of the gamma ray and an electron, the gamma ray is scattered with reduced energy while the electron recoils with the energy which the gamma ray lost.

A second process occurs when the gamma energy is high enough to be converted into an electron-positron pair. Here the gamma ray disappears and the total energy of the electron pair is equal to the original gamma-ray energy. This process occurs most frequently in heavy elements.

The third process is the photoelectric effect. In this process a gamma ray, which is incident on an atom, yields up all its energy in ejecting an electron from one of the inner shells. The vacant energy level is filled by electrons from lower energy levels, and lower energy gamma rays are emitted. This process also is favored by high mass.

As a result of these processes, there is a buildup of the total gammas. The degraded or less penetrating gammas are built up numerically in accordance with the following table based on the amount of attenuation. The buildup factor is the ratio of the emergent gamma flux to the undeviated primary gamma flux.

Attenuation	10	10^2	10^4	10^6	10^8
Buildup factor	2.8	4.2	7.1	10.5	13.9

Thus if we have an attenuation of primary gammas by a factor of 10^6 , the actual decrease in gamma radiation is effectively 10^5 . The resultant gammas are of longer wave length.

Actual attenuation takes place by scattering, lowering the energy level until absorption can take place, and absorption as heat without emission of new rays.

We would expect attenuation to follow

$$I = I_0 e^{-\tau x}$$

in which $\tau = \tau_{ph} + \tau_{Comp} + \tau_{pair}$, but we must add the buildup factor, τ/ρ . The mass absorption coefficient is practically constant for all elements for a given frequency provided the energy is below the pair production range. The energy ranges in which each of the three effects are most prominent are, in Mev:

Element	Photo	Compton	Pair
Al	< 0.02	$\sim 0.02 - 15$	> 15
Pb	< 0.02	$\sim 0.2 - 2$	> 2

From this table it is obvious that in reactors pair formation is not very important and Compton scattering is most important in the thermal shield, and finally, the photoelectric effect is most important in the outer sections

of the biological shield. Gammas for (n, γ) capture reactions for thermal neutrons vary from 2.2 Mev for the $H^1(n, \gamma)H^2$ reaction to 8 to 9 Mev for heavy target nuclei.

Neutron Attenuation

Absorption of thermal neutrons is very rapid in high neutron absorbing materials. Two difficulties occur, however: absorption in Fe results in the emission of a strong gamma ray of 6 Mev; the other is that the new nuclei may be radioactive and emit a gamma ray. Boron absorbs thermal neutrons without the emission of gammas and without becoming radioactive. It is used to remove the thermal neutrons. The fast neutrons pass on through. Since these will be more readily absorbed at low energies, we try to thermalize them. The process of thermalization in a shield will be different than in a moderator. In the shield the presence of heavy elements will result in inelastic scattering. In this process the high energy neutron collides with the nucleus of a heavy element and loses a large part of its energy in raising the nucleus to an excited state. The absorption is a resonance process and the energy of the neutrons can be reduced by a factor of 10 to 20. The nucleus is left in an excited state and, upon its return to the unexcited state, a gamma ray of energy equal to that lost by the neutron is emitted. Thus, we have a slower neutron at the expense of a gamma ray. Elastic scattering also takes place, in which momentum is exchanged in proportion to the respective masses of the colliding nuclei. In case the masses are equal the energy of the neutron will be reduced by a factor of e on the average in each collision. After thermalization, the neutron will be absorbed by (n, γ) reaction in general. Some (n, α) reactions will occur.

The attempts made to date to calculate neutron attenuation have been unsuccessful. Measured relaxation lengths for various concretes and the concrete compositions are tabulated in Table I.

Actual Shielding Design

A practical shield design for a central station power plant involves a preliminary estimate of the fraction of neutrons and gamma rays that will enter the shield. Suppose we have a power reactor at 750,000 kw heat. This reactor will produce several billion curies of gamma rays at full power. However, the fraction that reaches the shield will depend on the physical size and composition of the reactor. The self-shielding within a power reactor using natural or slightly enriched uranium will be large, and only a few per cent of the gammas and neutrons will reach the thermal shield. Perhaps, 5% of the gammas and 2% of the neutrons will reach the shield. At high power densities the amounts are very large.

Table I

MEASURED NEUTRON RELAXATION LENGTHS AND COMPOSITIONS OF CEMENTS AND CONCRETES

Material	Density gm/cm ³	Measured Relaxation Length, cm	Elemental Components, mols/liter													Total
			Fe	H	B	O	Mg	Cl	Ca	Ba	Si	Al	Mn	S	C	
Portland Concrete	2.3	11.1	0.13	4.8		71.8	0.2		14.5		15.6	0.44		0.06	10.8	118.3
Boron Cement	2.0	7.6		79.1	7.98	68.2	16.42	6.93	2.66							181.3
Brookhaven Concrete	4.3	6.3	54.7	21.1		47.7	0.35		6.45		2.10	0.79	1.22	0.11		134.5
W-1 Concrete	3.6	6.6	40.5	20.1	3.28	38.8	0.62		12.15		3.65	1.36	0.21	0.18		120.8
Barytes Concrete	3.5	8.0	0.11	5.17		63.5	0.2		3.4	11.9	4.1	0.4		12.0		100.8

Portland Concrete: 4 parts limestone gravel, 2 parts river sand, 1 part Portland Cement

Boron Cement: 1 part MgO, 1 part Colemanite, gauged with saturated MgCl₂

Brookhaven and W-1 Concretes: Portland Cement and iron aggregate

Barytes Concrete: BaSO₄ aggregate and Portland Cement

For 750,000 kw of heat, there will be about $3 \times 10^{13} \times 750,000 = 2.3 \times 10^{19}$ fissions per second. Each fission produces 2.5 neutrons and perhaps 5 gammas including the radioactive decay. The neutron flux at the edge of the core will be 10^{12} to 10^{13} n/cm² sec and the gamma flux 1 to 2×10^{13} gammas/cm² sec.

The shield must do several things. It must thermalize the fast neutrons, absorb the thermal neutrons, soften the gamma rays, and absorb the soft gamma rays. Since absorption of a neutron may result in the emission of a hard gamma, these processes are really opposites. To attenuate the neutrons we need lightweight elements, preferably hydrogenous materials, and to attenuate the gamma rays we need heavy elements. Therefore, all shields are compromises.

Other requirements on shields vary with the use. For mobile reactors lightness and compactness are important, and for stationary power plants cheapness and compactness are important. Since concrete, water, steel, and wood are our cheapest materials, power reactor shields will be made from these materials with minor additions of other materials.

Concrete is rather dense, cheap, and contains considerable amounts of hydrogen. However, it will not stand great thermal shock or high temperatures. For this reason, we will probably use so-called thermal shields of iron-water inside of concrete biological shields.

Ideally, a shield should contain elements with high inelastic scattering cross sections, light elements for elastic scattering, elements with high thermal neutron cross section, the material should be high density to absorb gammas, and the whole thing should be cheap and occupy little volume. The various materials that we may seriously consider are:

Hydrogen - High elastic scattering

Iron - Fair absorption; fair density

Boron - High absorption without gamma emission

Concretes - Various concretes with additions of colemanite or dense aggregates

Lead - High density; excellent inelastic scattering; expensive

Thorium and Uranium - Can be used indirectly to reduce the gammas in such reactors as breeders.

Actual Shield Design

In actual shield design for a power reactor there are two methods that may be used. The first of these is based on actual calculations of a water shield. Neutron collisions are treated as absorptions and a buildup factor applied later to account for the scattered component. Gamma-ray attenuation is calculated on the basis of a simplified picture which leads to a linear buildup factor. Corrections are then applied for the secondary gamma rays.

In the second method of calculation the data from a full-scale experiment are transformed to the geometry of the reactor to be shielded so that a direct comparison in dose for a given power is available. Frequently this process requires new experimental data.

Without going into the details of reactor design, we may use comparative data from various reactor experiments and estimate the thickness of the thermal shield and biological shields, and from the thickness, the costs. The attenuation data are reasonably accurate if adequate buildup factors are added.

Suppose we have 10^{13} neutron flux and 2×10^{13} gamma flux at the inner edge of a shield. Four to six inches of iron and six to eight inches of water will reduce the gamma and neutron fluxes by a factor of 10. Thus, to reduce the neutron and gamma fluxes to, say, 10^9 will require 15 inches of iron and two feet of water. About 13 inches of concrete will reduce the neutron and gamma fluxes by a factor of 10. This includes the necessary buildup factors and allows for thermalization and absorption of the fast neutrons. For a reduction from 10^9 to 10^2 a concrete shield of 8 feet will be required. These figures can be used for preliminary cost estimates.

In power reactor shields there will be few openings and no experimental holes so the shield design is relatively straightforward.

Experimental Shielding Facilities

Experimental facilities which have been used extensively for shield development have been the Lid Tank and Bulk Shielding Facility at Oak Ridge. The Lid Tank consists of a large water tank placed in contact with a large hole, about 2 ft square, in the shield of the Oak Ridge graphite reactor. The tank is filled with water. Tests are conducted by placing various shielding materials in the tank and noting the attenuation.

The Bulk Shielding Facility consists of a highly enriched reactor in a large tank of water known as the Swimming Pool Reactor. The reactor consists of MTR type fuel elements mounted on a movable frame - all under

some 20 ft of water. Various shielding materials are set up and the reactor moved into place against the shielding materials for a check of attenuation. Attenuation curves for water obtained with this reactor are presented in Figure 1.

The top of the BNL reactor has also been used for shielding tests.

Attenuation Curves

The data obtained in shielding experiments are normally reported as curves similar to the attenuation curves shown in Figure 1.

Heavy Concretes

Various types of heavy concrete, as indicated in Table I, have been developed. These materials require special handling in placement because of their high density. The MTR shield was made of barytes concrete which was placed by the Prepakt Method. In this method the heavy aggregate is placed dry and then flooded with water. The light aggregate and cement are pumped into the bottom of the heavy aggregate and displace the water.

Mixed Shields

An example of attenuation in an actual shield is presented in Figure 2.

Cost of Shielding

The cost of ordinary concrete shielding is about \$200/yd³ installed. Steel for the thermal shield will cost perhaps \$75/ton installed.

Heating of Shielding

A thermal neutron current of $1 \times 10^{13}/\text{cm}^2 \text{ sec}$ corresponds to a heating of 5.6 watts/cm². If a boral coating is used on the inside of the thermal shield, we may assume something like 10 to 20% of the neutrons will be fast so that we will get a thermal flux drop to 10^8 to 10^9 which will buildup in the thermal shield to 10^{10} or higher outside of the boral sheet.

Gamma heating in the thermal shield can readily be calculated from the flux and attenuation. It will vary from something like 0.005 to 0.0001 watt/cm³ as we go outwards for 15 cm from the inside of the thermal shield.

Figure 1 - Bulk Shielding Facility Pure Water Data Work Sheet

Figure 2 - Flux Distribution through Moderator and Shield of MTR

LECTURE 20

SEALS, PUMPS, HEAT EXCHANGERSReferences:

- "Liquid Metals Handbook," R. N. Lyon, Navexos P-733, Supt. of Doc.
- "Nuclear Power Plants - Design and Performance of Liquid Metal Heat Exchangers and Steam Generators," R. D. Brooks, A. L. Rosenblatt, Mech. Eng., May 1953, p. 363.
- "The Once-Through Boiler in Nuclear Power Systems," J. P. Silvers, Paper to be presented at Univ. of Cal. at Los Angeles Conference, April, 1955.

Introduction

Specialized equipment is needed for several purposes in nuclear power plants. This equipment includes pressure vessels, pumps, heat exchangers, valves, seals, and occasionally special piping. The special instruments and reactor control elements are not discussed here. There are a number of reasons for use of special equipment as follows:

1. The leak tightness of equipment handling radioactive coolants or solutions should be much higher than that required for cooling water in regular steam plants. Leaks permit the spread of radioactivity, as well as possible loss of D_2O or other valuable solutions. A D_2O leak of $1 \text{ cm}^3/\text{min}$ (liquid) will cost about \$60,000/yr. The definition of tight as used in the nuclear energy field means that absolutely no leakage occurs. A tight air duct means no air can leak out and tests are usually conducted by He leak detectors.
2. Maintenance and repair of equipment during operation is impossible and difficult even with the reactor shut-down. While the N^{16} in water decays rapidly, corrosion products retain their radioactivity for a long period of time. Na (15-hr half-life) decays slowly and must be removed from the system before any maintenance or repair work can be done. Na may be removed by alcohol or steaming followed by a water wash.
3. Hydrocarbon lubricants must be avoided because they form a film on heat transfer surfaces or become carbonized due to decomposition by radiation. This results in lowered heat transfer rates. In addition, carbon may form CO_2 and cause increased corrosion by its effect on pH. This is not serious in deionized water.

4. Liquid metals have special problems. For example, Na has a high vapor pressure which can cause trouble due to freezing out of solid Na.

5. Special alloys may be required in order that we may have suitably low corrosion rates.

6. Nitrogen will react with water and oxygen to form nitric acid.

The coolants usually considered are H_2O , D_2O , and Na for heterogeneous reactors and solutions of salts, acids, bases, fused chlorides, and liquid metals for homogeneous reactors. Water and Na systems are discussed below with some information concerning homogeneous systems.

Water Coolants

Water used in reactors is usually deionized, except for the Hanford and NRX reactors in which the water is used on a once-through basis. This is discussed under coolant maintenance.

Piping systems are ordinarily designed differently for reactor coolants than in normal industrial practice. For example, all piping for H_2O must be shielded. So, it is run under ground or in shielded ducts. These are costly and the minimum pipe lengths above ground are used. With D_2O the water cost is so great that the minimum pipe diameters, as well as lengths, are used. In addition, the maintenance is very difficult.

Valves and Pumps

Valves and pumps are normally designed with the operating mechanism placed outside of the active system. A packing gland or seal is used to prevent leakage along the pump shaft or valve stem. In normal steam and water practice the loss of several pounds of water or steam per day is of little consequence. Any leakage of D_2O through a seal must be recovered by evaporation into dry air and subsequent refrigeration and drying.

For radioactive materials there are two general methods of approach - enclosed drive mechanisms and double seals. In the enclosed drive mechanism for a pump, an induction motor is used and a thin can is placed between the starter magnets and the rotor. In such a pump the water fills the space around the rotor and floods the bearings so that the bearings must be designed for water lubrication. This means that the bearing loads must be a small fraction of ordinary hydrocarbon lubricated bearings or, inversely, the bearings must be very large. Westinghouse Electric Company makes such pumps. Allis-Chalmers' engineers have suggested a "float" journal in which the shaft is kept from contact with the journal by high pressure water or liquid metal streams.

In the canned pump the inner can is made very thin. An outer heavy can is placed over the windings to prevent a major loss of coolant in case of failure of the inner can. The space around the windings may be oil or gas filled. These pumps are expensive and not yet as reliable as standard pumps.

In the double seal type pump, high pressure deionized water is introduced between the seals so that any leaks are into the system. This works fine for H_2O systems, but not for D_2O systems. In the D_2O systems a vacuum is placed between the seals so D_2O is recovered.

Valves are used as sparingly as possible. Instead of a valve, extra piping may be used so that a heat exchanger may be drained and removed without draining the reactor tank. Where valves are used, they may be capped with welded bonnets during normal operation or bellows may be used.

In general, seal materials should be of the metallic type.

Whenever a mechanism such as a control rod drive must be placed in a water reactor system, it must depend on water lubrication. The mechanism must be corrosion resistant. In almost all cases the high Co materials like Stellite 3 have been found superior in wear resistance to other materials, such as Martensitic steel, in tests up to $440^\circ C$. Precipitation-hardened stainless steels and other Co materials, such as Haynes 25, are used for light load applications where ductility is important. These materials can be operated in water at only 1 to 2% of their design loads. This statement holds for gears, ball bearings, ball bearing lead screws, and in standard wear tests. Some of the typical test results are presented in Table I.

In the case of boiling reactors the problems of tightness are expanded to include the turbine and condenser, but the heat exchangers are eliminated. The leakage from the turbine probably would not be serious for H_2O systems, since there is little reactivity carry over from the reactor other than the N^{16} . In the case of the D_2O system the turbine must either be made leakproof or the leakage must be recovered. Designs have been suggested that permit covering the bolts and side splits of the turbine and recovering the D_2O leakage. The usual turbine shaft seal is a labyrinth. For a D_2O -driven turbine an improved shaft seal would be required. A labyrinth with more passages and a vacuum or pressure at the midpoint should be adequate. Care must be exercised to prevent leakage of H_2O in, as well as of D_2O out.

Heat exchangers, boilers, and condensers can be designed to be leakproof. Careful design and construction would improve commercial items, but double headers, rolled at the inner and welded at the outer tube sheet, and even double tubes have been suggested. Such systems may not be required, but certainly careful workmanship and frequent inspections will be required. Cored tubes may be used in heat exchangers to cut down the value of D_2O present.

TYPICAL PERFORMANCE OF WEARING PARTS USING WATER LUBRICATIONBEARINGS

<u>Races</u>	<u>Material Balls</u>	<u>Retainer</u>	<u>Manufacturer Size</u>	<u>Number of Revolutions</u>	<u>Test Load</u>	<u>Manufacturer Rated Load</u>	<u>Per Cent of Rated Load</u>
Stellite 3	Stellite 3	"W" SS, H.T.	3304	70,000,000	30# radial	1350	2.2
"W" SS, H.T.	Stellite 3	416 SS, H.T.		70,000,000	30# radial	2150	1.4
Stellite 6 inlay	Stellite 3	Nylon		8,000,000	50# thrust	2150	2.3
Stellite 3	7/7 Stellite 3	None*	20208	14,600,000	76# thrust	4390	1.73

NOTE: Per cent of rated load = test load/rated load (from New Departure Handbook)

GEAR TESTS

<u>Gear Material</u>	<u>Speed Run, rpm</u>	<u>Temperature Applied, F</u>	<u>A Oil Lube** Load Rating, lb</u>	<u>B Applied Test Load, lb</u>	<u>Per cent B of A</u>	<u>Remarks</u>
"W" SS, Malc.	108	450	1000	16-20	1.6-2	After seven million revolutions teeth show good polish with light pitting in some cases. Very little tooth wear.
Stellite 3	100	450	1000	20	2	After seven million revolutions teeth are polished with light radial scoring. - Little wear.

NOTE: All gears - 1" face, 3/16" pitch, 2-15/16" pitch diameter, 30 teeth.

*Spacer ball 7/7

**Using Lewis formula $W = F S_0 y \frac{c}{p}$ where F = active face width; S_0 = stress (assumed 30,000 psi for heat treated gear steel) y = form factor; c = velocity factor (used 1 for low speed); p = diametral pitch

More compact and improved designs of heat exchangers and boilers probably will be developed. These probably will be much tighter and cheaper than present types. In the case of condensers the first tubes are frequently injured by water carried by the high velocity steam. This can be corrected by design. The quality of the steam produced in some types of reactor systems will be low and no reheat is practicable. In Na-cooled reactor systems, the high temperature Na could be used both for superheating and reheating.

The metals which can be used in water-cooled reactors are Al, Zr, and the steels. Recent tests with Al indicate that it can be used up to 230°C or more. Zr alloys can be used up to 350°C and stainless steel somewhat higher. To date stainless steel has been used as the external system with both Al and Zr. However, at the higher temperatures stainless steel has appreciable corrosion. It may turn out that Al can be used at quite high temperatures provided the entire system is Al so that corrosion inhibitors can be used.

In D_2O piping, if flanged joints are used, electric drip detectors should be used to notify the operators of gasket failures.

Na Coolant

With Na coolant higher temperatures can be used provided the entire system is kept clean by elimination of O_2 and H_2O . Here the problem is not only to keep the Na from leaking outwards, but also to keep air and water vapor from leaking into the system. He or A can be used as blanketing gas. Ordinary grades of gases are not pure enough for use. Lamp grade A, double charcoal purified He is satisfactory. Passing the gas through Na in a packed tower at 750°F , purifies the gas sufficiently for use.

All piping or parts of the system in which Na may freeze must be provided with electrical heating pads. All joints should be pressure-welded to code so that there are no cracks in which cleaner or Na may be held when the system is drained.

Na has an appreciable vapor pressure. It will condense on any cold surfaces, and if it freezes it may cause difficulties in operating equipment.

Before putting Na into a system the system should be completely flushed, degreased, and dried. The system should then be leak tested and filled with inert gas before use. Insofar as possible the system should be designed for complete draining and for the minimum amounts of gas pockets. Inert gas under pressure should be used to force the Na into the system. A stainless steel micrometallic filter should be used in the fill line. While a dump tank is advisable in most industrial systems, this should be avoided

in the case of the reactor since the fuel elements should be kept covered with the metal at all times. Means should be provided for transfer of the Na from the reactor to a storage tank in case major repairs are needed and provided the reactor is unloaded or the decay heat is low enough for gas cooling of the fuel elements.

Na may be kept clean by filtering cold through steel wool, since the oxide is less soluble in the cold than when hot.

Pumps

Canned motor pumps can be used for Na. However, wear and galling of the bearings has resulted in continuous trouble. Standard centrifugal pumps may be used if the pump casing is enclosed in a He tank and leakage is taken care of while the air is kept out by a pressurized seal. Care in design is necessary to avoid oil contamination.

Electromagnetic pumps have been developed and used very successfully. Their efficiency is less than that of centrifugal pumps in that they range around 20 to 60% while centrifugal pumps may go up to 80% or more. The efficiency of the electromagnetic pumps is climbing rapidly with improved designs.

The great advantage of the electromagnetic pump is its lack of moving parts. For this reason the pipe containing the Na may be welded completely since there are no seals or bearings operating in the Na, or the pump may be immersed in a tank of liquid Na using slip joints.

The Na is usually carried through the pump in thin-walled tubing. Cavitation in this tube must be kept to zero. Since Na will form vortices readily, it is necessary to keep this down and also eliminate cavitation by a static head at least 2.5 times the velocity head in the pump. Gas entrainment in the pump causes very great increases in resistance and loss of pumping.

Three types of pumps have been developed: (1) linear, AC, induction pump; (2) helical flow, induction pump; and (3) the DC Faraday pump. The characteristics of some of the pumps built to date are presented in Table II.

In the design of the Faraday pump a current flows through the Na perpendicular to the line of flow and this section of the pump is held in a strong magnetic field. The principles are similar to a motor design. Either AC or DC may be used. Since low voltages are used with high amperage, DC is usually used. The DC can be generated by use of a homopolar generator, usually at 1 volt or so. In this pump electrical insulation is obtained by use of stainless steel bellows as a resistance.

Table II

ELECTROMAGNETIC PUMPS

Characteristic	Linear induction	Linear induction	Round induction	DC Faraday	Centrifugal
Builder	General Electric	General Electric	General Electric	Argonne National Laboratory	Allis-Chalmers
Location	KAPL	KAPL	KAPL	ARCO, EBR	Milwaukee
Capacity, gpm	1400	2500	400	500	275
Head, ft	100	100	54	50	55
Speed, rpm	---	---	---	---	1600
Power requirements	350 v, 60 cps, 3 phase, 80 kw	440 v, 60 cps, 3 phase, 130 kw	200 v, 25 cps, 3 phase, 28 kw	0.8 v, 18,000 amp, DC	25-hp drive, field excitation 30 amp
Dimensions	8.5 ft long 37 in. wide 24 in. high	7 ft 8 in. long 37 in. wide 15.5 in. high	38 in. long 25 in. diameter	---	---
Weight, lb	6430	3900	---	---	---
NPSH,* psia	20	21	---	---	---
Cooling	Forced air, 2000 cfm	Forced water, 50 gpm	Forced air, 1000 cfm	None	Air
Insulation	Class B	Class H	Class B	Stainless Steel	Class B
Thermal Insulation	Phosphoasbestos	Stainless Steel	Phosphoasbestos	None	---
Control of flow	Vary voltage	Vary voltage	Vary voltage	Vary current	Vary speed or excitation
Efficiency	37% Na	37% Na	20%	40% NaK	17% Na
Power factor	50%	35%	80%	---	---

*Net positive suction head.

In the linear induction pump a multipole field travels axially down the fluid duct. The field is produced by polyphase AC windings located on two sides of the duct. Since the windings must be near the hot metal, they must be air or water-cooled.

Valves

Valves for use with liquid metals are usually special in respect to valve seat and stem packing. Ta or W is used on the seat facing and a bellows is used as a seal of the valve stem.

Instruments

The development of special instruments for measuring flow rates, etc., is of interest. The high vapor pressure of Na causes clogging of many conventional instruments in that the Na will vaporize into the lines and freeze.

Pressure may be measured by use of a double diaphragm pressure transmitter which operates a bleed line on a constant volume high pressure air source. The pressure is read by a gage on the inlet air line.

Flow may be measured by magnetic flowmeters which induce current and are very similar in design to magnetic pumps. Difficulties with variations in materials are encountered and local emf's have been generated. Wetting takes time. Electropolishing decreases wetting time. Once the surface is wet by the Na it apparently retains its ability to be wet.

Heat Exchangers

Several applications of heat exchangers are apparent in the nuclear energy field. These include the following:

Water - Water
Water - Boiling Water
Na - NaK
NaK - Boiling Water

The design of these heat exchangers follows conventional practice in most respects but special requirements are added. The most obvious requirements are leak tightness, reliability, and compactness. Design work carried out to date has pointed up most of the questions and have led to improvements in the design of heat exchangers, but much remains to be done in respect to compactness, cheapness, and simplicity.

Suggestions for improvements in the conventional tube type heat exchanger have included the use of double headers, welded tube ends, double tubes with a circulating liquid between the tubes to indicate leaks, cored tubes to reduce volume, long rigidly held tubes, and the use of fabricated pressed corrugated plates rather than tubes.

The actual requirements will vary for the different reactor systems. For example, in a pressurized H_2O reactor the interchanger will be in reality a boiler heated with high pressure water. The reactor water will be highly radioactive due to its N^{16} , but will contain very small amounts of other impurities except perhaps 100 psi H_2 to react with any oxygen formed by decomposition of the water. This water will not be particularly corrosive and its radioactivity will be low a few minutes after a heat exchanger is removed from service. A conventional heat exchange boiler could be used because leaks would not be serious nor difficult to repair.

In a pressurized D_2O reactor, the heat exchanger would be more rigidly designed. The leak tightness requirement would be more important and the volume of the D_2O side would be kept to a minimum. In such a heat exchanger two headers might be used with the tubes rolled into the inner header and welded into the outer header. Dry air or He would be circulated between the headers to pick up any D_2O that might leak for recovery. In such a heat exchanger, a decision must be made whether the D_2O or boiling H_2O side is at highest pressure. H_2O might dilute the D_2O moderator or D_2O might be lost.

In the Na-NaK heat exchanger the Na will always be at a higher pressure than the NaK and a check made for Na radioactivity in the NaK. In the NaK to boiling water exchanger concentric tubes might be used with Hg or diphenyl pumped between them. One way is to use concentric tubes which have slots milled in the outside of the inner tube. Hg is pumped through these slots to locate leaks or simply a gas such as air may be pumped through.

Brooks and Rosenblatt present results of tests made on forced circulation and natural circulation water boilers. In both units, Hg was used to separate the double-walled tubes. Since the steam side film resistance is less than the tube or NaK side, the heat transfer was similar in the two units. The tube resistance was, of course, the greatest. Their results for boiling and superheating are presented in Figures 1 and 2. Their results for the over-all heat transfer coefficients for flat and round tube Na to NaK heat exchangers are presented in Figures 3 and 4.

Silvers discusses the use of once-through boilers with their advantages and disadvantages and presents results of calculations.

Figure 1 OVER-ALL HEAT-TRANSFER COEFFICIENTS OF SODIUM TO NaK FLAT-TUBE HEAT EXCHANGER

Figure 2 OVER-ALL HEAT-TRANSFER COEFFICIENTS OF SODIUM TO NaK SHELL-AND-TUBE HEAT EXCHANGER

Figure 3 OVER-ALL HEAT-TRANSFER COEFFICIENTS OF NaK TO WATER EVAPORATORS

Figure 4 OVER-ALL HEAT-TRANSFER COEFFICIENTS OF NaK TO STEAM SUPERHEATERS

LECTURE 21

LOADING, UNLOADING, COOLING AFTER SHUTDOWNReference:

"Heat Generation in Irradiated Uranium," S. Untermeyer, J. T. Weills, ANL-4790

Loading and Unloading

The early graphite reactors were designed for through-hole refueling. This was used at the X-10 and has been followed in the design of the Brookhaven reactor. Other heterogeneous reactors have utilized variations of the single end loading system. In the graphite reactors the fuel channels have been horizontal while in almost all other reactor designs, the fuel elements have been vertical. In recent years the attempt has been made in design to incorporate a larger amount of fissionable material in a fuel element and to handle this fuel element or assembly by crane.

The X-10 and Hanford fuel elements are slugs 4 or 8 in. long. The Hanford slugs are 1.360 in. in diam which gives a canned slug of somewhat larger diam and about 4-1/2 or 8-1/2 in. long. These are loaded singly by hand.

The front face of the Oak Ridge reactor is provided with shielding and an elevator that carries the operators to any cooling tube on the reactor face. To discharge and recharge a tube, the operators remove the front plug. Then the operators put in new slugs one by one and push the contents of the tube to the rear. This is done until the entire tube has been discharged. The irradiated slugs fall out of the tube on to a rubber mat and roll into the canal. The plug is then replaced.

In those reactors in which the fuel assemblies are fed in and removed through the top of the reactor, the handling equipment becomes complicated and expensive. There are several designs that have been developed for protection against the radioactive fuel elements during discharge. These are: (1) the use of a rotating eccentric top shield and a coffin; (2) the use of large cave rooms over the reactor; (3) for H_2O reactors the fuel elements may be pulled out into a deep well of water over the reactor. Most designs of power reactors for D_2O and Na coolants make use of the coffin principle, while H_2O reactors are mostly designed along the canal principle.

The MTR differs from other H_2O reactor designs in that the canal is below the reactor and the fuel assemblies are discharged through a pipe which has suitable valves. In this case the depth over the fuel elements is great

enough to give protection to personnel handling single fuel elements. The sequence of operations follows: (1) remove the top plug with the control drives by crane; (2) remove the upper grids by hand tools and light crane; (3) remove a section of the Be reflector over the discharge chute and store with the upper grids over the Be; (4) pick up a fuel element or shim control rod by hand and drop down the discharge chute; (5) feed in a new fuel element through the top and position in the lattice. The discharge chute is fitted with a hydraulic gate valve. The lower end of the discharge chute connects with and is sealed by a rotating tube that accepts the fuel element, rotates to a horizontal position, and discharges the fuel element in the bottom of the canal. The fuel elements are picked up and stored in a vertical position.

In some designs of proposed reactors the top shield is rotated until the discharge hole is over the fuel element to be discharged. A coffin is then placed over the hole and the fuel element withdrawn into the coffin. A coffin door is closed and the coffin removed for discharge of the fuel element to the canal. It is cumbersome, slow, and rather dangerous in that a fuel element might get stuck when partially removed. The possibility of a leak of radioactive material is also present - especially with Na-cooled reactors. An experimental full-scale setup of the unloading mechanism, less shielding, has been built and tested for the SGR.

In general, the methods suggested for loading and unloading power reactors have been complicated and would be difficult to keep functioning over many years. Pressurized water-cooled reactors may be considered in two groups: (1) those in which the core is placed in a pressure vessel and the top of the pressure vessel removed for loading and unloading, and (2) those in which the pressure vessel is pressurized independently of the tubes. In this latter case, the tubes are depressurized and reloaded one at a time. The advantage of the independent tubes is that there is no large opening that must be frequently unsealed and resealed, but this must be weighed against the disadvantages of the increased complexity of construction and maintenance.

A number of designs have been suggested for loading and unloading by means of various semi-automatic devices that reach into the reactor and move the fuel elements by means of grapples on the ends of folding arms. The locations of the grapples are indicated by various mechanical indexing devices.

If we were to attempt to set up a set of criteria for design of loading and unloading equipment, we might include the following considerations:

- a. There should be the minimum number of moving parts that can be contaminated with the coolant.

- b. The fuel elements should be designed with seats or bearings with angles of perhaps 20° so that they cannot bind.
- c. The fuel element cap to which the grapple is attached should extend above the coolant during the unloading operations if the coolant is opaque such as Na.
- d. The unloading should be simply a movement in the vertical direction, then horizontally, and finally vertically. These are the simplest motions that appear possible.
- e. The active portion of the fuel element should not be removed from the coolant during unloading.

The above criteria are somewhat mutually exclusive and no completely satisfactory procedure has been developed. The MTR procedure is perhaps the simplest that could be applied to large scale power reactors, but even this does not meet all our criteria.

Cooling After Shutdown

The heat production due to the radioactive decay of the fission products introduces a problem peculiar to nuclear reactors, i.e., the continued production of heat after the reactor has been shut down. This heat must be removed continuously or there will be danger of the fuel elements melting with the release of gaseous fission products. The heat production is given by the equation:

$$P = 0.08t^{-0.2}P_0$$

where P_0 is the heat production during operations and P is the shutdown heat. For the MTR the equation will be, in kw,

$$P = 0.08 \times 30,000t^{-0.2} = 2400t^{-0.2}$$

The maximum actual heat production per square foot of fuel element in the MTR during operation is 540,000 BTU/ft²hr.

On the assumption that the MTR has been in operation for a sufficient period to build up fission products, the heat production for various times after shutdown will be:

Time	BTU/ft ² hr	%	
-1 sec	540,000	---	100
1 sec	43,200	100	8.0
10 sec	28,800	66.5	5.3
1 min, 60 sec	19,000	44.0	3.5
1 hr, 3600 sec	8,400	19.4	1.5
1 day, 816 x 10 ⁴	4,500	10.4	0.83
1 week, 6.0 x 10 ⁵	3,000	6.9	0.56
1 month, 2.6 x 10 ⁶	2,200	5.1	0.41
1 year, 3.2 x 10 ⁷	1,400	3.2	0.26
10 years, 3.2 x 10 ⁸	860	2.0	0.16

The equation developed by Untermeyer and Weills for natural U reactors is:

$$\frac{P_s}{P_o} = 0.1 \left[(T_s + 10)^{-0.2} - 0.87(T_s + 2 \times 10^7)^{-0.2} \right] -$$

$$0.1 \left[(T_s + T_o + 10)^{-0.2} - 0.87(T_s + T_o + 2 \times 10^7)^{-0.2} \right]$$

The MTR fuel elements must be stored under water for about two weeks. After this time they can be stored in air and depend on free convection cooling. The actual cooling rates will depend on the geometry of the fuel elements, the flux, and the time of operation. For short operational times there will be a much faster decay rate, because the long life fission products will be present in such small amounts that they will not add significantly to the reactivity. The equation given is not very accurate for times over a month.

LECTURE 22

OPERATIONAL PROBLEMSReference:

"Summary Report of the Reactor Safeguard Committee,"
WASH-3, no date

Introduction

The operational problems involved with nuclear reactors are considered throughout the design, and many features of the operations are fixed by the designer in the choice of reactor type, method of maintenance, etc. For example, provisions for removing and cleaning up pumps and heat exchangers must be built into the reactor system. Other problems such as the hazards involved with a reactor are considered early in the design. The true operational problems which are met during daily operation have to do with the shipment, storage, handling, etc., of the fuel elements; loading and unloading, cleanup of radioactivity, maintenance and repair, flattening of the reactor during operating, maintenance and checking of the control system, and correction for long term reactivity changes.

Reactor Hazards

The hazards involved with the production, control, and storage of the large amounts of radioactive fission products were recognized as soon as fission type reactors were considered. It was for this reason, as much as for security reasons, that Oak Ridge, Los Alamos, and Hanford were located in isolated areas and such large tracts of land around these facilities were brought under security control.

Soon after the Atomic Energy Commission took over the Project, the General Advisory Committee recommended the creation of a Reactor Safeguard Committee. Such a committee was formed and held its first meeting on November 7, 1947. The Committee was requested to review the design of each proposed reactor and make appropriate recommendations to the Commission. The Committee has been changed slightly in the past few years: its title has been changed, but its work has been essentially unchanged.

The Committee meets from time to time to consider proposals for various reactors or for changes in power level, etc. The Committee functions by review of design reports submitted to it. In addition, it may take up problems related to hazards and request specific research or development work.

The greatest danger in the normal operations of a reactor system comes from the chemical plants and the storage of the fission products. During solution of the irradiated U, the gaseous fission products escape. These include Xe, Kr, and sometimes I_2 . The great danger inherent in a nuclear reactor itself is the possible release of the fission products due to a catastrophe or to a series of accidents that result in the loss of fission products.

Perhaps the three most important factors to consider in the safety of a reactor are the temperature coefficient, the loss of coolant, and the design of the control system. Every effort should be made to design and build only those reactors that have strongly negative temperature coefficients. With care, tank-type reactors can be designed so the loss of coolant is practically impossible. A gas-tight building will retain any gases emitted during a power surge such as the NRX incident. The design of the control systems can be carried out so that they are essentially fool-proof and, with strongly negative temperature coefficients, will prevent power surges.

The Reactor Safeguard Committee has listed those things which must be considered in order to permit an adequate safety appraisal of a proposed reactor:

1. Description of the reactor.
2. Detailed plan of the normal operation.
3. Plan of the experiments which will be performed on the reactor, including the limitations which will be placed on experiments.
4. The normal schedule of chemical processing and disposal of reactor products.
5. The description of the safety mechanisms of the reactor.
6. A discussion of the special hazards connected with the reactor, including the evaluation of the possibility of a major catastrophe.
7. The distribution of population near the proposed site.
8. A list of the vital industrial or war installations near the proposed location.
9. Frequency of occurrence of wind directions and velocities both at the surface and aloft; frequency of precipitation and its correlation with wind directions; and information as to the frequency of stable and turbulent wind conditions.

10. The occurrence of inversion layers and precipitation and their correlation with wind directions.
11. The methods proposed for the control of stack gases and the possibilities of the correlation of reactor operation with meteorological conditions.
12. The normal methods of disposal of aqueous coolant or other solutions.
13. Hydrological data on the area, the expected drainage of liquids in the case of a major accident, and the distribution of activities in the drainage system under such conditions.
14. The evaluation of the probability of earthquakes and their intensities.
15. The evaluation of the probability of an ordinary fire.
16. Discussion of hazards and risks to surrounding population as influenced by meteorological, geological, and hydrological conditions.

Similar data must be presented with plans to build separations plants. With the above information for background, how do we proceed in designing a nuclear reactor for power? Obviously, we must design so that we cover all possibilities of hazards which could arise during normal operation. It is significant that the two accidental power surges for which we have detailed data occurred during special experiments and under conditions that some of the safety devices were not operating in their normal manner. Possibly a third set of safety devices which were fully automatic and not connected to the regular control system in any way would have greatly reduced the power levels in these accidental surges.

If we were to put forward a philosophy of design for civilian power reactors, what should we include? Some of the items that should be considered are:

1. Auto vs. Non-autocatalytic Reactors - It would appear worth while not to consider seriously any reactor type that cannot be designed so that it is non-autocatalytic. For example, the H₂O-cooled and moderated, graphite-moderated reactor may be either auto or non-autocatalytic. We should not consider a design for construction which is autocatalytic. The only exceptions to this rule are those reactors of an experimental nature built to determine if a given type is in fact auto or non-autocatalytic.

2. Temperature Coefficients - Only reactors having negative temperature coefficients should be considered for construction, except as noted above.

3. Fuel Element Size - The use of thin fuel plates or wires should be favored over heavy bars of fuel. The reason for this is that the heavy bars will melt or vaporize in a power surge before sufficient heat is transferred to the coolant to appreciably change the density of the coolant. High heat transfer coefficients would be beneficial. For example, if it is desired to use oxides, these may be subdivided into fine wires or mixed with metal to give higher heat transfer rates.

4. Self-Quenching - The H_2O and D_2O -cooled and moderated reactors can be built to be self-quenching by boiling, and thus inherently safe. While this is a tremendous advantage over other types of coolants, it is probably not necessary to restrict reactors to these coolants.

5. Control Systems - The control system for the reactor should consist of at least two, and preferably three, different systems. The actual fine control (running control rod) should be carried out by rapid response and high rate of movement of a control rod that can control less than $0.75\% \delta k/k$ and preferably not over $0.5\% \delta k/k$. Since 0.75% of the neutrons are delayed, this control rod cannot make the reactor prompt critical even if it is injected instantaneously. The large increments of control should be built into shim or shim safety rods which can be moved only at very low rates through induction motor driven gear trains. With such a system, an error on the part of the operator will not permit addition of large increments of power in short periods. With slow increases in power, the regular safety circuits based on rate of power rise or period meters and the regular control instruments can shut the reactor down by operating the safety circuits.

6. Third Safety Devices - Completely independent third safety devices should be built into the reactors to shut them down in case of power surges. Such devices are in a primitive state of development. Work is going forward and should be continued.

7. Reactor Tank - The reactor should be built in a tank or vessel so that the coolant will not drain from the fuel elements in case of tank failure. This means that the reactor should not be built as a boiler - above ground - but the reactor should be placed in a pit or at least in a heavy concrete structure. This is readily done and the shielding becomes the secondary tank. The use of dump tanks should be avoided, and the coolant should be removed from the reactor only by special pumps. If an operating room is required below the reactor, the volume should be small and the excess coolant over the fuel should exceed this volume.

8. Reactor Building - The reactor building should be designed to hold any gases that might be given off by a power surge. Some provision should be built into this building for removal of the gaseous fission products and particulate matter rapidly and completely in case there is a power surge. Dilution of reasonable amounts of gaseous fission products will permit their disposal. Xenon and argon have short lives, but krypton has a long life. Efficient filters will remove much of the solid particulate matter.

9. Cooling After Shutdown - Reactors which operate at low specific powers can be air-cooled after shutdown. Thin fuel elements would help this by increasing the surface area for cooling.

10. Total Amount of Fission Products - The reactor type which has the minimum amount of fission products present should be favored.

It has been demonstrated by a number of economic studies that several reactor types potentially can be developed to produce economical heat for electrical power. These reactors are:

1. H₂O-cooled and moderated, slightly enriched U or U²³³-Th.
2. D₂O-cooled and moderated, natural U.
3. Sodium-cooled, graphite-moderated reactor U²³³-Th or slightly enriched U.
4. Homogeneous, H₂O or D₂O solution of uranyl sulfate.
5. Fast Pu breeder.
6. Homogeneous, liquid metal Pu breeder.

A designer of a chemical plant, for example, chooses a process on the basis of economics and adds the necessary safety devices considered necessary after the design layouts are well along. In the case of the nuclear reactor, safety is nearly as important as economics. Fortunately, the two seem to go together in that the most economical types appear to be the safest.

In a power reactor we may have several hundred kilograms of Pu or U²³³ and a billion or more curies of fission products. Perhaps a fourth of the fission products will be gaseous.

The homogeneous reactor permits separation and separate storage of the gaseous fission products almost as rapidly as formed. In case of a power surge, the danger of spread of these products is greatly reduced. Incidentally, the xenon problem also disappears. Separation processes for the fission products from uranyl sulfate solutions show promise for continuous operation. If these processes are successful, the homogeneous reactor would become, perhaps, the safest of all. However, in breeder or U²³³ homogeneous reactors, alpha emitters would be present in large amounts.

Boiling experiments should be run on D₂O lattices and on both H₂O and D₂O at high pressures. Similar types of experiments should be run on all types of reactors.

When the reactor system has been chosen for design on the basis of economics and safety, the design can proceed on the basis of regular reactor design principles. The central design problem centers around the heat removal. This problem is intimately associated with the power production through the temperature of reactor operation. The temperature in turn is intimately connected with the materials of construction.

The choice of reactor type will fix the moderator, if any, the coolant, and whether the reactor is homogeneous or heterogeneous.

The initial design studies will serve to fix the velocity of coolant, the over-all temperature, and the size of the fuel elements. In addition, the clad material will be fixed. The specific power level in kw/liter will be settled early in the design along with the flux and the heat production per unit area of the fuel plates. Except for breeders, the amount of fuel needed for heat removal purposes will be larger than a critical mass so the surface of the fuel plates and the volume of the coolant will fix the design.

The maximum temperature of the fuel and the maximum temperature of the clad surface will limit the reactor power. These temperatures will be dependent upon the properties of the fuel and clad. For natural U the maximum fuel temperature is usually kept 50°C or so below the alpha-beta transition temperature of 660°C . There have been some tests at temperatures above 660°C . These indicate that there is some possibility of operating at a higher temperature - at least in the middle of the fuel elements.

The control systems of all reactors have been given care in design and are based on well thought out principles.

LECTURE 23

ECONOMIC CONSIDERATIONSReference:

"Cost Estimating Procedure for Evaluation Work," D. W. Altwein, E. D. Kane, N. B. Shumate, LRL-101, February, 1954.

Introduction

The purpose of this discussion is to outline the type of evaluations that can be made from an economic viewpoint as far as power reactors are concerned. Unfortunately, we cannot discuss actual costs of U^{235} , natural U, irradiation costs, or chemical processing costs. A series of prices has been established by the Commission for the period July 1, 1955, to July 1, 1962. This list covers the prices the Government will pay for certain items, inventory charges, and the prices that the Government will pay for production of some materials. This list of prices will be made available to a limited number of persons making reactor evaluations.

Studies of the types discussed here have been extremely valuable in pointing up what types of reactors show promise of eventual economic value and in pointing out the places in the designs that need especial attention. The studies have been extremely useful in aiding the Commission in its decisions.

In addition to studies made by the Commission, studies have been made by the Ferguson Company and by the North American Aviation groups at the request of the Division of Reactor Development. These studies were the first to point up the desirability of using light water-cooled and moderated and sodium-cooled, graphite-moderated reactors with slightly enriched uranium and with $Th-U^{233}$. Later cost studies made by Argonne, Massachusetts Institute of Technology, AEC, and California Research and Development Company, as well as the NAA studies, have confirmed the values of such things as the recycle of Pu in power reactors. The cost studies made by the industrial teams have also been most helpful in indicating what lines of reactor development are worth while. In fact, it is actually difficult for us to separate our thinking from the results of the economic studies.

The criticism has been made that the economic studies are not worthwhile since they are not accurate. Usually, the statement is made that the construction costs are too low. A recent check of such a study has been made in which detailed cost studies on a firm basis using bids, etc., was made. This study showed that the previous estimates were too high, but not as inaccurate as we all supposed they were. The industrial teams have all used bids for the equipment external to the reactor, perhaps 85% of the total.

One problem that always comes up in power reactor studies is the question of state of development. Should we assume that the known problems have been solved and an industry has been developed, or should we assume that the costs must be based on present technology? Can we recycle Pu and U²³³? We don't know how at present, but we are quite certain that satisfactory methods can be developed.

In carrying out cost studies on various reactor types there is no real reason why all details must be evaluated as the designs are in a very early and incomplete stage of development. There is real reason to keep the comparisons on the same bases.

Care should be used to differentiate estimated costs as used by an engineer for estimating purposes and actual operating costs. The estimated cost for a plant is the cost for which the engineer believes that it can be built and placed in operation.

Cost Studies

If we are making a comparative study between two types of reactors, it is permissible to drop out items which are equal for the two reactors, or we may assume quite arbitrary unit costs for these items. If we are attempting to determine actual costs or if we are comparing reactors with regular steam plants, we must consider all items which differ between the plants.

Startup and inventory costs must be treated carefully. All startup costs incurred until the reactor reaches the design power level may be added to capital costs. Plant amortization then starts at the end of the startup. Initial inventory for a reactor may be quite high. This includes source and fissionable materials and heavy water. In order to discourage large inventories, artificially high inventory charges have been applied in the past. We may break up cost studies into six categories:

1. Direct material costs
2. Direct labor costs
3. Indirect construction costs
4. Technical service costs
5. Contingencies
6. Labor and weather factors

1. The direct material costs include the costs of all equipment delivered to the site. If shop labor away from the site is required, it is included in the cost of the materials. Freight, taxes, and delivery charges are included, but scaffolding, etc., are not included. If freight and taxes are not known, 5% of base material charge is added for freight and 3% for taxes.

2. The direct labor costs are those costs associated with on-site fabrication, erection, and construction. Shop fabrication labor away from the site is a part of the direct material cost. Premium pay, travel allowances, insurance, premium for overtime are not included. These are included in the indirect construction costs.

3. Indirect construction costs include overhead and construction facility costs. The indirect costs as referred to percentages of direct labor costs may be somewhat as follows:

Temporary Construction Facilities

Temporary office, etc.	1%
Temporary utilities	1
Scaffolding	1
Equipment Rental	12
Total	<u>15%</u>

Overhead

Small tools	4%
Consumable supplies	5
Field office expense, supervision, office overhead, etc.	8
Material handling	1
Unallocated labor	2
Payroll taxes	15
Premium allowance for overtime, travel, etc., (travel allowance at NRTS has been \$3/day)	0
Fee or profit	25
Home office overhead supervision	10
Completion bond	3
Total	<u>73%</u>

Total Indirect Costs 88%

4. Technical service costs are the normal architectural and engineering costs. They include preparation of all plans, drawings, and specifications, soil investigations, engineering estimates, and cost control. Usually field inspection and supervision to insure that plans and specifications are followed are included. Preparation of as-built drawings is included. Research and development costs are excluded. Based on total construction costs, or the sum of the direct and indirect costs, technical services will cost about:

5% for conventional plants
 10% for reactors
 15% for completely new plants

Our work is generally 7.5% or so since we usually go beyond the usual manufacturer in preparing plans and specifications.

5. Contingencies are added to allow for inaccuracies in estimating. These are based on the sum of the design and construction costs. They vary from 5% for a conventional building or plant to 25% for unique designs for which no cost data are available. For nuclear reactor power plants about 15% would be adequate at present.

6. Labor and weather factors are sometimes added to account for differing costs for plants built in one locality by a contractor who bases his estimates on his home locality. For example, a Chicago contractor might deduct 10 and 5%, respectively, for work in the San Francisco or Los Angeles areas and even more for work in some of the Southern cities. Freight charges, however, might be higher.

A summary of the costs might be something like the following:

Direct material	60%
Direct labor	40%
Direct construction	100%
Indirect construction (88% of direct labor)	35%
Construction cost	135%
Technical services (10% of above)	13%
Design and construction cost	148%
Contingency (15% of above)	22%
Base cost	170%
Labor and weather factors	±

One problem that arises in nuclear power plants is that of charging the fuel and blanket materials. Perhaps the first fuel and blanket loading, as well as the startup costs, should be charged as investment costs. Subsequent fuel loadings would be charged as operating expenses.

Once a nuclear power plant has been built and is in operation, there are the annual charges and the operating charges to be made. The annual charges will be dependent on the estimated life of the plant as the plant must be written off in that time. Since no data are available on the life of nuclear reactors, we can only make estimates. Since the fuel elements are renewed frequently, the only things that can deteriorate are the reactor vessels, pumps, piping, etc. Since these approach regular process plant equipment in complexity and materials of construction, we may make reasonable lifetime estimates. There appears no reason why 20 years should not be a reasonable reactor life. The remainder of the plant, such as the power plant, can be assumed to have a normal life expectancy.

Another factor is the type of financing one talks about. We actually have three systems: government, public utility, and private industry. In government financing, interest rates are lower than for private industry, and no taxes are collected. In public utility finance, profits are limited by the utility commissions, but, in return, the markets are protected so that reasonable profits are quite certain. In private industry, there is always an urge to use money for several different purposes, and the private company will invest in that field which has the greatest potential profits.

The fixed charges on investment may be about 15% for utilities for depreciating items for nuclear plants. A charge of 12% is reasonable for non-depreciating items such as land, nuclear fuels (D_2O , Be, BeO), and source materials. The initial loading of a reactor is considered a non-depreciating item.

Land costs are usually small for public utilities, perhaps \$1 per kw of installed capacity. If we apply the Reactor Safeguard formula, the land costs would be very high. So, for the moment, we must assume that we can develop nuclear power plants that can be built near population centers. The enclosure of the reactor in a sealed building such as those used for CP-5 and the SIR reactors should permit construction of reactors near industrial centers. The added costs of these sealed buildings must be included in the cost analysis.

Example of Cost Calculation

We may assume the following costs of nuclear fuels and fissionable materials as approximate to indicate methods of making cost studies:

U^{235}	\$20/gm
D_2O	\$50/lb
Natural U as oxide	\$10/lb
Sodium	\$0.16/lb
Graphite	\$0.25/lb

Slightly enriched U costs are unknown. Since costs of making UF_6 , recovering the UF_6 from the separation plant, and producing the metal must be high, let us add \$10/lb to the cost of the U, and use a straight line enrichment cost from natural U of \$20/lb to \$9,000/lb.

Fabrication may be assumed: Graphite \$0.25/lb; natural or slightly enriched U \$5.00/lb; U^{235} or Pu^{239} in Al-clad rods \$1.00/gm; U^{235} or Pu^{239} in Zr-clad rods \$2.00/gm.

Recovery of Pu and U from natural or slightly enriched U may be assumed to cost \$10/lb of feed material and recovery of enriched U or Pu from Al or Zr rods may be assumed as \$1.00/gm. The value of Pu may be assumed equal to that of U^{235} .

Using the above data, we may calculate the value of recycling Pu in a light water-cooled and moderated reactor. Let us assume that we use 1.00% U^{235} in slightly enriched U or U to which Pu is added to give 1.00% fissionable material content as a fuel and that we burn 2 atoms/100 with an overall conversion ratio of 0.80.

We need not know the critical mass nor the power level of the reactor. We assume a basis and calculate the costs on this basis:

Basis: 2000 lb of feed

$$\text{Cost of feed per lb} = 20 + \frac{8980 \times 0.3}{99.3} = \$47/\text{lb or } \$94,000/\text{ton}$$

$$\text{Fabrication costs } 5 \times 2000 = \$10,000/\text{ton}$$

$$\text{Gm/ton } \text{U}^{235} \text{ feed: } 0.010 \times 2000 \times 454 = 9080$$

$$\text{U}^{235} + \text{Pu in reject fuel: } 9080 \times 0.80 = 7260 \text{ gms.}$$

This is an estimate only as the conversion ratio changes with time in the reactor. It will also be dependent upon the power level, etc. Fuel recovery costs/ton $2000 \times 10 = \$20,000$. About 20% of the reject fissionable material will be U^{235} or $7260 \times 0.20 = 1452$ gms, leaving 5810 gms of Pu.

Since we have no data on cost of making Pu fuel elements, let us assume the fabrication costs are double those for U, or \$10/lb. Then $10 \times 2000 = \$20,000/\text{ton}$.

The U^{235} added to natural U to enrich it to 1.00% is $0.00289 \times 2000 \times 454 = 2630$ gm.

There are several alternatives that may be followed. First, we may buy natural U and enrich to 1.00% U^{235} with fully enriched U^{235} or the U^{235} might be used in a spiked reactor; and second, we may re-enrich the rejected fuel by buying (or selling) Pu. These alternatives and the original conditions are discussed in Table I along with the conditions discussed above. For simplicity, it is assumed that the reactor has been operating long enough that rejected Pu is available.

Table I

COST OF FUEL PER TON FOR WATER COOLED REACTOR

<u>Basis 2000# Feed</u>	<u>Slightly Enriched U</u>	<u>Nat U + U^{235}</u>	<u>Re-Enriched Fuel</u>
Cost of Feed			
Natural U, 20 x 2000	-	40,000	-
Natural U Makeup 0.02 x 20 x 2000	-	-	800
Slightly Enriched U	94,000	-	-
Full Enriched U or Pu 2630 x 20	-	52,600	-
Buy (9080 - 5810 - 1452) = 1820 g Pu	-	-	36,400
Fabrication			
No Pu, 5 x 2000	10,000	10,000	-
With Pu 10 x 2000	-	-	20,000
Recovery Costs 2000 x 10	20,000	20,000	20,000
Sale Price for Pu, 5810 x 20	116,200	-	-
Excess Pu (5810 - 2630) 20	-	63,600	-
Net Cost/Ton of Fuel	7,800	59,000	77,200

The low cost of the slightly enriched U is due to the high burnout assumed and the fact that U^{235} in natural uranium oxides at \$10/lb of U is valued at only \$3.10/gm while we have valued the Pu at \$20/gm. Both U^{235} and Pu have a heat equivalent value of about \$16/gm which is equal to that of coal of 12,000 BTU/# with the coal cost of \$8.00/ton.

In the above calculations, we have two obvious errors. We have assumed that the reactivity of Pu^{239} equals that of U^{235} and that the Pu is all Pu^{239} . The second is that the value of Pu should be less than that of U^{235} by at least the increased fabrication costs.

Similar calculations might be made on the assumption that Pu has market values of 15, 10, and 5 dollars/gm. Different results would be obtained for lowered irradiation times and for different enrichments.

Using the above cost of fuel as \$8,000/ton, the fuel charge for electric power on the basis of 25% thermal efficiency may be calculated:

$$\text{Energy per gm of U}^{235} \text{ or Pu fissioned} = 2.16 \times 10^4 \text{ kw hr}$$

$$\text{Grams per ton} = 2000 \times 0.02 \times 454 = 18,160$$

$$\text{kwhr/ton electric} = 2.16 \times 10^4 \times 1.816 \times 10^4 \times 0.25 = 9.8 \times 10^7$$

$$\text{Cost/kwhr for fuel} = \frac{8.0 \times 10^6}{9.8 \times 10^7} = 0.08 \text{ mills}$$

This holds for all loadings after the first.

On inventory charges we may use the mean value between the feed and reject material:

$$\text{Feed cost} \quad \$95,000/\text{ton}$$

$$\text{Reject value} \quad \$116,200 - 20,000 = \$96,000$$

Reject value is considered value of Pu less fabrication costs.

Average inventory must be charged on 1.5 loadings since there would be, on the average, a half loading in the external processing plants including fuel elements awaiting charging.

We assume inventory charges at 12%/annum.

The inventory charge will vary with the rate of burnup. Let us assume 1 and 2 years:

Inventory cost

$$0.12 \times 1.5 \times 95,500 = \$17,200/\text{yr}$$

$$\text{Cost/kwhr} = \frac{1.7 \times 10^7}{9.8 \times 10^7} = 0.17 \text{ mills}$$

Thus for reloading each year the total cost of fuel would be 0.25 mill/kwhr and 0.42 mill/kwhr for two-year inventory charge. The amount of fuel held up by the separation and fuel assembly plants would not be proportional to the burnup in actual plant practice. Note that the above cost of fuel is for loadings after the first.

Probably all the above costs are far too low but they illustrate the low fuel charges which are potentially possible with high burnout and high rates of fuel burnup.

As a second example, let us compare a sodium-cooled, graphite-moderated reactor with a light water cooled and moderated reactor in respect to fuel charges only. Here the differences under study are the conversion ratio and the thermal efficiencies. Let us assume the following values:

	<u>H₂O</u>	<u>NaC</u>
Average conversion ratio	0.80	0.70
Thermal efficiency	25%	35%

Basis 1 ton of fuel with 2 atoms/100 burnout

For water, fuel will cost \$8,000/ton

For the NaC reactor the fuel would be 1.25% U²³⁵.

Gm/ton: U²³⁵ initial 1.25% = 11,300

U²³⁵ and Pu in reject: 11,300 x 0.70 = 7910

Using the same fabrication and separation costs mentioned above, we may calculate:

U²³⁵ added to natural U to give enriched

$$0.00539 \times 2000 \times 454 = 4890 \text{ gm.}$$

Enriched fuel will cost/ton:

$$20 + \frac{8980 \times 0.55}{99.3} = \$69.8/\#$$

$$69.8 \times 2000 = \$139,600$$

Fabrication and recovery \$30,000

Pu in reject = 7910 x 0.80 = 6328

Sale price for Pu = 6328 x 20 = \$126,600

Net cost per ton of fuel = \$43,000

$$\text{kwhr/ton electric} = 2.16 \times 10^4 \times 18,160 \times 0.35 = 1.37 \times 10^8$$

$$\text{Cost/kwhr for fuel} = \frac{4.3 \times 10^7}{1.37 \times 10^8} = 0.31 \text{ mills}$$

Inventory charge:

Fuel value in \$149,600

Fuel value out \$ 106,600 (no value for U)

Average value \$128,000

$$\text{Inventory charge/yr} = 0.12 \times 128,000 \times 1.5 \text{ for 1.5 charges} = \$23,000/\text{yr}$$

$$\text{Cost/kwhr} = \frac{2.3 \times 10^7}{1.37 \times 10^8} = 0.17 \text{ mills}$$

Total cost of fuel/kwhr = 0.48 mills compared with 0.25 for H₂O on one year's inventory charge.

The use of D₂O in a reactor requires a high inventory charge for the D₂O plus a makeup loss which might be 5% of the D₂O per year. For a reactor using 150 tons of D₂O these costs would be:

$$\text{Cost of D}_2\text{O} = 50 \times 150 \times 2000$$

$$= \$1.5 \times 10^7$$

$$\text{Inventory cost} = 0.12 \times 1.5 \times 10^7$$

$$= \$1.8 \times 10^6 \text{ per year}$$

$$\text{Makeup at 5\%} = \$7.5 \times 10^5 \text{ per year .}$$

LECTURE 24

PRINCIPLES OF SOLVENT EXTRACTION*References:

- "Absorption and Extraction," T. K. Sherwood, R. L. Pigford, McGraw-Hill Book Co., New York, (1952)
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Introduction

The irradiation of solid natural or slightly enriched U results in the production of Pu and fission products intimately mixed in the crystals of the U. The irradiation of solid Th or a Th-U²³³ alloy results in a similar mixture of products. Likewise, the irradiation of Al-U²³⁵, Zr-U²³⁵, or Al-Pu alloys result in part of the U²³⁵ or Pu present being converted to fission products. In homogeneous reactors, a solution is formed in which the Pu and fission products are dissolved or precipitated as they are formed during irradiation. Only a small fraction of the fissionable materials and only a very small part of the U²³⁸ or Th react during the irradiation. Nevertheless, the materials have to be removed from the reactor due to one or more of the following items: irradiation damage of solid fuel elements, buildup of fission products, and loss of reactivity.

After removal from the reactor, it is necessary to "recover" the fissionable materials present and the unused U²³⁸ or Th in "separation" plants. The methods used to date have consisted of dissolving the fuel elements in acid (or forming an acid solution) and subsequently separating the Pu and U by precipitation or solvent extraction. The precipitation procedures and the

* Most of this Lecture was prepared by Robert S. Voris.

use of "carriers" to increase the size of the precipitate were described briefly in the Manhattan Project literature made public at the end of the War. This lecture discusses the principles of solvent extraction while the equipment is discussed in the next lecture.

The fission products are extremely radioactive and emit beta particles and gamma rays of very high energy. For this reason the separated or recovered fissionable materials, U^{238} , and Th must be very completely freed of the fission products or the materials can only be manufactured into new fuel elements behind heavy shielding. Only one part of the fission products in 10^{10} parts of U or Pu can be tolerated without gamma shielding. The Pu and U^{233} must be handled behind light shielding at all times because of the spontaneous emission of alpha particles. Thus, decontamination factors of the solutions in which the U and Pu are dissolved of 10^7 to 10^8 are required. Because of the high value of pure fissionable materials, of the order of \$20/gm, the recovery of the Pu and U^{233} must approach 100%. These are the two requirements on separation plants - high recovery of small amounts of materials and high purity of products.

The amount of Pu present in irradiated U will vary 0.1% to perhaps 0.8%. To recover 99% of 0.1% means that only one part Pu in 100,000 parts of U may be rejected and only one part in several million parts of solution may be rejected. The amounts of Pu and U^{235} in the Al and Zr alloys mentioned range from 1 to 10% of the fuel element after irradiation.

Solvent extraction has been used as described here for about 100 years. The first use was in France. Liquid SO_2 was used to purify (French raffiner) kerosene by removal of odors, presumably S compounds. In the past 30 to 40 years, extensive uses of solvent extraction have been developed in petroleum refining.

The use of solvent extraction to recover Pu and U from irradiated materials is very expensive compared to non-solvent extraction processes now under development, e.g., distillation of fluorides and pyrometallurgical processes, such as slagging as used in the open hearth steel furnaces.

Definitions

Solvent extraction may be defined as the separation of two components or groups of components by means of the transfer of one of the components to a solvent and the subsequent recovery of the desired component by a second solvent extraction, evaporation, or precipitation.

In order for solvent extraction to be of use, a solute must be present in a solution, the solute must be valuable either for its recovery or removal, and the solute must be soluble in some other liquid that is not soluble or only

slightly soluble in the first solvent. Separation depends on the transfer of this solute, which may consist of one or more components, between two essentially immiscible phases by virtue of the difference in solubilities of the components. Liquid extraction can be simple or fractional (depending on whether a one-component transfer or multicomponent separation is made), single or multiple contact (depending on the number of contacting stages), and batch or continuous.

All extractions require four steps: (1) mixing of solvent and solution, (2) separation of the phases, (3) recovery of the solute from the "extract," and (4) recovery of solvent from both "extract" and "raffinate" by a subsequent extraction or by distillation. The product may be recovered by stripping (a second extraction) away from the first extract; distillation may be used to recover solvent.

Contact between the original solution and the solvent may take place in a variety of equipment types such as mixer-settlers, open, plate, or packed towers, or centrifugal contactors. For economic reasons, continuous countercurrent extraction is preferred in most operations, and the most common extraction is in a simple column which merely strips one component from a solution for recovery or separates two components by selective solubility of one component in the solvent. Compound columns employ a center-feed and can be treated as having enriching and exhausting sections above or below the feed. In many processes, the compound columns have extraction and scrubbing sections while the simple columns are for stripping only.

A column is often simply a vertical pipe or tube in which some sort of packing, such as Raschig rings, has been placed to break up the droplets of the disperse phases and form renewed contact surfaces. A Raschig ring is simply a cylindrical sleeve with height equal to the diameter. The column will be filled at all times with one of the two phases and the second dispersed phase will pass up or down the column. To operate, the two phases must have sufficient difference in gravity to separate rapidly. In the separation of U, Pu, and fission products, a nitric acid solution in water may be used as the feed and some organic material such as ordinary ether may be used as the solvent. In this case the water solution or aqueous phase fills the column. The U is transferred to the ether in the first column and re-extracted into a water phase in a second column under slightly different operating conditions. The Pu can be separated in a third step.

In compound columns a scrub is used to back-extract the impurities that dissolve in the extract in equilibrium with the more concentrated feed solution. Thus, water-soluble contaminants in the extract are pulled back into the scrub solution and join the raffinate. The raffinate may be a waste solution or the product. In the nuclear energy plants, the raffinate contains the fission products.

A liquid-liquid extraction column works on the countercurrent principle, i.e., the organic and aqueous phases flow in opposite directions. This is the most efficient way for operating any diffusion process, see Figure 1.

FIGURE 1

Typical Stripping and Decontaminating Columns

It should be noted that the raffinate is in contact with the fresh solvent and the extract is in contact with the feed or scrub.

A quantitative understanding and design of extraction equipment requires three basic tools:

- (1) Phase equilibrium concepts and data;
- (2) Appropriate material balances and their stoichiometry;
- (3) Diffusion-rate equations.

Phase Equilibria

In the ideal case for a solute partitioned between two immiscible phases, the distribution law holds and

$$y = mx \tag{1}$$

where

y = solute concentration in extract phase
 x = solute concentration in raffinate phase
 m = distribution coefficient

However, m usually varies with concentration and other factors such as salt content, pH, temperature, etc. Therefore, the "equilibrium curve" must be determined experimentally under conditions approximating those existing in the column.

Such equilibrium and solubility data can be represented on triangular coordinates, but graphical procedures on ordinary coordinates are normally used in calculations.

Material Balances

The equation for the balance of solute in and out of a column section determines the "operating line," which has a slope equal to the flow ratio of raffinate/extract (also called aqueous/organic or L/V). L and V are carry-overs from the McCabe-Thiele theory of distillation in which L is the moles of liquid down the column and V is the moles of vapor up the column per unit time, see Figure 2.

FIGURE 2

Mathematical Quantities in Solvent Extraction

$$V(y - y_2) = L(x - x_2); V(y_1 - y) = L(x_1 - x) \quad (2)$$

where solute is transferring from the L (aqueous raffinate) to V (organic extract).

The units here are represented as mole fractions of solute, but any consistent set of units may be chosen. Moles of solute per mole of non-solute (x and y) are often used, in which case L and V are in moles of non-solute.

Theoretical Stages

By plotting the operating line and equilibrium curve on a y versus x plot in consistent units, it is possible to step off the "theoretical stages" between the top and bottom ends of the column, see Figure 3. This provides a measure of the difficulty of such a separation. For a given column, the column height divided by the theoretical stages is the HETS (height equivalent to a theoretical stage) which is often used as a design criterion. Since HETS varies widely with operating conditions such as type of system, flow rate, concentration, and tower geometry, it is necessary to have very specific HETS data for design by this method.

Fundamentally, this method is unsound but in view of the incomplete state of the art for more exact treatments, it is a good tool practically. It is particularly useful for extraction in a series of mixer-settlers, where values for stage efficiencies allow the number of actual stages to be estimated from the stepwise estimate of the theoretical stages required.

Rate Equations

In a continuous countercurrent packed tower, the concentrations vary from point to point; if the equilibrium and operating lines have appreciably different slopes, the transfer unit method is a better design criterion than is the HETS method because the driving force for the transfer unit method is based on an integrated driving force in preference to a constant driving force over a theoretical stage. The number of transfer units, unlike the theoretical stages, however, must be based on a particular phase (organic or aqueous). It turns out that the number of theoretical stages is intermediate between the two transfer unit numbers.

Liquid extraction rate equations are based on the two-film theory and on the assumption that the interphase transfer rate is proportional to a driving force expressed as concentrations of the diffusing substance. It is assumed that equilibrium exists at the interface and that the resistances of the two films are additive, see Figure 4.

$$N \, dA = k_R(x - x_i)dA = k_E(y_i - y)dA \quad (3)$$

where

N = rate of transfer of solute in lb moles/(hr)(unit area)

A = phase contact area

k_R, k_E = film coefficients for raffinate and extract phases in
lb moles/(hr)(ft²)(lb mole/ft³)

Figure 3

PLOT OF "THEORETICAL STAGES" IN SOLVENT EXTRACTION

Figure 4

REPRESENTATION OF EXTRACTION TERMS

x, y = concentrations of solute in main body of raffinate and extract phases, in lb mole per cu ft

x_i, y_i = concentrations of solute in raffinate and extract phases in equilibrium at interface

Since it is difficult to isolate individual film coefficients, the over-all coefficients K_R and K_E are usually considered in terms of an over-all driving force, i.e., concentrations between the two main streams. Since solute concentrations as such in different solvents cannot be subtracted to get over-all driving forces, it is necessary to take the over-all driving force as the difference between the actual bulk concentration in one phase and the concentration corresponding to equilibrium with the main body of the other phase.

$$N \, dA = K_R (x - x^*) dA = K_E (y^* - y) dA \quad (4)$$

where x^* is that concentration in the x (or R) phase which would be in equilibrium with a concentration of y in the E phase, and y^* is that concentration of y which would be in equilibrium with x . Such driving forces can be estimated at any point from the equilibrium and operating lines.

Where the distribution law holds, i.e., $y = mx$, $x - x^*$ is proportional to $y^* - y$ and

$$\frac{1}{K_E} = \frac{1}{k_E} + \frac{m}{k_R} \text{ and } \frac{1}{K_R} = \frac{1}{k_R} + \frac{1}{mk_E} \quad (5)$$

It can be seen that, depending on the equilibrium slope m , the over-all K may approach an individual film k , in which case that film is said to be controlling.

In most types of equipment, the interfacial contact area (A) is not known, so it is convenient to express the unknown area per cu ft of equipment (a) with the transfer coefficient giving

$$N a dV = K_R a (x - x^*) dV = K_E a (y^* - y) dV \quad (6)$$

where $K_E a$ is the capacity coefficient in lb moles/(hr)(ft³)(lb moles/ft³) if the solute concentrations are in lb moles/ft³. For large values of m (i.e., high extract solubility) $K_R a$ is apt to be controlling and for small values of m (i.e., high raffinate solubility), $K_E a$ may be controlling. In such cases, the experimental data should be expressed in terms of the controlling phase.

For constant flow rates of extract (E) and raffinate (R), a material balance gives $SRdx = SEdy = NadV$. Therefore,

$$\text{tower height} = Z = \frac{V}{S} = \int_{x_2}^{x_1} \frac{Rdx}{K_{Ra}(x - x^*)} = \int_{y_2}^{y_1} \frac{Edy}{K_{Ea}(y^* - y)} \quad (7)$$

where S is the column cross-section and V is the column volume. Here R and E are the flow/unit column area corresponding to the L and V flow rates mentioned earlier.

These integrals may be found graphically, plotting $\frac{1}{x - x^*}$ (from the difference between x on the operating line and x^* on the equilibrium line) versus x, or $\frac{1}{y^* - y}$ versus y.

Using the concept of over-all transfer units, the difficulty of a given separation is indicated by

$$N_{OR} = \int_{x_2}^{x_1} \frac{dx}{x - x^*} \text{ or } N_{OE} = \int_{y_2}^{y_1} \frac{dy}{y^* - y} \quad (8)$$

so that tower height = $(HTU)_{OR}N_{OR} = (HTU)_{OE}N_{OE}$. Thus,

$$\frac{R}{K_{Ra}} = HTU_{OR} \text{ and } \frac{E}{K_{Ea}} = HTU_{OE} \quad (9)$$

here N_{OR} = number of transfer units based on raffinate

HTU_{OR} = height of a transfer unit based on raffinate.

In the general case, the number of over-all transfer units must be obtained by graphical integration of

$$\frac{1}{x - x^*} \text{ vs } x \text{ or } \frac{1}{y^* - y} \text{ vs } y \quad (10)$$

For those systems where the distribution law holds (straight equilibrium line) and the extracting solvent is pure, an algebraic conversion can be made to give the transfer units based on the phase being extracted.

$$N_{OR} = \frac{2.3 \log[(1 - P)M + P]}{(1 - P)} \quad (11)$$

where P = extraction factor = $\frac{R}{mE}$ or $\frac{L}{mV}$ and $M = \frac{x_{in}}{x_{out}}$ for extraction from the raffinate (aqueous phase) to extract (organic phase).

The number of stages is given by

$$N_S = \frac{[\log (1 - P)M + P]}{\log(1/P)} \quad (12)$$

This type of Colburn equation is frequently used to calculate transfer units, a measure of the difficulty of a separation. Graphs are also available for solving these equation (see Perry, page 554).

In the use of such equations, the literature lists a variety of nomenclatures, so the proper form for direction of the diffusing component, e.g., extracting or stripping, must be considered.

Nuclear Fuel Processing

The value of the distribution or partition coefficient is generally designated as E_a^O which refers to the ratio of solute (e.g., uranyl nitrate) in the organic phase to that in the aqueous phase. The volume basis (weight/unit volume) and mole ratio (moles solute per mole non-solute) are often used, although any other units such as weight/weight or moles/volume can be used, as long as they are kept consistent. This E_a^O is a function of several factors, which may influence the molecular form of the solute in solution or shift the ionization equilibrium of the solute.

1. Solvent

This is, of course, an important element in devising a separation process. Factors to be considered include: selectivity, aqueous solubility, chemical stability, radiation stability, vapor pressure, flash-point, density, viscosity, ease of purification and recovery, availability and cost, corrosiveness, and toxicity. Diethyl ether has been used in U purification. Other solvents which have been investigated at some length include hexone (MIBK, methyl isobutyl ketone), TBP (tributyl phosphate), diisopropyl ether, penta-ether (dibutoxytetraethylene glycol), diisopropyl carbinol, and Butex (dibutyl carbinol). In general, those molecules which have an electronegative oxygen functional group (ketones, alcohols, ethers) are good solvents. A larger proportion of oxygen increases the extractive power of a solvent for water-soluble ions; higher molecular weight decreases it.

TBP represents an unusual case because it shows a strong, specific uranium solvent action which results from the formation of a definite coordination complex.

Thorium forms $\text{Th}(\text{NO}_3)_4 \cdot 4\text{TBP}$; because of this, separation of U from Th in breeder blankets is difficult with TBP. The TBP may be diluted with non-polar hydrocarbons (kerosene-type) or carbon tetrachloride to improve characteristics such as density, viscosity, emulsification, solubility, and improve U stripping.

2. Solvation

The relative binding of solvent molecules by solute affects the E_a^O . Uranyl salts are hydrated by water and the ion probably transfers with 3 - 4 molecules of water. In hexone solvent the uranyl ion probably is solvated by two molecules of hexone. In TBP, the solvation of uranyl ion is quite pronounced.

3. Salting

A common ion, e.g., nitrate, can shift the equation below by the mass-action law and increase the transfer of uranyl ion to the organic phase.

Nitric acid functions as a salting agent, as do $\text{Al}(\text{NO}_3)_3$, and NH_4NO_3 . $\text{Al}(\text{NO}_3)_3$ is frequently used because, in addition to the large number of nitrate ions per mole, the aluminum ion has a high affinity for water of hydration. It, therefore, tends to decrease the H_2O concentration, increase the relative concentration of nitrate, and increase the E_a^O . It has the disadvantages of being difficult to recover and occupies large storage volume as a waste.

In an organic solvent such as hexone, the amount of HNO_3 that can be added as a salting agent is limited because of reaction with the solvent. At high temperatures danger of explosion exists. Some solvents, such as TBP, can tolerate relatively large quantities of HNO_3 . The HNO_3 can be recovered from the wastes by distillation whereas $\text{Al}(\text{NO}_3)_3$ cannot.

Some of these effects are illustrated in Table I.

In all comparisons of E_a^O 's, one must specify carefully the conditions.

4. pH

The hydrogen-ion concentration may control the ionic or molecular species and thus influence relative solubility. For example, uranyl ion is dimerized in water above pH 3 (0.025 M uranyl nitrate) so that low pH (more acid) favors the monomeric ion.

Table I

EFFECTS OF SALTING AGENTS ON E_a^0

$\text{UO}_2(\text{NO}_3)_2$ gm/l	HNO_3 M	$\text{Al}(\text{NO}_3)_3$ M	E_a^0 in (gm/l organic) nexone (gm/l aqueous)
1	0.3	0	0.007
1	0.3	0.73	0.5
1	0.3	1.46	8
10	0	0.53	0.07
10	0.3	0.53	0.11
10	0.6	0.53	0.30
1	0.3	0	0.007
10	0.3	0	0.01
100	0.3	0	0.02
1000	0.3	0	0.50

5. Chelation

For specific removal of a given ion, a process called chelation has been demonstrated on a small scale and finds use in analytical separations. Chelates are molecular complexes of specific geometry; the β -diketones, particularly TTA (thenoyltrifluoroacetone) are excellent for removing Pu (in the IV state) from aqueous solutions.

in which HK represents the chelate former, TTA.

This extraction is very dependent on the acidity and hence great selectivity is possible for various ions. Those ions appreciably extracted from 0.5 M HNO_3 are Pu(IV), Zr(IV), Np(IV), Ce(IV), Fe(III), and Sn(IV). The organic solvent may be benzene, o-dichlorobenzene, or sec-butylbenzene. The oxidation state must be carefully controlled in the Pu(IV) state. The Pu(IV) can subsequently be reduced to Pu(III) and stripped from the organic layer or stripped as Pu(IV) by stronger HNO_3 .

6. Concentration

Depending on acid and salting concentrations, the E_a^0 of uranyl nitrate in hexone can increase or decrease with uranium concentration in the organic phase; in TBP the E_a^0 decreases with uranium saturation. This can be regarded as a self-salting effect. The uranium concentration can also affect the distribution of other ions.

A procedure for separating U, Pu, and fission products by solvent extraction has been described by Bruce. This involves extraction of Pu and U away from fission products in a nitric acid solution using ether solvent and a scrub solution, then selective partition of Pu in reduced valence state into water, and finally stripping of U from solvent into water. This is outlined in Figure 5. A very similar system was described in a previous lecture on separation of U and Th from their ore concentrates.

Figure 5

SOLVENT EXTRACTION PURIFICATION OF IRRADIATED URANIUM

LECTURE 25

ENGINEERING AND SOLVENT EXTRACTION DESIGN*References:

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Introduction

The basic requirements for the design of solvent extraction plants for recovery of Pu, Th, and U from irradiated fuel elements are:

- (1) High recovery efficiencies of 99.9% or so.
- (2) High decontamination factors, 10^7 based on the volume of solution or 10^{10} based on volume of enriched materials.
- (3) Protection of personnel from the beta and gamma rays emitted by the fission products and also from the alpha particles emitted by U^{233} and Pu.

In addition to the basic requirements, there are the normal economic and engineering requirements such as:

- (1) Low costs
- (2) High throughput
- (3) Ease of maintenance
- (4) High solvent recoveries
- (5) Low waste volumes or readily disposable wastes
- (6) Low fire hazard
- (7) Low labor, laboratory, and supervisory personnel requirements.

These two sets of requirements are incompatible in respect to protection of personnel and low costs for the reason that the protection requirement combined with maintenance of contaminated equipment is just horribly expensive. The equipment and chemicals required would be costly enough without the shielding, maintenance, and waste storage problems.

Maintenance Philosophy

There are at least three types of solvent extraction plants that can be built and operated for U and Pu recovery. These plants differ in respect to method of maintenance. All three plants are readily designed for remote operation and control - in fact they would be so designed for operation if no radioactivity were present. Further, if no leaks ever occurred and if the equipment could be readily decontaminated, any design could be utilized.

Actually, two types of plants have been built and a third type studied in this country. These are:

(1) Remote maintenance. In this plant the equipment is so designed and located that the piping can be disconnected and the equipment removed by remote operation. After removal, the damaged equipment is either buried or decontaminated and repaired.

(2) Direct maintenance. In the direct maintenance plant, the equipment is fixed in location and most of the piping installed by welding. In case repairs or alterations are required, the equipment and piping are decontaminated in place and then repairs carried out by men working directly on the equipment.

(3) The third type of plant is designed for installation in a water-filled canal. Operation and maintenance is carried out remotely, but quite directly, through the water.

The advantages of the remote maintenance over the direct maintenance plant are:

(1) Immediately after a breakdown or difficulty occurs, the equipment in a given cell or section of the plant can be drained, flushed, and disassembled for removal and replacement. The plant-down time for repairs is low. In the direct maintenance plant, it is necessary to decontaminate the entire cell's equipment before maintenance can be carried out.

(2) A leak does not become a major problem as it need not be cleaned up before repairs are carried out. In the case of the direct maintenance plant, the material which leaks out must be cleaned up remotely before repairs can be started. This is frequently difficult to do and is almost always time consuming.

The disadvantages of the direct maintenance plant as compared with the indirect maintenance plants are:

(1) Greater complexity of design. In order to carry out remote disassembly operations each pipe line must be connected to each piece of equipment by a jumper pipe. A jumper pipe is connected to the vessel and outside

pipe by gasketed joints which are pulled up by screw-jack connectors which can be screwed up by a remotely operated electrical wrench. In the direct maintenance plant, the lines are welded in place. In the remote maintenance plant, extra lines between cells must be provided in case a line fails. In the direct maintenance plant, lines are usually run through sleeves set in the shielding walls and the lines can be renewed.

(2) The large number of jumpers and remote connections require that more space be provided around equipment than is needed in the direct maintenance plants. Therefore, the shielded space may become greater.

(3) Leaks are more apt to occur in the remote maintenance plant because of the large number of connections. This is not an important consideration. As noted, provision must be made for cleanup of leaks in the direct maintenance plants. Such facilities include stainless steel sheets welded together and covering the entire inside surface of the cell. Strippable paint may also be used.

Over-all Plant Design

A separations plant consists of several cells in which the dissolvers, extraction columns, receiving tanks, solvent recovery, and other equipment in which the fission products are present, surrounded by control rooms, rooms for solution makeup, etc. A cross section of such a plant is presented in Figure 1. The dissolvers and other reaction vessels are indicated in a central gallery which might be several hundred feet long and separated into cells by thick, shielding-type cross walls. The walls surrounding the reaction vessels are several feet thick.

The ceiling above the reaction vessels is made of 6-ft thick, removable concrete blocks. Maintenance is carried out by removing these blocks, thus opening the cell. The defective piece of equipment is disconnected and replaced and the connections and shielding replaced. The total enclosed space becomes very large compared to the size of the reaction vessels.

The non-radiative portion of the plant is very similar in appearance to a fine chemicals plant. There are extensive control panels for the normal remote operation of the plant, data indicating and recording equipment. Solvents, solutions, etc., are prepared in separate rooms and fed to the reaction vessels by pumps or gravity.

The transfer of radioactive materials is a very difficult procedure since pumps are to be avoided due to their frequent maintenance problems. Gravity flow is used as much as possible. When necessary, liquids may be moved by steam ejectors. Valves are avoided when possible within cells and only bellows type valves are used.

Figure 1 - Cross Section of Separations Plant

Materials of construction of the radioactive portion of the plant are few. All equipment including pipelines, instrument lines, etc., is made of stainless steel, either 347 Nb-stabilized or 304 ELC. All welds are made by deep groove welding and grinding smooth on the inside when possible. Welding and passification of the steel are specified in detail and inspected with unusual care. As a result, leaks and corrosion difficulties are infrequent.

In so far as possible, all equipment is designed to eliminate all moving parts and a minimum number of gaskets are used. In pulsed columns and mixer-settlers there are moving parts, but these are the exceptions, not the rule. Gaskets are usually Teflon, well supported and held in place.

Decontamination of equipment is carried out by steaming and hot acid and caustic washes.

Equipment

The basic decisions that must be made in the design of a given fuel recovery and separation plant besides the type of maintenance, plant capacity, solvent, etc., are the types of contactors that are to be used. The equipment may consist of packed columns, mixer-settlers, or other types described below. Pumps, valves, etc., may be special.

Solvent Extraction Equipment

The available types of contacting equipment include:

A. Mixer-settlers

1. Tanks with propeller or turbine agitators.
2. Flow or in-line mixers such as jets, injectors, orifices, nozzles, or pumps.
3. Scheibel column.
4. Compartmented settlers, such as the Fenske contactor.

B. Continuous columns

1. Film
2. Spray
3. Baffle (e.g., donut-disk)
4. Perforated or sieve-plate
5. Packed (e.g., rings, saddles)

C. Centrifugal Extractors (e.g., Podbielniak)

The choice of a particular type will depend on reliability of data, emulsification properties, available space, sedimentation, settling rates, etc.

The mixer-settler sequence has the advantage that the individual stages approach theoretical stages and experimental data can be scaled up with more assurance. In addition, there is less limitation on flow rates and hydraulic balance and there is more flexibility for handling a variety of systems and unfamiliar systems. It avoids high head room which may be expensive (e.g., if shielded), although columns may protrude through the bottom of the canyon into the ground.

Mixer-settler devices have been developed to accomplish (1) intimate phase dispersion without "back-mixing" between stages, (2) complete phase settling, and (3) hydraulic stability. The net flow of phases can be provided by gravity or by "pump-mix" agitators. In some of the work at KAPL, for example, centrifugal pump-mix impellers are driven by vertical shafts and the horizontal settlers provide 1-2 minutes hold-up. Baffles allow the separated bottom and top phases to pass to the mixing chambers of the preceding and following stages, see Figures 2 and 3.

An ingenious stacked-stage extractor developed by M. R. Fenske has shown promise in pilot-scale work. This consists of a vertical counter-flow assemblage of flat stages in which the two phases are agitated by oscillating perforated plates, all driven by a common shaft. This extractor is characterized by low hold-up, high through-put, and high stage efficiency.

The Scheibel column consists of alternate empty and packed sections, with a vertical shaft fitted with agitator blades in each empty section. The agitated sections provide mixing; the packed "calming" sections permit coalescence and vertical flow of the dispersed phases. HETS values as low as 2 in. on small columns have been reported and stage efficiencies of 85-110 per cent can be obtained. (The 110 per cent figure indicates that some additional extraction occurred in the coalescence sections.) The efficiency depends on rotor speeds, which range up to 600 rpm.

Film towers have been used primarily for control of interface area in wetted-wall towers. Spray towers are simple, cheap, have high flow rates, and are easily cleaned. Proper design usually calls for a flared wall at the dispersion point (to raise the flooding point) and droplets in the vicinity of 1/8-in. diam.

Baffle towers are versatile and fairly efficient; the donut-disk type can be equipped with rotary scrapers to remove accumulated solids. Perforated plate or sieve-plate columns are widely used, particularly in the oil industry. Perforations are usually 1/16 - 3/8 in. diam, and tray

Figure 2 - Box Extractor

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Figure 3 - Cell Assembly

spacings 6 - 24 in. The Koch Cascade is a recent version giving up to 0.75 theoretical contact per tray at high through-put. Pulsed perforated-plate towers particularly can be made to give high extraction efficiency. Bubble caps are not extensively used. Packed towers are simple and cheap; acid-resistant systems can be obtained at low cost.

The Podbielniak extractor is a centrifugal contactor which can attain 4 - 8 theoretical stages. Two liquids are pumped (through rotary seals) counter-currently through a spiral channel inside a steel casing which revolves at high speed. This unit has high through-put and short residence time; it has found wide acceptance in recovery of penicillin from the fermentation broth.

A packed column consists simply of a pipe filled with some type of packing material. One of the common types of packing is the Raschig ring which is merely a piece of tubing cut into short pieces with the diameter equal to the height. These are simply dumped into the tower. Packing sizes are $1/2$ to 2 in.

The extraction efficiency of a column can be increased by a factor of 2 or 3 by means of "pulsing." In a pulse column the liquid contents are pulsed up and down through a series of horizontal, perforated plates by imposing a hydraulic pressure cycle from a piston or bellows pump on one of the feed streams. This produces the net countercurrent flow of the two phases and at the same time causes constant formation of new bubbles of the dispersed phase. In a sense, a pulse column is a series of small spray columns. In an ordinary packed column, a droplet may rise to the top without exposing a new surface; the breaking off of new droplets by the alternate "squeezing" of light and heavy phases through the perforations constantly exposes new surfaces for diffusion. Typical conditions would be plates ($1/8$ in. holes, 23% free area) spaced at 2 in. and pulses of 0.25 - 1.5 in. amplitude and 30 - 120 cycles/min. frequency. From pilot plant studies, it has been found that the capacity and efficiency (as measured by HTU) can be correlated with the product of amplitude and pulse frequency; the optimum amplitude-pulse product is 50 - 70 in./min.

The chief advantages of pulsed columns over packed columns are the reduced height, more reproducible extrapolation to larger sizes, and more reliable hydraulic balance. The power consumption is relatively large, however, and mechanical complexity is increased.

Mixer-Settler Calculations

The number of theoretical stages can be stepped off from equilibrium and operating lines, and then a stage efficiency (usually 75 - 100%) applied to calculate actual stages. Proper design of the settling and phase-separating sections is just as important as providing adequate mixing.

Knowing the terminal conditions and the solvent ratio, the operating line can be constructed and the ideal stages determined. If the number of stages is set in advance, trial and error choice of operating lines is required to satisfy the desired terminal conditions. The slope then gives the solvent quantity required.

For countercurrent multistage contact involving the ideal distribution law ($y = mx$) and fresh extracting solvent, the fraction of the solute unextracted after the N 'th stage is

$$Z_n = \frac{P - 1}{P^{N+1} - 1} \quad (1)$$

Where P is the extraction factor ($\frac{m}{L/V}$) i.e., ratio of slopes of equilibrium and operating lines. Here L is the weight of solvent in the feed and V the weight of pure extracting solvent.

Continuous Column Calculations

In continuous countercurrent columns, a similar procedure for stepping off the theoretical stages is used, but in this case, the physical identity of the actual stages vanishes and it is necessary to assume some value of the HETS (height equivalent to theoretical stage) to calculate the tower height. For cases involving ideal distribution and pure extracting solvent, an algebraic solution is possible for the number of stages required:

$$N_s = \log \frac{[(1 - P)M + P]}{\log(\frac{1}{P})} \quad (2)$$

where P = extraction factor: $\frac{L}{mV}$ for extraction and $\frac{mV}{L}$ for stripping (i.e., ratio of slope of equilibrium and operating line); and

$$M = \frac{x \text{ in}}{x \text{ out}} \text{ for extraction or } \frac{y \text{ in}}{y \text{ out}} \text{ for stripping.}$$

For a rough estimate of tower height, published values of HETS may be consulted, but these vary widely with operating conditions, such as flow rates, solvent system, and tower geometry. In small columns, the HETS may be as low as 1 - 2 ft, but in larger diameter columns may reach 100 ft. Experimental data on HETS can also be misleading because of end effects, particularly in spray towers. This means that a large part of the observed extraction occurs in the first few inches of contact and tower heights cannot be extrapolated.

Distributor devices to minimize channeling are essential in larger columns. Therefore, it is important that pilot plant runs be made on the particular system if reliable extrapolation is necessary.

A more fundamentally correct procedure is to use the transfer unit rather than the theoretical stage method. The number of transfer units and the HTU (height of a transfer unit) take into account the rate of transfer, based on over-all concentration driving forces across the film resistances. Since the transfer units can be based on either solvent, i.e., continuous or discontinuous phases, one solvent film may be controlling. Presumably, the relative magnitude of the film resistances depends on the relative solute solubilities (i.e., on m), but, actually, it appears that it may depend more on the phase-contacting area per unit volume. As a general rule, the phase fed at larger volumetric rate should be dispersed; in packed columns the phase which wets the packing should be continuous regardless of the flow rate. Radiation exposure of organic solvent in contact with hot aqueous feed can be reduced by dispersing the organic phase. Most available literature data have been based on the aqueous phase continuous and controlling.

The transfer units and HTU values are generally based on the phase from which the solute is transferring, i.e., water for extraction and organic for stripping. The organic phase is generally dispersed, making the interface level control simpler.

Much of the data on extraction performance has been correlated in terms of K_a (e.g., K_{Da} where the discontinuous phase resistance is controlling), usually as a function of flow rate of one liquid phase. The usual units for K_{Da} , an experimentally determined quantity, are $\text{lb mol}/(\text{hr})(\text{ft}^3)(\text{lb mol}/\text{ft}^3)$. For example, if the flow rate of the controlling phase (L_D) is expressed as ft/hr , then the HTU in feet (for the ideal system) is

$$\text{HTU}_{OD} = \frac{L_D}{K_{Da}} \quad (3)$$

To calculate the over-all transfer units required for transferring solute from a continuous aqueous raffinate to a dispersed organic extract, the general equation is

$$N_{Ox} \text{ or } N_{OR} = \int_{x_2}^{x_1} \frac{(1-x) O_M dx}{(1-x)(x-x^*)} \quad (4)$$

where

x = concentration in raffinate phase

x^* = concentration corresponding to the equilibrium value for x

$$(1-x)_{OM} = \ln [\text{mean of } (1-x) \text{ and } (1-x^*)]$$

For dilute solutions, this becomes

$$N_{OR} = \int_{x_2}^{x_1} \frac{dx}{x - x^*} \quad (5)$$

even though the equilibrium line is curved.

If both equilibrium and operating lines are straight, an algebraic solution can be made according to

$$N_t = \frac{2.3 \log (1 - P) M + P}{(1 - P)} \quad (6)$$

where P and M are the same as defined for the earlier stage calculation.

The extraction factor, which is the ratio of slopes of equilibrium and operating lines, $(\frac{mV}{L})$, is of great importance in locating the film of principal resistance and determining the economics of the separation. It is dimensionless. Since, for ideal dilute systems,

$$HTU_{OC} = HTU_C + (\frac{L}{mV}) HTU_D \quad (7)$$

and

$$HTU_{OD} = HTU_D + (\frac{mV}{L}) HTU_C \quad , \quad (8)$$

then for large $\frac{mV}{L}$, the principal resistance is in the L (here the continuous) phase; for small values, the resistance is in the V (here the discontinuous) phase. The relationship between the two N's is

$$N_{OD} = \frac{L}{mV} N_{OC} \quad , \quad (9)$$

If $\frac{mV}{L}$ is smaller than 1, the equilibrium and operating lines will intersect and a limiting raffinate composition is reached, regardless of stages. The greater the $\frac{mV}{L}$, the fewer transfer units required, but the more extract which must be recovered. The economic balance is probably in the range of 1.5 - 2.0.

Even though m and $\frac{L}{V}$ may vary widely, the range of $\frac{mV}{L}$ in industrial processes is small, usually from 1.25 - 2.0. (This considers solute transferred from raffinate L or x phase to extract V or y phase; for reversed direction, the inverse value, 0.8 ~ 0.5, applies) There is also evidence that the efficiencies of phase contacting devices change with $\frac{mV}{L}$.

The total transfer units or stages are calculated by graphical integration wherever the equilibrium curve is not straight (such as in the more concentrated region); to this is added the value obtained by algebraic solution of that region where an average m can be assumed (such as in the pinch region).

Flooding Points

The height of a tower is determined by the HTU or HETS, but the minimum cross section is determined by the allowable flow rates of the phases. Maximum throughput for extraction towers is limited by the flood point above which the discontinuous liquid is carried back with the continuous liquid; this in turn depends on density differences, drop size, relative flow rates, and type of packing. Spray towers have 2 - 6 times the capacity of packed towers.

Correlations for flooding point are still being developed and are being reported in the literature.

Example I

The equilibrium distribution of uranyl nitrate, $\text{UO}_2(\text{NO}_3)_2$, between diethyl ether and water has been reported by L. G. Jodra, A. P. Luina, and A. Oroz. If an ether solution of $\text{UO}_2(\text{NO}_3)_2$ (35% by weight) is to be stripped with water in a tower to transfer 95% of the U to the H_2O , what is the minimum ratio of H_2O /ether that can be used? At a H_2O /ether ratio of 1.1, calculate the number of theoretical stages (N_s), and the number of transfer units based on both ether ($N_{t, \text{org.}}$) and H_2O ($N_{t, \text{aq.}}$) for this separation.

Solution

The equilibrium data in Table 1 are recalculated to weights of $\text{UO}_2(\text{NO}_3)_2$ (UN) per weight non-solute and plotted in Figure 4. Weight fractions could be used but, in view of the relatively large solute transfer, a non-changing basis is preferable.

For the conditions given, the ether feed concentration is $Y_1 = \frac{0.35}{0.65} = 0.54$ gm UN/gm solute-free ether phase and $X_2 = 0$. For 95% transfer, $Y_2 = 0.54 \times 0.05 = 0.027$; the value for X will depend on the ratio of the two flow rates.

Table 1

EQUILIBRIUM IN SYSTEM: $\text{UO}_2(\text{NO}_3)_2 - \text{Et}_2\text{O} - \text{H}_2\text{O}$ (25°C)*

Water Phase, wgt % UN	Ether Phase, wgt % UN	Water Phase, g. UN	Ether Phase, g. UN
		<u>g. non-solute</u>	<u>g. non-solute</u>
8.9	0.0008	0.0977	0.000008
17.8	1.0	0.217	0.0101
23.6	3.1	0.309	0.0320
24.7	3.3	0.328	0.0341
26.6	4.7	0.363	0.0493
27.3	5.7	0.375	0.0605
30.0	12.8	0.429	0.147
31.7	16.2	0.464	0.194
36.2	25.5	0.568	0.343
40.9	32.8	0.694	0.488
43.6	36.2	0.773	0.567
46.5	41.6	0.870	0.713

* Reported by L. G. Jodra, A. P. Luina, and A. Oroz.

The minimum ratio of H_2O /ether flow will be the slope of a line from (X_2, Y_2) intersecting the equilibrium line at $Y_1 = 0.54$. This can be found by graphical or algebraic means and is equal to $\frac{0.54 - 0.027}{0.75 - 0} = 0.685$.

This would require an infinite number of stages; therefore, a higher ratio must be used, compromising the amount of H_2O with number of stages.

For a flow ratio of 1.1, for example, a material balance gives $X_1 = \frac{0.54 \times 0.95}{1.1} = 0.467$ gm UN/gm solute-free H_2O . Stepping off the theoretical stages in Figure 4 gives $N_s = 1.9$ theoretical stages. The transfer unit numbers are determined by graphical integration in Figure 5 using data for the driving forces $(Y - Y^*$ or $X^* - X)$ read from Figure 4 and

DISTRIBUTION IN SYSTEM: $\text{UO}_2(\text{NO}_3)_2 - \text{Et}_2\text{O} - \text{H}_2\text{O}$

Figure 4 - Equilibrium Distribution in Water-Ether System

tabulated in Table II. There are found to be 1.6 units based on the aqueous and 2.2 based on the ether phase. The ether phase is controlling and extrapolations for design should be based on it.

Table II

DATA FOR GRAPHICAL INTEGRATIONS

<u>Y</u>	<u>Y*</u>	<u>Y - Y*</u>	<u>$\frac{1}{Y - Y^*}$</u>
0.540	0.200	0.340	2.94
0.450	0.080	0.370	2.70
0.432	0.055	0.377	2.65
0.411	0.043	0.368	2.72
0.375	0.030	0.345	2.90
0.335	0.020	0.315	3.18
0.270	0.010	0.260	3.85
0.235	0.005	0.230	4.35
0.200	0.001	0.199	5.03
0.100	0	0.100	10.0
0.040	0	0.040	25.0
0.027	0	0.027	37.1

<u>X</u>	<u>X*</u>	<u>X* - X</u>	<u>$\frac{1}{X^* - X}$</u>
0.467	0.750	0.283	3.55
0.385	0.660	0.275	3.64
0.295	0.575	0.280	3.58
0.203	0.500	0.297	3.37
0.110	0.435	0.325	3.08
0.040	0.380	0.340	2.94
0.020	0.365	0.345	2.90
0.010	0.345	0.335	2.97
0	0.305	0.305	3.28

Figure 5

GRAPHICAL INTEGRATIONS FOR TRANSFER UNITS

Example 2

Operation of a packed laboratory column with aqueous phase continuous and ether phase dispersed indicates that HTU_{aq} values are around 3.5 ft at this flow ratio and $6 \text{ gm}/(\text{min})(\text{cm}^2)$ flow for the organic phase. To strip 100 tons/month of $UO_2(NO_3)_2$ under the above conditions (i.e., 35% UN in ether feed, 95% transfer), what size column will be needed?

Solution

Assuming the laboratory column performance to be typical of that pertaining to a larger column, the minimum tower height would be 2.2 units $\times 3.5 \text{ ft/unit} = 7.7 \text{ ft}$. This may or may not be reliable; the more experimental data over a range of variables such as flow rates and packings, the better. On the other hand, a few feet of column one way or the other is a small factor in over-all cost, so a generous safety factor can be allowed.

To choose the column diameter, it would be desirable to have some data on flooding point for the particular packing and HTU vs % of flood point. In the absence of such data the assumption is made that the HTU will not change with scale-up. This indicates an allowable feed rate of at least

$$\frac{6 \times 60 \times (2.54 \times 12)^2}{454} = 740 \text{ lb solution/hr ft}^2. \text{ At 35\% by weight UN and 24-hr}$$

operation, the required column area is $\frac{100 \times 2000}{740 \times 24 \times 30 \times 0.35} = 1.07 \text{ ft}^2$. This would correspond to a pipe size just slightly over 14 in. diam, so a 16 in. diam pipe would be used.

Example 3

If an aqueous solution of $UO_2(NO_3)_2$ is to be extracted into ether, what is the minimum ratio of ether/ H_2O that will give an aqueous raffinate of 20 weight %? Can 95% of the $UO_2(NO_3)_2$ in a 35% by weight aqueous solution be transferred to ether? What can be done about it?

Solution

For extraction of $UO_2(NO_3)_2$ from water into ether, the operating line will be below the equilibrium curve. A straight-edge pivoted at an effluent H_2O concentration of $X = 0.25$, $Y = 0$ (i.e., 20 wt%) becomes tangent to the equilibrium curve at a slope of about 0.25. This is the maximum H_2O /ether ratio that can be used, so the minimum ether/ H_2O ratio is 4/1.

Achieving 95% removal of UN from a 35 wt % feed would require an $X_2 = 0.027$. Because of the extreme flat pinch at the dilute end of the equilibrium curve, it would take almost an infinite ratio of ether/aqueous flows, (i.e., a horizontal line); so this separation is not feasible.

The addition of a salting agent to the aqueous feed should be investigated as a means of getting around this limitation. From the data on distribution coefficients given in the preceding lecture, it can be seen that hexone systems salted with $\text{Al}(\text{NO}_3)_3$ or even HNO_3 show appreciable distribution coefficients even at low X 's. These are higher than the coefficients for the unsalted hexone system. Katzin's and Sullivan's data show negligible E_a^0 's up to 10% $\text{UO}_2(\text{NO}_3)_2$ (aqueous). Furthermore, the data of Furman, Mundy, and Morrison show that the nitrates of iron, aluminum, zinc, copper, and calcium are effective agents for salting uranyl nitrate from water to ether.

LECTURE 26

SOLVENT EXTRACTION PROCESSESOre Purification

Historically, the purification of U by ethyl ether extraction was the first in AEC technology and played a large part in achieving the first self-sustaining chain reaction. Today, tributyl phosphate (TBP) is the preferred solvent because it is more effective and less flammable.

The U_3O_8 concentrates from various purification schemes operating on pitchblende or carnotite ores are dissolved in HNO_3 and the uranyl nitrate then transferred to the organic solvent. The U is scrubbed back into water and recovered as uranyl nitrate hexahydrate (UNH). This is decomposed into UO_3 by heating.

The purification of Th is similar - a TBP-dibutyl ether mixture extraction of $Th(NO_3)_4$ to reduce rare earths and U content. For ordinary use, extraction to remove rare earths is unnecessary and removal of U by oxalate precipitation is sufficient.

Fuel ProcessingDissolution

In many fuel processing schemes, it is necessary to decan elements or slugs and dissolve them in HNO_3 . The evolution of H_2 results in an explosion hazard which could cause serious spread of radioactivity. Extreme care or use of other solvents than nitric acid is used in the decanning process.

The noble gases (Kr, Xe) which are liberated during dissolution can be vented with dilution. Venting of radioactive I_2 , which can be fixed by plant and animal life, must be reduced by long cooling for decay, retention in solution or off-gas scrubbing.

Separation of U from Fission Products

A typical process is described briefly for recovery of U from Al-U alloys. In such a system, no Pu is present, while the U is present in small quantities. The fuel assembly here contains over 90% Al by weight (the fuel meat contains about 15% U by weight); solution in HNO_3 automatically supplies most of the salting agent.

The flowsheet for the process is presented in Figure 1. The relative concentrations are given in Table I.

Homogeneous Fuels

Since homogeneous fuels are undergoing rapid development, their processing is still quite tentative. For a fuel such as uranyl sulfate, it should be possible to process by batch, intermittent, or continuous removal of fuel.

Figure 1

TYPICAL U RECOVERY SHEET

Table I

TYPICAL FLOW SHEET - CONCENTRATIONS

Stream Variables	Feed	Solvent	Scrub	Aqueous Raffinate	Organic Extract	Strip	Organic Raffinate	Aqueous U Product
Cycle 1								
	1AF	1AX	1AS	1AW	1AU	1CX	1CW	1CU
Relative flow rate	100	125	25	127	123	32.7	122	33.7
U, gm/l.	1.125	---	---	0.0001	0.914	---	---	3.34
Al(NO ₃) ₃ , M	1.60	---	1.00	1.47	---	---	---	---
NH ₄ NO ₃ , M	0.66	---	---	0.53	---	---	---	---
NH ₃ , M	0.20	---	0.20	0.20	---	0.04	---	---
Hg(NO ₃) ₂ ·0.5H ₂ O, g/l.	0.45	---	---	0.43	---	---	---	---
HNO ₃ , M	---	---	---	---	---	---	0.001	0.04
Hexone, Wt %	---	98.1	---	0.71	98.1	---	98.1	1.80
Gr	1.258	0.801	1.15	1.234	0.802	0.998	0.800	1.001
Decontamination Factor, Beta	1.0	---	---	---	---	---	---	1 x 10 ⁴
Gamma	1.0	---	---	---	---	---	---	1 x 10 ⁴
Pu	1.0	---	---	---	---	---	---	20
Cycle 2								
Relative flow rate	1.6	21.6	20	21.9	21.3	5.7	21.3	5.7
U, gm/l.	69.4	---	---	---	5.27	---	---	19.46
Al(NO ₃) ₃ , M	---	---	1.60	1.47	---	---	---	---
NH ₄ NO ₃ , M	---	---	---	0.17	---	---	---	---
NH ₃ , M	---	---	0.284	0.20	---	---	---	---
Fe(SO ₃ NH ₂) ₂ , M	---	---	0.05	0.047	---	---	---	---
HNO ₃ , M	0.83	---	---	---	---	0.04	0.001	0.04
Hexone, Wt %	---	98.1	---	0.78	98.16	---	98.1	1.66
Gr	1.12	0.801	1.240	1.226	0.81	0.998	0.800	1.02
Decontamination factor, Beta	1 x 10 ⁴	---	---	---	---	---	---	3 x 10 ⁶
Gamma	1 x 10 ⁴	---	---	---	---	---	---	2 x 10 ⁶
Pu	20	---	---	---	---	---	---	2 x 10 ³
Cycle 3								
Relative flow rate	1.6	11.7	10.1	11.8	11.6	3.1	11.5	3.1
U, gm/l.	69.4	---	---	---	9.71	---	---	35.65
Al(NO ₃) ₃ , M	---	---	1.72	1.47	---	---	---	---
NH ₄ OH, M	---	---	---	0.02	---	---	---	---
NH ₃ , M	---	---	0.25	0.20	---	0.04	0.001	---
HNO ₃ , M	0.14	---	---	---	---	---	---	0.04
Hexone, Wt %	---	98.1	---	0.79	98.22	---	98.1	1.52
Gr	1.11	0.801	1.258	1.237	0.814	0.998	0.803	1.22

LECTURE 27

METALLURGICAL PROCESSES*Reference:

"Principles of Zone Melting," W. G. Pfann, AJME, 194, 747 (1952)

Introduction

Base metals are purified by preferential oxidation of the impurities to oxides and their removal as oxides in the slag. This slagging separation process depends on the general physical phenomenon that molten oxides are soluble in other oxides and molten metals are frequently soluble in one another, but oxides are not soluble in metals or the metals in the oxides. There are a few exceptions to this rule, notably ZrO is soluble in Zr.

Other methods of purifying a metal are to utilize insoluble metals that extract third metals from the first ones. For example, molten zinc is mixed with lead to extract silver from the lead. Fused salts can also be used as extraction agents. Electrolysis, hot wire disassociation, and distillation are other means used to purify metals. As pointed out in an earlier lecture, if electrolysis is used, the oxides may be purified and pure metals produced as is the case with Al.

The methods of purification depend on their applicability primarily on the place in the Electromotive Series of the metal being purified. No detailed analysis of the various systems is given here.

Thus, we have several possible pyrometallurgical processes which might be applicable to the separation of U and Pu from the fission products. Several methods of possible interest are discussed below.

Attempts were made during the Manhattan District days to apply pyrometallurgical procedures to the separation of Pu and U from fission products. However, the amount of Pu present in U is below the level of many impurities in commercial metals and these methods are not too applicable. Also, Pu is soluble in U, so extraction would be expected to be difficult.

The possible utilization of Th, U, and Pu in power reactors has indicated that another look at the pyrometallurgical operations is justified. Here, complete separation with decontamination factors of 10^6 to 10^7 are not necessarily required, complete separation of U and Pu is not required, the concentrations of Pu and fission products are much higher, and there is a great

* Prepared by H. M. Feder

economic incentive for lowered costs and increased concentrations of fission products. Therefore, several laboratories have started development of these processes. If these methods are successful, the present fuel cycle can be shortened.

Liquation and slagging, if successful, will produce metal that must be refabricated by remote means as it will be only partially decontaminated. The development of these remote fuel fabrication techniques is a particularly difficult and challenging problem as well as being expensive. In the liquation and slagging sequence the fission products would be in a highly concentrated form.

We might define a pyrometallurgical process as one in which physical metallurgy is applied to a metallic fuel in order to recover its fissionable or fertile constituents in a relative pure metallic or oxide form.

Liquation and Slagging

This operation of liquation and slagging involves melting the U with its impurities such as fission products in a crucible, holding in the molten state for some time to permit coagulation and separation of the impurities (liquation) by floating to the top or by settling, and removal of the impurities as molten oxides, carbides, nitrides, etc., (slagging). The oxides are usually lower density and float.

The available phase diagrams of uranium metal systems indicate that of the fission elements only the alkali and alkaline earth metals have very low solubility in liquid uranium. The rare earth metals and silver have limited solubilities ($<1\%$ near the melting point) and the remaining elements have appreciably higher solubilities. The low solubility metals also have high vapor pressures and presumably will be lost by volatilization rather than liquation. A more general approach involves the formation of insoluble stable binary or ternary compounds of the fission elements. In essence, the preferential oxidation procedure is of this type. One might also use carbides, nitrides, etc., if the thermodynamic data show favorable stabilities. Some data of a sketchy nature are presented in Table I. Intelligent application of the data requires some knowledge of the actual species formed. For instance, in applying the data for the carbides, it is imperative to know which of the uranium carbides are formed. This in turn may depend on the mode of formation. When graphite is immersed in liquid uranium, the compound formed at the surface is the dicarbide. It would therefore be expected that zirconium carbide would be preferentially formed and removed provided its solubility is not too large. Molybdenum and ruthenium would not be removed in such a process. The same type of argument may be applied to the nitrides and sulfides. The data for silicides, phosphides, and borides are even sketchier so that no generalizations can be made at present about liquation processes involving the addition of silicon, phosphorous, or boron.

Table I

ESTIMATED FREE ENERGIES OF FORMATION AT 1500°K

Kilocalories per gram atoms of non-metallic form

<u>Carbides</u>	<u>Nitrides</u>	<u>Sulfides</u>
TiC -54	UN -54	SrS -93
UC -35	ZrN -47	BaS -91
ZrC -32	TiN -46	1/3La ₂ S ₃ -79
SiC -24	HfN -45	1/3Pu ₂ S ₃ -59
1/2ThC ₂ -17.6	1/4Th ₃ N ₄ -44	Rb ₂ S -58
PuC -16.5	1/3U ₂ N ₃ -41	Cs ₂ S -57
1/2UC ₂ -16.2	CeN -41	US -54
NbC -16	1/4U ₃ N ₄ -37	ZnS -29
1/2CaC ₂ - 7.8	LaN -35	CdS -16
Mo ₂ C -3.0	YN -34	Cu ₂ S -13
WC -1.4	NbN -29	1/2MoS ₂ -9
1/2Na ₂ C ₂ +3.0	AlN -25	Ag ₂ S -3
Carbides of Grp. VIII d	BN -14	1/2RuS ₂ -2
	1/2Sr ₃ N ₂ -4	1/3Sb ₂ S ₃ +3
	1/2Ba ₃ N ₂ -1	
	Mo ₂ N +15	
	Nitrides of Zn, Cd, In, Ga, and Group VIII d	

d = Spontaneously decompose

Halogenation

Methods of purification using partial halogenation have been investigated recently. Tables II and III present some of the relevant thermodynamic data for processing. In addition to the rare earths, the alkaline earths, and zirconium, partial fluorination or chlorination concentrates the alkali metals and plutonium in the halide layer. The use of the halogens must be carefully controlled in order to obtain only partial halogenation.

Distillation and Volatilization

A number of metals, Zn, Cd, Ca, Mg, Hg, and Na, may be purified by distillation. The vapor pressure of uranium is too small to make this approach feasible, but some of the impurities may be removed by volatilization in a vacuum. Table IV presents some of the available vapor pressure data. The rare gases and the metals which do not show compound formation with uranium, Rb, Cd, Cs, Zn, Sr, Ra, may be readily removed by volatilization whereas Se, As, Te, Sb, In, which may form compounds with uranium are not necessarily removable by volatilization despite their high vapor pressures as pure substances above the melting point of uranium. Iodine and bromine are certain to form the compounds indicated which have sufficient volatility to permit ready removal.

Oxidation

Metals such as uranium and plutonium may be purified by slagging off the impurities by a small amount of controlled oxidation to form sufficient oxides to permit concentration of the baser elements in the oxide slag. The thermodynamic data are given in Table V. The rare earths, the alkaline earths, and zirconium may be expected to be removed by this method to greater or lesser extents, whereas niobium and the elements below it cannot be removed in the same manner. The quantitative calculation of the extents of removal in the favorable cases, especially for the close elements Zr, Pu, and Ba, cannot be made with the data of Table V because the necessary data on the free energies of solution of the elements in liquid uranium and of the oxides in solid UO or UO₂ are lacking. Only a small separation between uranium and plutonium is to be expected.

Comments

The above discussion indicates that some separation of uranium and plutonium from the fission products might be expected. However, the impurities in the uranium include oxides, nitrides, carbides, and perhaps others. While the amounts of these materials are low, they certainly will

Table II

NEGATIVE FREE ENERGIES OF FORMATION PER
GRAM ATOM OF FLUORINE, Kcal.

<u>T(°K) =</u>	<u>1000</u>	<u>1500</u>	<u>2000</u>
1/2SrF ₂	125.0	116.0	104.0
1/2BaF ₂	124.0	114.5	105.5
1/3LaF ₃	122.0	113.3	106.3
1/3CeF ₃	120.0	111.3	104.3
1/3NdF ₃	118.3	109.0	101.7
RbF	108.0	89.7	79.6
CsF	106.0	87.3	80.1
1/3PuF ₃	105.0	95.7	86.0
1/3ZrF ₃	100.0	93.3	89.0
1/3UF ₃	93.7	84.0	76.0
<hr/>			
1/5NbF ₅ ^a	77.2	73.8	70.6
1/2ZnF ₂	70.0	59.0	50.0
1/3GaF ₃	67.3	62.3	59.7
1/2CdF ₂	66.0	53.0	42.5
1/3SbF ₃	60.3	54.0	51.7
InF	60.0	66.0	71.0
1/6MoF ₆ ^a	55.3	49.5	44.0
CuF	46.0	41.0	46.0
1/2PdF ₂	44.5	43.5	36.5
1/6TeF ₆ ^a	39.0	32.0	24.8
1/2RhF ₂	38.5	?	?
AgF	37.6	36.3	44.6
1/6SeF ₆ ^a	27.3	20.1	12.0

Notes: a. May not be most stable valence state

b. Fluorides of Ge, Sn, Ru, Ir are much less stable than UF₃

Table III

NEGATIVE FREE ENERGIES OF FORMATION PER
GRAM ATOM OF CHLORINE, Kcal.

<u>T(°K) =</u>	<u>1000</u>	<u>1500</u>	<u>2000</u>
1/2BaCl ₂	84.0	74.5	67.5
RbCl	82.1	60.1	55.5
1/2SrCl ₂	81.0	73.5	63.5
CsCl	80.9	63.0	55.8
1/3CeCl ₃	70.3	68.3	60.7
1/3NdCl ₃	69.0	62.0	56.7
1/3LaCl ₃	68.3	62.3	57.0
1/3PuCl ₃	58.8	51.4	43.1
1/2ZrCl ₂	57.0	52.5	50.5
1/3UCl ₃	53.7	45.7	42.0
1/2ZnCl ₂	34.3	31.7	27.4
InCl	34.1	40.2	46.2
1/3GaCl ₃	32.0	29.7	27.0
1/2CdCl ₂	30.4	21.5	17.5
1/2SnCl ₂	27.8	26.6	25.1
1/4GeCl ₄ ^a	25.8	24.0	19.0
CuCl	22.0	20.0	18.0
AgCl	19.4	17.0	18.0
SeCl	16.4	20.4	14.3
1/3SbCl ₃	14.3	12.5	8.8
1/2MoCl ₂ ^a	9.0	5.0	4.5

Notes: a. May not be most stable valence state

b. Chlorides of Ru, Rh, Pd are much less stable than UCl₃

c. Data not available for Te, Nb chlorides

Table IV

VAPOR PRESSURE OF SOME ELEMENTS AND COMPOUNDS, Mm of Hg

<u>T(°K) =</u>	<u>1400</u>	<u>1500</u>	<u>1600</u>
Se, Kr, Xe	>> 10 ⁵	>> 10 ⁵	>> 10 ⁵
Rb	26,500	33,200	49,000
As	21,000	31,800	46,000
Cd	16,700	29,600	49,300
Cs	11,000	16,200	19,500
Zn	5,400	10,500	19,100
Te	980	1,860	3,400
UI ₃	140	330	710
Sr	125	265	550
Sb	34	105	290
Ba	22	42	112
UBr ₃	9.1	30.4	92
In	2.8	9.6	30
Sn	~ 0.5	~ 1	~ 3
Ag	~ 0.04	~ 0.4	~ 1.5
Ga		0.076	
In		0.72	
Ge		0.0076	

Table V

NEGATIVE FREE ENERGIES OF FORMATION PER
GRAM ATOM OF OXYGEN, Kcal.

<u>T(°K) =</u>	<u>1500</u>	<u>2000</u>	<u>2500</u>
1/3La ₂ O ₃	112.0	100.7	89.7
1/3Ce ₂ O ₃	107.0	94.0	83.0
1/3Nd ₂ O ₃	106.7	94.3	82.0
SrO	102.0	80.4	54.8
1/2UO ₂ (also UO)	98.9	88.5	78.5
1/2ZrO ₂	97.0	86.5	76.0
1/2PuO ₂	95.2	84.5	74.3
BaO	95.0	80.0	63.0
NbO	66.0	57.0	47.0
1/3Ga ₂ O ₃	49.0	35.7	28.7
ZnO	38.9	15.5	d.
1/2MoO ₂	38.0	30.0	22.0
1/3In ₂ O ₃	36.3	21.0	11.7
Cu ₂ O	12.0	6.6	1.3
CdO	11.8	d.	d.
Rb ₂ O	5.7	d.	d.
Cs ₂ O	3.6	d.	d.
Oxides of Ru, Rh, Pd, Ag	d.	d.	d.

Notes: a. d = Spontaneous decomposition to metal and oxygen

b. Estimated accuracy ± 2 kilocalories

c. Oxides of Ge, As, Se, Sn, Sb, Te are less stable than UO₂.

affect the separation of these impurities. The determination of what happens in liquation and slagging must be determined by experiment. The above data indicate the type of results that might be expected and the types of experiments of interest.

Application

The nature of the chemical metallurgical manipulations under discussion does not lead to the expectation that further development can achieve the kind of fission product decontamination now required to make possible non-remote handling of the end product. This circumstance strongly mitigates against the notion that these processes will find application in the processing of the fuel of any existent nuclear reactor.^(a) More likely the appropriate applications of such new processes will be found in connection with reactors planned for future building in which the remote handling and refabrication feature can be made integral with the over-all design.

Electrolytic Migration

A well known process for the purification of copper involves the electrolytic dissolution of an impure metal anode and the simultaneous deposition of the purified metal on a suitable cathode immersed in a copper solution. Purification occurs by two processes: (a) insoluble impurities are trapped in a bag around the anode and gaseous impurities are liberated and (b) the applied potential is so regulated that impurity metals less noble than copper, or if more noble having a greater overvoltage for deposition, are left behind in solution. This process, of course, cannot be directly applied to metals like uranium and plutonium since these metals cannot be electro-deposited from aqueous solution.

Zone Melting

Metallurgists have frequently improved the purity of metals by slowly withdrawing the molten metal from the furnace. Empirically it had been observed that impurities tended to segregate in the first and last portions to freeze and these could be readily cropped. Tin is an example. Many metals are purified to some extent by cropping the top of the ingot. This also removes blow holes.

(a) A chemical metallurgical operation might find application as a head-end step in connection with existent processes and reactors. Such an operation would serve to effect a bulk removal of fission products in concentrated form.

The principle involved is that dissolved impurities either raise or lower the melting point of a pure metal. Impurities which raise the melting point concentrate in the first portion to freeze while those that depress the melting point concentrate in the last portion. This may be illustrated for binary systems by use of the usual temperature composition diagrams, see Figure 1.

A process known as zone melting has been developed based on repeated melting and freezing of a small zone of a metal rod. The process is carried out by placing the rod inside of an induction coil and moving the coil, and the molten zone, slowly along the length of the rod. Thus, we may have repeated melting and freezing with a chance for the impurities to segregate in the two ends of the rod. The process has been applied to purifying germanium by the Bell Laboratories. The impurities which raise the melting point concentrate in the end initially melted while impurities which lower the freezing point concentrate in the far end.

If we take a hypothetical example as illustrated in Figure 2, we may indicate qualitatively the build up of the impurities. Suppose the dotted line on the composition diagram represents the limit of solubility of a given impurity, B, in the metal, A, then the full lines indicate the qualitative change in composition with repeated zone melting.

Application of zone melting to purification or removal of fission products from uranium has been tried at ANL. The tests run have been of the scouting type in an attempt to find the best rate of movement, bar size, temperature, etc.

Figure 1

LIQUID - SOLID EQUILIBRIUM DIAGRAMS

Figure 2

ZONE MELTING DIAGRAM

LECTURE 28

EFFLUENT CONTROL

References:

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Introduction

The effluents or wastes of nuclear reactors and their associated fuel fabrication and fuel separation and recovery plants may be classified into gaseous, particulate, liquid, and solid wastes. The particulate wastes, either liquid or solid, are carried by the gas streams. Problems involved with effluents are encountered with removal of this particulate matter from gas and cooling air streams, concentration and storage of liquid wastes, concentration and storage of solid wastes, and in the concentration of gaseous and liquid wastes by plant life, clays, and other materials.

The amount of reactivity that can be discharged from a plant depends on the dilution that can be obtained, the half-life of the particular isotope being discharged, and the possibility of utilization and concentration by plant and animal life. For example, relatively large amounts of A^{41} (1.8 hr) or Na^{24} (15 hr) might be released while I^{131} (8.14 d) and Sr^{90} (20 y) are more dangerous.

The limits for Pu and U^{233} are also very low since these are alpha emitters. The maximum permissible amounts of the alpha emitters were presented in Table VI of Lecture 10.

As might be expected, the various laboratories permit different amounts of activities in their liquid wastes insofar as dilution and isolation are considered. Some of the requirements are shown in Table I.

Table I

AMOUNTS OF ACTIVITIES IN LIQUID WASTES

<u>Site</u>	<u>Activity</u>	<u>Amount of Activity</u>
KAPL	α	$<1.70 \times 10^{-4} \mu\text{c/gal}$
Oak Ridge		$<5 \text{ curies/day}$
BNL		$<0.03 \mu\text{c/l}$ per batch and an average of $<0.03 \mu\text{c/l}$
Sr	$\beta - \gamma$	$<0.01 \mu\text{c/l}$
	α	$<0.001 \mu\text{c/l}$
Hanford	Pu	$<0.5 \mu\text{g/l}$ to cribs <u>plus</u>
	β	$<4 \mu\text{c/l}$
	Pu	$<0.0001 \mu\text{g/l}$ to ponds <u>plus</u>
	β	$<0.5 \mu\text{c/l}$

The maximum permissible concentration of certain isotopes in air and water is presented in Table II.

Table II

MAXIMUM PERMISSIBLE CONCENTRATIONS
OF CERTAIN RADIOISOTOPES¹

	<u>Permissible Concentration, $\mu\text{c}/\text{cm}^3$</u>	
	<u>In Air²</u>	<u>In Water³</u>
Fission Products, Mixed	1 $\times 10^{-9}$	-
Ra ²²⁶	8 $\times 10^{-12}$	4 $\times 10^{-8}$
Pu ²³⁹	2 $\times 10^{-12}$	1.5 $\times 10^{-6}$
U, natural	1.7 $\times 10^{-11}$ (25 $\mu\text{g}/\text{m}^3$)	-
I ¹³¹	4 $\times 10^{-9}$	3 $\times 10^{-5}$
H ³ as HTO	5 $\times 10^{-5}$	0.4
as HT	5 $\times 10^{-2}$	-
Sr ⁹⁰	2 $\times 10^{-10}$	8 $\times 10^{-7}$
Na ²⁴	-	8 $\times 10^{-3}$
P ³²	-	3 $\times 10^{-4}$
C ¹⁴ as CO ₂	1 $\times 10^{-6}$	-
Co ⁶⁰	7 $\times 10^{-9}$	1 $\times 10^{-5}$
A ⁴¹	1 $\times 10^{-6}$	-
Xe ¹³³	1 $\times 10^{-5}$	-
Xe ¹³⁵	3 $\times 10^{-6}$	-

¹"Handling Radioactive Wastes in the Atomic Energy Program"

²Based on 168 hr/wk

³Based on an intake of 2.5 l/day

Gases and Particulate Matter

There are three different kinds of gases which must be treated in nuclear energy work. These are the cooling air from air-cooled reactors using the once-through system, as in the X-10, BNL, and MTR reactors, ordinary ventilation air for such plants as fuel fabrication and fuel recovery cells, laboratories, hoods, etc., and off gases of chemical dissolvers. All of these air or gas streams may carry radioactive solids in the form of dusts or particulate matter. While minute liquid droplets might be present

in dissolver off gases, these are usually neglected since the water or solvents will usually evaporate and leave solid particulate residues. In any case, the means used to remove the solids from the particulate matter will also remove fogs.

The cooling air used in the X-10 and BNL reactors would be expected to carry radioactive dusts since the incoming air is dust laden, the graphite can give off dust particles, and it is possible for fuel slugs to fail and on oxidation of the U, transfer to the cooling air stream large amounts of particulate matter containing fission products and Pu. Normal ventilation air from fuel fabrication and chemical plants would be expected to carry very little radioactivity. However, in case of machining radioactive materials or Pu, the air may become heavily laden with radioactive particles. Off gases would be expected to contain large amounts of radioactive Xe, Kr, and I. The removal of I would require absorption while Xe and Kr could only be recovered by low temperature absorption. All reactor cooling air will contain large amounts of A^{41} .

Commercial filters, washers, and electrical precipitators have been developed for removal of dusts from stack gases, ventilation air, etc. Usually these are not adequate for final cleanup in the nuclear energy field since their efficiencies are too low. Frequently the particulate matter in the uncleaned air is far below the normal discharge level for commercial filters. Electrostatic precipitators frequently arc, or fail to give continuous satisfactory operation. For heavy loads, electrostatic precipitators have large throughputs and low pressure drops. The efficiency of an electrostatic filter may be high during trouble-free operation.

Cloth filters such as bag filters have large capacity and low pressure drops but low efficiencies for particles in the 0.5 to 5 micron range. They are very efficient for larger particles.

In order to be satisfactory for use in nuclear energy plants, a filter must remove at least 99.99% of the particulate matter. Special filters developed by the Chemical Warfare Service do this. Other requirements are low flow resistance, a minimum¹ of maintenance for continuous operation, a life measured in years, and ease of disposability. In some cases the filters must also be fireproof.

Three types of filters have been developed for use. These are the CWS #6, the fiberglass, and the sand filter. The sand filter as developed and used at Hanford consists of several layers of successively smaller grains of sand in a large underground chamber. The efficiency of collection is 99.7% of the submicron sized particles. The mechanism of removal of the particulate matter is believed to be dependent on the van der Waals' and residual ionic adsorption forces at the surface of the filter media since the particles being removed are much smaller than the average opening.

At low gas velocities, the transfer is believed to be one of diffusion, while at rates above 75 ft/min, the stagnant film becomes thinner and the centrifugal force of the particles tends to carry them through the films to the point at which absorption forces are operative. At very high gas velocities, particles are practically all removed by jets impinging on flat plates.

Fiberglass filters have been developed for fair efficiency, but the highest efficiency filters of the CWS type are made from asbestos. The CWS #6, developed during World War II, contains Bolivian asbestos as the active ingredient. AEC Filter #1 is equivalent and contains domestic asbestos.

Most installations make use of a preliminary inlet filter of the coarse fiberglass type such as used in household furnaces, followed by a finer filter, such as a 1-in. pad of fiberglass #25 backed by a 1-in. pad of #50 (American Filter Co.), both compacted together to 1 in. The exit filters are made up of the two fiberglass filters compacted into 1 in. and followed by a CWS #6 or AEC #1 filter bank. Such an installation has fairly high pressure drop, but two to three year life.

The specifications for the CWS #6 and AEC #1 filter papers are given in Table III.

Table III

SPECIFICATIONS FOR FINAL FILTERS

		<u>CWS' #6</u>	<u>AEC #1</u>
Thickness	in.	0.035 - 0.045	0.033 - 0.045
Minimum tensile strength in machine direction	lb/in.	7.3	> 1-1/2
Resistance at air velocity in 28 ft/min	mm H ₂ O	< 110	< 115
Penetration of 0.3 μ dioctyl phthalate smoke at 28 ft/min for one ply	%	< 0.12	< 0.15
Paper shall fold back on itself without surface rupture			

The filters are normally pleated and cemented in boxes which fit into welded steel frames. The filter boxes are sealed to the frames by rubber gaskets.

Ramsey and Cagle report the installation of a filter building and filters for cleaning the cooling air emitted from ORNL graphite reactor. The Oak Ridge installation utilizes the coarse filters followed by the CWS #6 fine filters. The coarse filters are removed through removable plugs on the top of the building by long tongs and placed in a water canal for outside removal.

In installations where Pu or U^{233} are handled as the metal, particular care must be exercised. Pu forms an adherent oxide coating which may break down into a white oxide which is not adherent. The white oxide readily forms aerosols of very small particle size. Essentially every particle must be removed before the air is discharged. The half lives of natural U and Th are so great that these can be used in the open, but any work on Pu or U^{233} must be done in hoods. Air circulated through hoods should be filtered at the inlet and outlet of the hood, and again at a sufficient distance away so that possible explosions or fires in the hoods will not affect the final filters.

The filters in a hood system must be changed with care. The outside filters may be changed by placing a second filter on top of the first and pushing the contaminated filter into the hood. The filter is placed in a contaminated plastic bag which is attached to the hood for solid removal. After the bag has been filled it is heat sealed electronically and torn off through the heat seal leaving the top of the bag attached to the hood and sealed off. A new bag is placed over the old top and the top disconnected and dropped into the new bag. Great care must be exercised in design of Pu and U^{233} hoods and ventilation systems as the oxides will penetrate very minute holes.

The discharge of off gases has been discussed earlier. The I is the only gas of great concern. Its concentration is followed in plant life near Hanford, for example. If required, it can be removed by absorption by $AgNO_3$.

Reactor cooling air containing A^{41} and off gases containing large amounts of Xe and Kr are normally discharged through high stacks. The dispersal is usually given by Sutton's equation. However, actual dispersal depends on the meteorological conditions. When there is a wind over a few miles per hour with turbulent flow or strong lapse conditions, released gases will be dispersed satisfactorily. Under lapse conditions the air at the surface of the earth is warmer than it is at higher levels. Smoke rises and mixes with the air. Under inversion conditions, the air is cooler at the ground than it is higher up. Smoke tends to leave a stack and float away in a straight line or to hang over the area. Under inversion conditions, discharged radioactivity may be carried in a fairly concentrated stream. When the inversion breaks up, the gas may be brought to the ground in concentrated form.

Particulate matter discharged within a gas stream settles out by gravity. The rate of settling may be such that the activity is spread over large areas or, under an inversion, much of it may settle out in the neighborhood of the stack.

Liquids

The great bulk of the waste products containing the fission products are present in the liquid solutions from the chemical separation plants. The amount of total reactivity present in fission products is enormous. Murray has calculated that if a man is exposed to one pound of fresh fission products, say 0.1 day old, at 10 feet, he will receive a fatal dose of gamma rays in about 10^{-8} second.

The liquid wastes are normally concentrated and stored. If means were available to separate the long lived fission products, the amount of storage over three years could be reduced to a minimum. However, to date no means of separation have been developed and the fission products and salting agents are concentrated and stored. In some of the proposed separation procedures, such as the pyrometallurgical processes, the volumes to be stored might be greatly reduced and the fission products used for irradiation of foods, production of chemicals, etc.

Rodgers gives Table IV which presents the accumulation of long lived fission products. In this table eight of the most important long lived isotopes are shown with their accumulation over 50 years, their ultimate concentration, and the time necessary to reach this concentration based on the assumption of 10 kg/da production for the period. This would be equivalent to 9000 Mw of heat, or perhaps 3000 Mw of electrical energy. The data are plotted in Figure 1.

Table V gives the dispersal volumes required for tolerance, assuming that a large part of the electrical energy of the country is generated from nuclear energy, or three tons of fission products per day are produced. The purpose of this table is to indicate the impractical nature of many of the suggestions that have been made to dispose of the fission products. In spite of all these suggestions, separation, concentration, and storage appear now to be the only practical methods of handling the wastes.

Sr-90 is the controlling isotope.

If we produce three tons of fission products per day, we may assume a 2% burn up of the fuel. This means that only 150 tons per day of uranium must be processed. Some of the present processes produce of the order of 1000 gallons of waste per ton of uranium processed. This means that 150,000 gallons of waste would be produced per day. If this is to be stored unprocessed at, say, \$1.00 per gallon, the cost would be $\$5.5 \times 10^7/\text{yr.}$, which is high but not prohibitive.

Table IV

ACCUMULATION OF LONG-LIVED FISSION PRODUCTSBasis: 10 Kg Fission Products/Day $\approx$ 9000 MW Installed Reactor Capacity

Isotope	Fission Yield	Half Life	Decay Constant	Ultimate Quantity FP at Equilibrium		Accumulation of FP Activity in Curies At Time Indicated				
	%			Kg	Curies	10 y	20 y	30 y	40 y	50 y
Zr ⁹⁵	6.4	65 d	1.23×10^{-7}	25	5.2×10^8	5.2×10^8	5.2×10^8	5.2×10^8	5.2×10^8	5.2×10^8
Ce ¹⁴⁴	5.3	280 d	2.86×10^{-8}	133	4.3×10^8	4.3×10^8	4.3×10^8	4.3×10^8	4.3×10^8	4.3×10^8
Ru ¹⁰⁶	0.5	1.0 y	2.2×10^{-8}	12	4.1×10^7	4.1×10^7	4.1×10^7	4.1×10^7	4.1×10^7	4.1×10^7
Pm ¹⁴⁷	2.6	2.6 y	8.46×10^{-9}	225	2.1×10^8	1.9×10^8	2.1×10^8	2.1×10^8	2.1×10^8	2.1×10^8
Sr ⁹⁰	5.3	19.9 y	1.1×10^{-9}	2150	4.3×10^8	1.3×10^8	2.2×10^8	2.8×10^8	3.2×10^8	3.5×10^8
Cs ¹³⁷	6.2	33 y	6.65×10^{-10}	6350	5.0×10^8	9.5×10^7	1.7×10^8	2.3×10^8	2.8×10^8	3.3×10^8
Tc ⁹⁹	6.2	2.1×10^5 y	1.05×10^{-13}	2.9×10^7	5.0×10^8	16,500*	33,000	49,500	66,000	82,500
Pu ²³⁹	---	2.4×10^4 y	---	---	---	2,300**	4,600	6,900	9,200	11,500

*Decay neglected.

**Loss of 0.1% assumed on processing Decay neglected.

Figure 1

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Table V

50-YEAR ACCUMULATION OF LONG-LIVED ISOTOPES
AND REQUIRED DISPERSAL AREAS

Basis: 2.2×10^6 MW Installed Reactor Capacity $\approx$ 3 Tons Fission Products/Day

Isotope	Accumulated Quantity in 50 years, curies	Maximum Permissible Concentrations in		Volume Required to Dilute to Tolerance	
		Water c/ml	Air c/ml	Water cubic miles	Air cubic miles
Zr ⁹⁵	1.3×10^{11}	4×10^{-3}	4×10^{-7}	7.8×10^3	7.8×10^7
Ce ¹⁴⁴	1.1×10^{11}	4×10^{-2}	7×10^{-9}	6.6×10^3	3.8×10^9
Ru ¹⁰⁶	1.0×10^{11}	0.1	3×10^{-8}	2.4×10^2	8×10^8
Pm ¹⁴⁷	5.1×10^{10}	1	2×10^{-7}	12	6×10^7
Sr ⁹⁰	8.6×10^{10}	8×10^{-7}	2×10^{-10}	2.6×10^7	1×10^{11}
Cs ¹³⁷	8.1×10^{10}	1.5×10^{-3}	2×10^{-7}	1.3×10^4	9.7×10^7
Tc ⁹⁹	2.0×10^7	3×10^{-2}	3×10^{-6}	0.2	1.6×10^3
Pu ²³⁹	2.8×10^6	1.5×10^{-6}	2×10^{-12}	4.5×10^2	3.4×10^8

This volume can be reduced, of course, by selecting processes which produce a smaller waste volume initially, or by several concentrating techniques. There will be, however, an irreducible volume which must be accepted. Taking as the most optimistic assumption that the inert salts can be held to a weight equal to the fission products themselves, six tons per day of concentrate will be formed. At a density of 100 lb/cubic foot the volume formed is 120 cubic feet/day; 44,000 cubic feet/year. This is not an impractical number. Even if storage space for this volume cost \$50 per cubic foot, the storage bill would be only \$2,000,000 per year. To this would have to be added, of course, the much larger cost of concentrating, plus the costs of inspecting and maintenance of storage. As concrete in stainless steel tanks would be used, maintenance should be low.

Several processes may be used in concentrating the solutions. The most important is, of course, evaporation. Decontamination factors at practical rates of operation are 10^4 . This varies with the concentration in the feeds, the foaming of the concentrated solution, etc. Costs range from 5 to 10 cents per gallon processed. Only single effect evaporations have been used. There is no incentive to get really high decontamination factors as ion exchange can be used to remove the traces carried over in the distillate and give a very low activity effluent.

Packed glass wool removes most of the activity from the steam probably as droplets, but bubble caps, etc., are not efficient decontaminant devices.

Evaporation is usually carried to the point that most of the residue is precipitated solids. These may be barreled for shipment or stored in tanks.

One of the important problems is that of recovery or removal of the salting agents from the concentrated solutions. If this could be done cheaply, the bulk could be reduced. Recrystallization of $\text{Al}(\text{NO}_3)_3$ or NH_4NO_3 has been tried, but not with great success. HNO_3 can be removed by distillation.

Ion exchange resins are useful on very low solid content feeds. The capacity of the resins and the concentration factor obtainable vary inversely with the solid content of the feed. Cation resins will give decontamination factors of about 50, mixed beds of about 10^5 .

As evaporator condensates contain only a trace of solid materials, the ion exchange resins are reasonably cheap. Cost for low solid feed is < \$0.01/gallon.

Flocculation has been studied and decontamination factors of 10 have been obtained. This is too low to be of interest on chemical plant wastes but is of interest in water treatment plants which may have to use water contaminated by bomb fall out. The advantage of flocculation is that it will operate at very low costs on feeds containing various amounts of solids.

Electrodeionization is similar to ion exchange except that it uses selective membranes and continuously regenerates the membrane by an electric current rather than by chemicals. Development is still under way on applications of the process.

The storage of liquids will always be hazardous, even though nearly every conceivable precaution is taken. Sr^{90} for 10 half lives will require over 200 years storage. Corrosion, earthquakes, floods, combat, etc., might cause loss. If storage of liquid wastes is rejected, then storage as solids will be required. Argonne and others have demonstrated batch and pilot scale methods of solid storage.

Wastes have been concentrated to the point that they solidify on cooling and the solids stored in drums. While this is an improvement, non-sealed or broken drums are subject to leaching. Two hundred years might see a percentage of the drums eroded through.

Wastes may be immobilized by addition of Portland cement at a volume increase of two. The concrete certainly would be leachable. Absorbing the wastes on clays or on glass or ceramic forming substances and vitrifying them appears more promising. Some reasonable volume of unleachable material might be found.

If precipitation or complete separation of those fission products which have relative long lives could be worked out, then vitrifying and permanent storage for several hundred years appears practical. The fission products of most interest are in Table VI. The short-lived products are presented in Table VII. They would decay prior to chemical separation, but would be present in pyrometallurgical separations processes, because of the shorter decay period prior to separation. For interest, some of the fission products initially formed are presented in Table VIII. To get a true picture, the fission product decay chains must be studied.

Much work has been done recently on the utilization of fission products for pasteurization and sterilization of foods and the manufacture of chemicals. The concentrations of radioactivity required for a practical rate of production are large. For example, to sterilize a can of food requires from 1 to 2×10^6 roentgen of gamma rays.

There are several sources of gamma rays. These include irradiated enriched U fuel elements, irradiated natural or slightly enriched U, and the separated fission products. Since irradiated fuel must "cool" or undergo radioactive decay for a period of months before it is used in the present extraction plants, it would be desirable to use this fuel element during this decay period as a source of gamma irradiation. The use of irradiated natural or slightly enriched U is impractical because of the self absorption of the U due to its high density. The use of irradiated, highly enriched fuel

Table VI

FISSION PRODUCTS OF INTEREST IN SEPARATIONS

<u>Fission Product</u>	<u>Fission Yield, %</u>	<u>Half-Life</u>
Ru 103	3.0	45 d
Ru 106 - Rh 106	0.5	290' d
Zr 95 - Nb 95	6.2	65 d
Nb 95	6.2	38.7 d
Ce 141	6.1	28 d
Ce 144 - Pr 144	5.2	275 d
Kr 85	0.33	9.7 y
Sr 89	4.7	54 d
Sr 90 - Y 90	5.2	25 y
Y 91	5.6	57 d
Te 129m - Te 129	0.4	32 d
Cs 137 - Ba 137	6.1	33 y
Ba 140	6.2	12.8 d
La 140	6.2	40 h
Pr 143	5.6	13.8 d
Nd 147	3.0	11.8 d
Pm 147	3.0	3.7 y

Table VII

SHORT-LIVED FISSION PRODUCTS

<u>Fission Product</u>	<u>Half-Life</u>	<u>Fission Yield, %</u>
Sr 91	9.7 h	5.6
Zr 97	17.0 h	6.1
Ru 105	4.4 h	1.0
I 131	8 d	3.0
Xe 133	5.3 d	4.7

Table VIII
SLOW FISSION YIELDS
 (From AECD-2817)

Mass No.	U^{233}	U^{235}	Pu^{239}	Activity	Half- Life
72		1.5×10^{-5}	11×10^{-5}	Zn	49 h
73		10^{-4}		Ga	5 h
77	0.018	0.0037		Ge	12 h
		0.0091		As	40 h
(78)		0.020		As	90 m
		0.020		Ge	2.1 h
81		0.008		Se	59 m
		0.125		Se	17 m
83		0.21	0.073	Se	25 m
	0.70	0.4		Be	2.4 h
84		0.65		Br	30 m
89	5.6	4.6	1.76	Sr	55 d
91		5.0	2.14	Sr	9.7 h
		5.9	2.86	Y	57 d
(92)		5.1		Sr	2.7 h
(94)		5		Y	20 m
95	5.7	6.4	5.57	Zr	65 d
97		6.1	5.17	Zr	17 h
99	4.7	6.2	5.93	Mo	67 h
103	0.85	3.7	5.56	Ru	42 d
105		0.9		Ru	4.5 h
			3.76	Rh	36.5 h
106	0.24	0.48	4.76	Ru	1 y
109	0.046	0.028	1.04	Pd	13.5 h
111	0.022	0.018	0.275	Ag	7.5 d
112	0.014	0.011	0.103	Pd	21 d

Table VIII (Contd.)

Mass No.	U^{233}	U^{235}	Pu^{239}	Activity	Half-Life
115	0.017	0.012	0.048	Cd	2.3 d + 44 d
117		0.01		Cd	2.83 h
121	0.02	0.014	0.041	Sn	26.h
125	0.03	0.012		Sn	9 d
125		0.024		Sb	2.7 y
(126)		0.1		Sn	70 m
127	$\left. \begin{matrix} 0.092 \\ 0.023 \end{matrix} \right\} 0.115$	$\left. \begin{matrix} 0.13 \\ 0.03 \end{matrix} \right\} 0.16$	0.358	Sb	93 h
129		$\left. \begin{matrix} 0.24 \\ 0.19 \end{matrix} \right\} 0.43$		Sb I	4.2 h 32 d
131	2.7	2.8	3.61	I	8 d
(132)		3.6	4.90	Te	77 h
133		4.5	4.95	I	22 h
(134)		5.7		I	54 m
135		5.9		Xe	9.2 h
		5.6	5.54	I	6.7 h
139		6.3	5.19	Ba	86 m
140	6.0	6.1	4.90	Ba	12.8 d
141		5.7	4.41	Ce	28 d
143		5.4	4.33	Ce	33 h
144	3.4	5.3	3.41	Ce	275 d
147		2.6		Nd	11 d
(149)		1.4		61	47 h
153	0.078	0.15	0.384	Sn	47 h
155		0.03		Eu	2 y
		0.031		Sm	25 m
156		0.013	0.126	Eu	15.4 d
		0.016		Sm	10 h
(157)		0.0074		Eu	15.4 h
(158)		0.002		Eu	60 m

elements such as the Al-U used in the MTR produces the most practical source. Gamma sources made up of several such fuel elements are being used in development work. The intensity of the irradiation is over 1×10^6 r per hour.

Most of the discussion has centered around the use of separated fission products. The wastes from the present separation plants which contain the fission products are too dilute to be of much practical value. However, fairly good sources have been made up by concentration of the wastes. The use of pyrometallurgical and volatility processes are expected to produce very concentrated wastes which will produce high gamma concentrations. Such processes are in early stages of development at present but the results obtained are favorable. Practical methods of chemically separating the fission products have not been worked out. Ion exchange has given some concentration.

The concentration of gamma rays produced in the reactor during the fission process is very high but the practical use is complicated by the neutrons present. It would appear possible to use a H_2O -B thermal shield and still have a very high gamma concentration with a low neutron concentration for chemical manufacture. Many chemical reactions could be aided by either gamma or neutron irradiation. The use of a reactor for these reactions appears to be favorable. There are actually hundreds of reactions that could be considered. Henley presents a number of reactions with the literature references.

Solids

Solid wastes may be classified into two groups. These are equipment and non-equipment items. By equipment, we mean tanks, pumps, etc., removed from separation plants, etc., but which have sufficient activity that they cannot be placed in the regular commercial scrap outlets. The non-equipment items include contaminated rags, paper, and general packaging. Much of this material is combustible. Precipitates that occur in liquid solutions are considered a part of the liquid wastes.

Disposition of equipment/items is usually open storage or burial. If equipment is contaminated only to a low level and the equipment is made of metal or even wood, burial in an isolated area or open storage may be adequate.

The combustible material is burned in special incinerators that collect the ash in a plastic bag under water. When the bag is filled, the ash is dewatered for permanent storage. The combustion gases are scrubbed to remove the large particles of fly ash and to cool the gas. The gas is further scrubbed by mixing a stream of water with it and separating in centrifugal separators. Finally, the offgas is heated slightly to evaporate any droplets present and prevent condensation. The gas is then filtered and exhausted to the stack. After exchange of filters, the old ones are burned in the incinerator.

APPENDIX A

The following references were included in the original classified lectures, but were omitted from the declassified lectures because they were not commonly available.

Lecture 1 - Design Considerations

None

Lecture 2 - Thermal vs. Fast Reactors

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Lecture 3 - Light and Heavy Water Cooled Reactors

MTR

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Lecture 5 - Homogeneous Reactors

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Lecture 6 - Industrial Study Groups

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Lecture 7 - Boiling Reactors

None

Lecture 8 - Fluid Flow

None

Lecture 9 - Heat Transfer

None

Lecture 10 - Materials

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Lecture 11 - Preparation of Pure Materials

None

Lecture 12 - Preparation of Alloys - Phase Diagrams

None

Lecture 13 - Preparation of Fuel Elements

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Lecture 15 - Radiation Damage

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