# *Part I*
# AQUEOUS HOMOGENEOUS REACTORS

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## PREFACE

This compilation of information related to aqueous homogeneous reactors summarizes the results of more than ten years of research and development by Oak Ridge National Laboratory and other organizations. Some 1500 technical man-years of effort have been devoted to this work, the cost of which totals more than $50 million. A summary of a program of this magnitude must necessarily be devoted primarily to the main technical approaches pursued, with less attention to alternate approaches. For more complete coverage, the reader is directed to the selected bibliography at the end of Part I.

Although research in other countries has contributed to the technology of aqueous homogeneous reactors, this review is limited to work in the United States. In a few instances, however, data and references pertaining to work carried on outside the United States are included for continuity.

Responsibility for the preparation of Part I was shared by the members of the Oak Ridge National Laboratory as given on the preceding page and at the beginning of each chapter.

Review of the manuscript by others of the Oak Ridge Laboratory staff and by scientists and engineers of Argonne National Laboratory and Westinghouse Electric Corporation have improved clarity and accuracy. Suggestions by R. B. Briggs, director of the Homogeneous Reactor Project at the Oak Ridge Laboratory, and S. McLain, consultant to the Argonne Laboratory, were particularly helpful.

Others at Oak Ridge who assisted in the preparation of this part include W. D. Reel, who checked all chapters for style and consistency, W. C. Colwell, who was in charge of the execution of the drawings, and H. B. Whetsel, who prepared the subject index.

Oak Ridge, Tennessee                                    James A. Lane, *Editor*
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CHAPTER 1

# HOMOGENEOUS REACTORS AND THEIR DEVELOPMENT*

## 1-1. BACKGROUND†

**1-1.1 Work prior to the Manhattan Project.** Nuclear reactors fueled with a solution or homogeneous mixture of fuel and moderator were among the first nuclear systems to be investigated experimentally following the discovery of uranium fission. In fact, it was only slightly more than a year after this discovery that Halban and Kowarski at the Cavendish Laboratory in England performed experiments which indicated to them that a successful self-sustaining chain reaction could be achieved with a slurry of uranium oxide ($U_3O_8$) in heavy water.

In these experiments, reported in December 1940 [1], 112 liters of heavy water mixed with varying amounts of $U_3O_8$ powder were used inside an aluminum sphere 60 cm in diameter, which was immersed in about one ton of heavy mineral oil to serve as a reflector. (Mineral oil was chosen to avoid contamination of the $D_2O$ in case of a leak in the sphere.) By measuring neutron fluxes at varying distances from a neutron source located in the center of the sphere, Halban and Kowarski calculated a multiplication factor of $1.18 \pm 0.07$ for this system when the ratio of deuterium atoms to uranium atoms was 380 to 1, and $1.09 \pm 0.03$ when the D/U ratio was 160 to 1.

Other experiments conducted at the same time by Halban and Kowarski [1]‡, using $U_3O_8$ and paraffin wax, indicated that with a heterogeneous lattice arrangement it would be possible to achieve multiplication factors as high as 1.37 in a system containing about 100 atoms of deuterium per atom of uranium.

It is interesting to note that the $D_2O$ supply used in the experiments had been evacuated from France. The $D_2O$ originally came from the laboratories of the Norwegian Hydroelectric Company, and with the destruction of this plant and its $D_2O$ stockpile in 1942, this was the sole remaining supply of purified $D_2O$. However, it was not enough to allow a self-sustaining chain reaction to be established with natural uranium.

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†This section is based on material supplied by W. E. Thompson, Oak Ridge National Laboratory.
‡See the list of references at the end of the chapter.

Even earlier (in 1939) Halban and Kowarski, as well as other experimentalists, had fairly well established that self-sustaining chain reactions with $U_3O_8$ and ordinary water are not possible [2,3,4]. Homogeneous systems of uranium with carbon, helium, beryllium, or oxygen were also considered, and were rejected as not feasible either for nuclear, chemical, or engineering reasons.

In November 1942, Kowarski, with Fenning and Seligman, reported more refined experiments which led to the conclusion that neither homogeneous nor heterogeneous mixtures of $U_3O_8$ with ordinary water would lead to self-sustaining chain reactions, the highest values of the multiplication factor being 0.79 for the homogeneous system and 0.85 for the heterogeneous system.

Because it was clear even by early 1942 that the only feasible homogeneous reactor using natural uranium would be one moderated with $D_2O$, and because no $D_2O$ was available at that time for use in reactors, interest in homogeneous reactor systems was purely academic. The atomic energy program, which was then getting well under way, devoted its attention to heterogeneous reactors.  By using a heterogeneous lattice arrangement with a core of uranium metal slugs spaced inside graphite blocks and a periphery containing $U_3O_8$ slugs (used after the supply of uranium metal ran out) spaced inside the graphite, the first successful self-sustaining chain reaction was achieved on December 2, 1942.

**1–1.2 Early homogeneous reactor development programs at Columbia and Chicago universities.**   Interest in homogeneous reactors lagged until early in 1943, when it became clear that American and Canadian efforts to produce large quantities of heavy water would be successful. At that time the group under H. C. Urey at Columbia University directed its attention to the development of slurried reactors utilizing uranium oxide and $D_2O$.

In March 1943, Urey and Fermi held a conference to review the situation with respect to homogeneous reactors.  They noted the value of 1.18 that Halban and Kowarski had obtained for the multiplication factor in a $U_3O_8$–$D_2O$ slurry reactor and pointed out that the value calculated from theory was only 1.02.  They realized, however, that neither the theory nor the experiment was free from serious objections, and that insufficient data were available to allow a trustworthy conclusion to be reached as to the feasibility of homogeneous systems.

If the results of Halban and Kowarski were correct, then a homogeneous system containing a few tons of heavy water would be chain reacting.  On the other hand, if the theoretical estimates were correct, the order of 100 tons of $D_2O$ would be required.

Urey and Fermi recommended [5] that the earlier $U_3O_8$–$D_2O$ experiments be repeated with the improved techniques then known, and that

consideration be given to incorporating a mixture of uranium and heavy water into the pile at Chicago to determine its effect on the pile reactivity.

From the theoretical considerations of E. P. Wigner and others, it appeared that the most favorable arrangement for a $U_3O_8$–$D_2O$ reactor would be one in which the slurry was pumped through a lattice of tubes immersed in $D_2O$ moderator. This was especially true because the neutron absorption cross section assigned to heavy water at that time made it appear that more than 200 tons of $D_2O$ would be required to reach criticality in an entirely homogeneous system in which the $U_3O_8$ and moderator were mixed. With a heterogeneous system it seemed likely that a much smaller quantity of $D_2O$ would suffice and every effort was directed toward preparing a design that would require about 50 tons of $D_2O$ [6].

It was estimated by E. P. Wigner that the uranium concentration in the slurry would have to be 2.5 to 3 grams per cubic centimeter of slurry. It became apparent immediately that no aqueous solution of a uranium compound could be made with such a density. With pure $UF_6$, 2.48 grams of uranium per cubic centimeter could be obtained, and piles utilizing this compound were considered. However, the corrosion problems in such a system were believed to be so severe that the development of a reactor to operate at a high power level would be extremely difficult, if not impossible.

Other compounds, such as uranyl nitrate dissolved in $D_2O$, were excluded because in the case of nitrate the neutron absorption of nitrogen was too high and in other cases sufficient densities could not be obtained. Thus the initial phase of the research at Columbia was directed toward the development of high-density slurries [6].

The reactor visualized by the Columbia group was one in which an extremely dense suspension of uranium in $D_2O$ would be pumped through a large number of pipes arranged inside a heavy-water moderator. It was planned that both the slurry and the moderator would be circulated through heat exchangers for cooling [6].

Then, in July of 1943, the experiments of Langsdorf [7] were completed, giving a much lower cross section for deuterium than was known earlier. As a result, the homogeneous reactor became much more attractive, since the critical size (neglecting external holdup) could then be reduced to about 30 tons of $D_2O$ with about 6 tons of uranium as oxide in an unreflected sphere [8]. This favorable development allowed emphasis to be shifted to less dense slurries, greatly simplifying the problems of maintaining a suspension of dense slurry, pumping it, and protecting against erosion. Experiments were directed toward developing a reactor design which would permit operation without continuous processing of the slurry to maintain its density [6].

By the end of 1943 preliminary designs had been developed at the University of Chicago Metallurgical Laboratory for several types of heavy-

water reactors, all using slurry fuel but differing in that one was completely homogeneous [9], one was a light-water-cooled heterogeneous arrangement [10], and another was a $D_2O$-cooled heterogeneous reactor [11]. These reactors were proposed for operation at power levels of 500 Mw or more (depending on external power-removal systems) and were intended as alternates to the Hanford piles for plutonium production in case satisfactory operation of the graphite-natural uranium, water-cooled piles could not be achieved.

At this point one might ask why it was that homogeneous *solution* reactors were not given more serious consideration, especially in view of the newly discovered cross section for deuterium, which permitted considerably lower concentrations of uranium. The answer is that the only known soluble salts of uranium which had a sufficiently low cross section to enable the design of a reactor of feasible size and $D_2O$ requirement were uranyl fluoride and uranium hexafluoride. (Enriched uranium was not then available.) These were considered, but rejected principally because of corrosion and instability under radiation. A second factor was the evidence that $D_2O$ decomposition would be more severe in a solution reactor where fission fragments would be formed in intimate contact with the $D_2O$ rather than inside a solid particle as in the case of a slurry.

Research on homogeneous reactors was undertaken at Columbia University in May 1943, and continued with diminishing emphasis until the end of 1943, at which time most of the members of the homogeneous reactor group were transferred to Chicago, where they continued their work under the Metallurgical Laboratory.

At the Metallurgical Laboratory, the principal motivation of interest in homogeneous reactors was to develop alternate plutonium production facilities to be used in the event that the Hanford reactors did not operate successfully on a suitable large scale, and studies were continued through 1944. With the successful operation of the Hanford reactors, however, interest in homogeneous plutonium producers diminished, and by the end of 1944 very nearly all developmental research had been discontinued. The results of this work are summarized in a book by Kirschenbaum [12].

**1–1.3 The first homogeneous reactors and the Los Alamos program.** During the summer of 1943 a group at Los Alamos, under the leadership of D. W. Kerst, designed a "power-boiler" homogeneous reactor, having as its fuel a uranyl sulfate-water solution utilizing the enriched uranium which was expected to become available from the electromagnetic process. However, this design was put aside in favor of a low-power homogeneous reactor designed by R. F. Christy. The low-power homogeneous reactor was built and used during the spring and summer of 1944 for the first of a series of integral experiments with enriched material (see Chapter 7).

There were two reasons for choosing $UO_2SO_4$ instead of uranyl nitrate as the fuel: there is less neutron absorption in the sulfate than in the nitrate, and the sulfate was thought to be more soluble. The latter reason was considered important because it was feared that with the maximum-enrichment material from the electromagnetic process, it might be difficult to dissolve the critical mass in the desired volume [13]. These objections to the use of uranyl nitrate, however, were subsequently found to be invalid.

After gaining experience in operating the low-power reactor, "LOPO," the Los Alamos group revised its plans for the higher power homogeneous reactor, known as the "HYPO," and after extensive modification of the design, the reactor was built and put into operation in December 1944 with uranyl nitrate as the fuel.

In April 1949, rather extensive alterations to the HYPO were begun in order to make the reactor a more useful and safer experimental tool. The modified reactor, known as "SUPO," is still in operation. The present SUPO model reached local boiling during initial tests, due to the high power density. A slight increase in power density above the design level produces local boiling between cooling coils, even though the average solution temperature does not exceed 85°C.

Interest in solution reactors continued at Los Alamos, and improved designs of the Water Boiler (SUPO Model II) were proposed [14]. These, however, have not yet been constructed at Los Alamos, although similar designs have been built for various universities [15].

The work on water boilers at Los Alamos led to the design of power reactor versions as possible package power reactors for remote locations. Construction of these reactors, known as Los Alamos Power Reactor Experiments No. 1 and No. 2 (LAPRE–1 and LAPRE–2), started in early 1955. To achieve high-temperature operation at relatively low pressures, LAPRE–1 and –2 were fueled with solutions of enriched uranium oxide in concentrated phosphoric acid. The first experiment reached criticality in March 1956 and was operated at 20 kw for about 5 hr. At that time radioactivity was noted in the steam system, and the reactor was shut down and dismantled. It was discovered that the gold plating on the stainless steel cooling coils had been damaged during assembly and the phosphoric acid fuel solution had corroded through the stainless steel. The cooling coils were replaced and operations were resumed in October 1956. However, similar corrosion difficulties were encountered, and it was decided to discontinue operations. In the meantime, work on LAPRE–2 continued, and construction of the reactor and its facilities was completed during the early part of 1958. The details of these reactors are given in Chapter 7.

**1–1.4 Early homogeneous reactor development at Clinton Laboratories (now Oak Ridge National Laboratory).** With the availability of enriched uranium in 1944, the possibility of constructing a homogeneous reactor became more attractive because, by using enriched uranium, the $D_2O$ requirement could be greatly reduced, or even ordinary water could be used. The chemists at Clinton Laboratories (now ORNL), notably C. D. Coryell, A. Turkevich, S. G. English, and H. S. Brown, became interested in enriched-uranium homogeneous reactors primarily as a facility for producing other radioisotopes in larger amounts, and a number of reports on the subject were issued by various members of the Chemistry Division (D. E. Koshland, Jr., W. J. Knox, and L. B. Werner).

In August 1944 Coryell and Turkevich prepared a memorandum [16] recommending the construction of a 50-kw homogeneous reactor containing 5 kg of uranium enriched to $12\frac{1}{2}\%$ $U^{235}$ or about 500 g of plutonium. The fuel proposed was to be in the form of salt solution in ordinary water. The following valuable uses of such a reactor were listed in this memorandum and enlarged upon in a later memorandum by Coryell and Brown [17]:

(1) The preparation of large quantities of radioactive tracers.

(2) The preparation of intense radioactive sources.

(3) Studies in the preparation and extraction of $U^{233}$.

(4) The preparation of active material for Hanford process research.

(5) Study of chemical radiation effects at high power levels.

(6) Accumulation of data on the operating characteristics, chemical stability, and general feasibility of homogeneous reactors.

The physicists were also interested in the homogeneous reactor, particularly as a research facility which would provide a high neutron flux for various experimental uses. The desirability of studying, or demonstrating, if possible, the process of breeding had been made especially attractive by the recent data indicating that $U^{233}$ emitted more neutrons for each one absorbed than either $U^{235}$ or $Pu^{239}$, and the physicists were quick to point out the possibility of establishing a $U^{233}$-thorium breeding cycle which would create more $U^{233}$ from the thorium than was consumed in the reactor. These potentialities were very convincingly presented in November 1944 by L. W. Nordheim in a report entitled "The Case for an Enriched Pile" (ORNL–CF–44–11–236).

The power output of such a breeder with a three-year doubling time is about 10,000 kw, and this was established as a new goal for the homogeneous reactor. The reactor, then, was conceived to be a prototype homogeneous reactor and thermal breeder; in addition, it was conceived as an all-purpose experimental tool with a neutron flux higher than any other reactor.

Work on the 10,000-kw homogeneous reactor was pursued vigorously through 1945; however, at the end of that year there were still several

basic problems which had not been solved. Perhaps the most serious of these was the formation of bubbles in the homogeneous solution. These bubbles appear as a result of the decomposition of water into hydrogen and oxygen by fission fragments and other energetic particles. Because the bubbles cause fluctuations in the density of the fuel solution, they make it difficult to control the operating level of the reactor. Nuclear physics calculations made at the time indicated that under certain conditions it might be possible to set up a power oscillation which, instead of being damped, would get larger with each cycle until the reactor went completely out of control. Minimizing the bubble problem by operating at elevated temperature and pressure was not considered seriously for two reasons: first, beryllium, aluminum, and lead were the only possible tank materials then known to have sufficiently low neutron-absorption characteristics to be useful in a breeder reactor. Of these metals, only lead was acceptable because of corrosion, and lead is not strong enough to sustain elevated temperatures and high pressures. Second, there had been essentially no previous experience in handling highly radioactive materials under pressure, and consequently the idea of constructing a completely new type of reactor to operate under high pressure was not considered attractive.

Other major unsolved problems at the end of 1945 were those of corrosion, solution stability, and large external holdup of fissionable material. Because it appeared that the solution of these problems would require extensive research and development at higher neutron fluxes than were then available, it was decided to return to the earlier idea of a heterogeneous reactor proposed by E. P. Wigner and his associates at the Metallurgical Laboratory. Experimental investigations in this reactor, it was hoped, would yield data which would enable the homogeneous reactor problems to be solved. The extensive effort on this latter reactor (later built as the Materials Testing Reactor in Idaho) forced a temporary cessation of design and development activities related to homogeneous breeder reactors, although basic research on aqueous uranium systems continued.

**1–1.5 The homogeneous reactor program at the Oak Ridge National Laboratory.** Early in 1949, A. M. Weinberg, Research Director of Oak Ridge National Laboratory, proposed that the over-all situation with respect to homogeneous reactors be reviewed and their feasibility be re-evaluated in the light of knowledge and experience gained since the end of 1945. Dr. Weinberg informally suggested to a few chemists, physicists, and engineers that they reconsider the prospects for homogeneous reactors and hold a series of meetings to discuss their findings.

At the meeting held by this group during the month of March 1949, it was agreed that the outlook for homogeneous reactors was considerably

brighter than in 1945 and that effort directed toward the design of a small experimental reactor should be resumed. By July 1949, interest in homogeneous reactors had increased further as a result of the preliminary studies which had been started, and it was decided to establish a small development effort on homogeneous reactors. A Homogeneous Reactor Committee, under the direction of C. E. Winters, was formed and reactor physics and design studies were undertaken on a somewhat expanded scale. By the latter part of August 1949, a preliminary design of the major components had been developed.

Construction of the reactor (Homogeneous Reactor Experiment No. 1) was started in September 1950, and completed in January 1952. After a period of nonnuclear testing with a natural-uranium fuel solution, HRE–1 reached criticality on April 15, 1952. Early in 1954 it was dismantled after successfully demonstrating the nuclear and chemical stability of a moderately high-power-density circulating-fuel reactor, fueled with a solution of enriched uranyl sulfate.

During the period of construction and operation of HRE–1, conceptual design studies were completed for a boiling reactor experiment (BRE) operating at 150 kw of heat and a 58-Mw (heat) intermediate-scale homogeneous reactor (ISHR). Further work on these reactors was deferred late in 1953, however, when it became evident from HRE–1 and the associated development program that construction of a second homogeneous reactor experiment would be a more suitable course of action.

The main reason for this decision was that HRE–1 did not demonstrate all the engineering features of a homogeneous reactor required for continuous operation of a nuclear power plant. Thus a second experimental reactor (Homogeneous Reactor Test, HRE–2), also fueled with uranyl sulfate, was constructed on the HRE–1 site to test the reliability of materials and equipment for long-term continuous operation of a homogeneous reactor, remote-maintenance procedures, and methods for the continuous removal of fission products and insoluble corrosion products. Construction of the reactor was completed late in 1956 and was followed by a period of nonnuclear operation to determine the engineering characteristics of the reactor. This testing program was interrupted for six to nine months by the need for replacing flanges and leak-detection tubing in which small cracks had developed, owing to stress corrosion induced by chloride contamination of the tubing. The reactor was brought to criticality on December 27, 1957, and reached full-power operation at 5 Mw on April 4, 1958. Shortly thereafter, a crack in the core tank developed which permitted fuel solution to leak into the $D_2O$ blanket. After consideration of the nuclear behavior of the reactor with fuel in both the core and blanket, operation was resumed under these conditions in May 1958.

TABLE 1–1

LEVELS OF EFFORT ON HOMOGENEOUS
REACTOR DEVELOPMENT AT ORNL

| Fiscal year | Millions of dollars | Man-years (technical) |
|:-----------:|:-------------------:|:---------------------:|
| 1949 | 0.15 | 5 |
| 1950 | 0.54 | 15 |
| 1951 | 2.2 | 75 |
| 1952 | 4.1 | 127 |
| 1953 | 3.4 | 119 |
| 1954 | 3.9 | 133 |
| 1955 | 7.7 | 219 |
| 1956 | 9.1 | 238 |
| 1957 | 10.0 | 316 |
| 1958 | 11.5 | 333 |

The ten-year growth of the ORNL effort on homogeneous reactors is indicated by Table 1–1, which summarizes the costs and man-years devoted to the program through fiscal year 1958.

Following the completion of construction and beginning of operation of HRE–2, the ORNL Homogeneous Reactor Project directed its attention to the design of a 60-Mw (heat) experimental aqueous thorium breeder reactor, designated as HRE–3, with the objective of completing the conceptual design during the summer of 1958. Work on slurry development and component development was accelerated to provide the information necessary for the start of construction of HRE–3 at the earliest possible date.

**1–1.6 Industrial participation in homogeneous reactor development.** Industrial participation in the homogeneous reactor program started with a number of studies to evaluate the economic potential of such reactors for large-scale power production [18–22]. The opinion of some who compared homogeneous breeder reactors with solid-fuel converters is reflected in the following excerpts from Ref. 19: "The two reactor types that offer the greatest possibilities for economic production of central station power are the thermal $U^{233}$ breeders of the circulating fuel type and fast plutonium breeders containing fuel easily adaptable to a simple processing system . . . The self-regulating features of fluid-fuel reactors and low fission-product inventory due to continuous chemical processing give these reactors the greatest possibility of safe and reliable operation . . . Both the pressurized

water and sodium-graphite systems suffer from the inability to consume (in a single cycle) a large fraction of the uranium necessary to result in low fuel costs that are attainable with breeder systems."

During late 1954 and early 1955, Westinghouse and Pennsylvania Power and Light Company, operating under Study Agreements with the Atomic Energy Commission, made a joint study [21] aimed at determining the economic feasibility of aqueous homogeneous-type reactor plants. The study indicated that a two-region solution-slurry plant and a single-region slurry plant appeared to have excellent long-range possibilities for producing competitive electric power. The study also indicated, however, that considerable development work would be required before the technical feasibility of either type of plant could be determined with any degree of certainty. The results of this and other continuing studies led the two companies to set up the Pennsylvania Advanced Reactor Project in August 1955. An initial proposal to build a 150-Mw (electric) power station financed with private funds was made to the A.E.C. by the Pennsylvania Advanced Reactor group at that time. This proposal was later modified and resubmitted as part of the power demonstration reactor program.

In spite of the formidable development program which appeared to be associated with the construction of a full-scale homogeneous reactor power plant, a second industrial group proposed building a homogeneous reactor as part of the power demonstration program in cooperation with the government. This proposal (made in response to a request by the Atomic Energy Commission for small-scale reactors) by the Foster Wheeler and Worthington Corporations in January 1956, considered construction of an aqueous homogeneous burner reactor. Plans were for a reactor and associated oil-fired superheater with a net electrical capacity of 10,000 kw for the Wolverine Electric Cooperative, Hersey, Michigan. Although this proposal was accepted in principle by the Atomic Energy Commission in April 1956, and money was appropriated by Congress for carrying out the project, in May 1958 the Atomic Energy Commission announced that plans had been canceled due to increases in the estimated cost of the plant (from $5.5 million to between $10.7 and $14.4 million).

The second proposal submitted to the Atomic Energy Commission jointly by the Pennsylvania Power and Light Company and Westinghouse Electric Corporation was determined by the Commission on February 26, 1958, as acceptable as a basis for negotiation of a contract but was later recalled, following a review by the Joint Congressional Committee on Atomic Energy. The proposal called for the construction of a reactor of the homogeneous type with a net electrical output of 70,000 to 150,000 kw to be operated on the Pennsylvania Power and Light Company system. The reactor would use a thorium-uranium fuel as a slurry in heavy water. Under the proposal, the Atomic Energy Commission would assume the

cost of research and development planned for 1958 and 1959, at which time a decision would be made either to begin actual construction of a plant or terminate the project. The cost of the project, scheduled for completion by December 1963, was estimated at \$108 million. The Westinghouse and Pennsylvania Power and Light Company's share of the cost included \$5.5 million for research since 1955, \$57 million for plant construction, and \$16 million for excess operating costs during the first five years of operation. The Atomic Energy Commission was asked to provide the additional \$29 million, including \$7 million for research and development in 1958–1959, \$18 million for research and development following a decision to construct the plant, and \$4 million for fuel charges during the first five years of operation.

## 1–2. General Characteristics of Homogeneous Reactors

**1–2.1 Types of systems and their applications.** Because of the large number of possible combinations of mechanical systems and compounds of uranium and thorium which may be dissolved or dispersed in $H_2O$ or $D_2O$, there exists in principle an entire spectrum of aqueous homogeneous reactors. These may be classified according to (a) the type of fissionable material burned and produced ($U^{235}$ burners, converters, breeders), (b) the geometry or disposition of the fuel and fertile material (one-region, two-region), or (c) the method of heat removal (boiling, circulating fuel, and fluidized suspension reactors). The possible materials which can be used in these various reactor types are given in Table 1–2; all combinations are not compatible.

### TABLE 1–2

### HOMOGENEOUS REACTOR MATERIALS

| Fuel | Fertile material | Moderator coolant | Corrosion-resistant metals of primary interest |
|---|---|---|---|
| $UO_2SO_4 + H_2SO_4$ | $U^{238}$ salt | $D_2O$ | Austenitic stainless steels |
| $UO_2F_2 + HF$ | $U^{238}$ oxide | $H_2O$ | Zircaloy–2 |
| $UO_2N^{15}O_3 + HNO_3$ | $ThO_2$ | | Titanium |
| $UO_2SO_4 + Li_2SO_4$ | | | Platinum |
| $UO_3 +$ alkali oxide $+ CO_2$ | | | Gold |
| $UO_3 + H_3PO_4, UO_2 + H_3PO_4$ | | | |
| $UO_3 + H_2CrO_4$ | | | |
| $UO_2, UO_3, U_3O_8$ | | | |

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TABLE 1-3

HOMOGENEOUS REACTOR TYPES AND APPLICATIONS

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| Reactor designation | Power level range, Mw heat | Fuel solution or suspension | Application |
|---|---|---|---|
| Water boiler | 0–0.05 | Enriched $UO_2SO_4$ or $UO_2(NO_3)_2$ in $H_2O$ | University nuclear research and training |
| Homogeneous research reactors | 800–2000 | Enriched $UO_2SO_4$ in $D_2O$ | Nuclear research at ultra-high thermal-neutron fluxes |
| $U^{235}$ burners | 40–500 | Enriched $UO_2SO_4$ in $H_2O$ or $D_2O$ | Small- to large-scale power plants in high-fuel-cost locations; mobile power plants |
| LAPRE type power reactors | 1–100 | Enriched $UO_3$ dissolved in 60 w/o phosphoric acid. Enriched $UO_2$ dissolved in 95 w/o phosphoric acid | Remotely located small- and intermediate-scale power plants |
| One-region power converters | 500–1000 | Slightly enriched $UO_3$ in $D_2O$ | Large-scale power production |
| One-region Pu producer | 1000–2000 | Slightly enriched $UO_2SO_4$ in $D_2O$ [with or without added $Li_2^7(SO_4)$] | Dual-purpose power plus plutonium production |
| Two-region Pu producer | 500–1500 | Enriched $UO_2SO_4$ in $D_2O$ (core). Depleted $UO_2SO_4$ in $D_2O$ (blanket) | Dual-purpose power plus plutonium production |
| Single-region thorium breeder | 500–1500 | Enriched $U^{235}$ or $U^{233}$ oxide plus $ThO_2$ in $D_2O$ | Large-scale power production |
| Two-region thorium breeder, solution core | 200–1000 | Enriched $U^{235}$ or $U^{233}$ as $UO_2SO_4$ in $D_2O$ (core) plus $ThO_2$ in $D_2O$ (blanket) | Large-scale power production and $U^{233}$ breeding or $U^{235}$ to $U^{233}$ conversion |
| Two-region thorium breeder, slurry core | 200–1000 | Enriched $U^{235}$ or $U^{233}$ oxide plus $ThO_2$ in $D_2O$ (core) plus $ThO_2$ in $D_2O$ (blanket) | Large-scale power production and $U^{233}$ breeding or $U^{235}$ to $U^{233}$ conversion |

The terms used in classifying homogeneous reactors may be defined as follows: *Burner reactors* are those in which fissionable fuel is consumed but virtually no new fuel is generated. To this class belong the water boilers, homogeneous research reactors, $U^{235}$ burners, and LAPRE-type reactors. *Converter reactors* produce a different fissionable fuel than is destroyed in the fission process, such as in the dual-purpose plutonium producers or single-region converters, while *breeder reactors* produce the same fissionable fuel as that which is consumed. *One-region reactors* contain a homogeneous mixture of fissionable and fertile materials in a moderator. Generally, these have large reactor diameters, in order to minimize neutron losses, and contain fuel plus fertile material in concentrations of 100 to 300 g of uranium or thorium per liter of solution or slurry. *Two-region reactors* are characterized by a core containing fissionable materials in the moderator surrounded by a blanket of fertile material in moderator. These reactors may have comparatively small diameters with dilute core-fuel concentrations (1 to 5 g of uranium per liter) and a blanket containing 500 to 2000 g of fertile material per liter. *Boiling reactors* are reactors in which boiling takes place in the core and/or blanket and heat is removed by separating the steam from the solution or suspension. *Fluidized suspension reactors* are those in which solid particles of fuel and fertile material are fluidized in the core and/or blanket, but are not circulated through the cooling system external to the reactor pressure vessel.

A summary of homogeneous reactor types and the primary application of each is given in Table 1–3.

**1–2.2 Advantages and disadvantages of aqueous fuel systems.** Aqueous fuel systems possess certain advantages which make them particularly attractive for numerous nuclear-reactor applications ranging from small reactors (for mobile units or package-power plants) to large, high-power reactors (for large-scale production of plutonium, $U^{233}$, and/or power). These advantages stem partly from the fluid nature of the fuel and partly from the homogeneous mixture of the fuel and moderator; i.e., an aqueous homogeneous reactor combines the attributes of liquid-fuel heterogeneous reactors with those of water-moderated heterogeneous reactors. If practical methods for handling a radioactive aqueous fuel system are developed, the inherent simplicity of this type of reactor should result in considerable economic gains in the production of nuclear power and fissionable material.

However, many apparently formidable practical problems are associated with continued operation and maintenance of systems involving radioactive fuel solutions. It is believed, therefore, that extensive experience in a series of small- to large-scale reactor installations will be required to demonstrate the reliability of aqueous homogeneous reactors; this will necessitate a long-range development program. In addition, the choice of

water as the fuel-bearing medium limits both the fuel concentration and operating temperature to values which may be less than optimum for production of power and fissionable material.

The principal advantages of aqueous fuel systems are:

(1) *High power density.*  Because of the homogeneous nature of the reactor fuel-fluid, virtually no heat-transfer barrier exists between the fuel and coolant.  Thus reactor power densities of 50 to 200 kw/liter may be possible, being limited by considerations other than heat transfer, such as radiation-induced corrosion and chemical reactions.

(2) *High burnup of fuel.*  In heterogeneous reactors, burnup is limited by radiation damage to fuel elements or loss of reactivity.  In liquid-fuel reactors, continual removal of poisons is possible, as well as continual additions of new fuel, thereby permitting unlimited burnup.

(3) *Continuous plutonium recovery.*  Continuous removal of neptunium or plutonium is possible in a liquid-fuel reactor.  This yields a product with a low $Pu^{240}$ content and increases the value of the plutonium [23].

(4) *Simple fuel preparation and reprocessing.*  The use of aqueous fuel solutions or slurries eliminates the expensive fuel-element fabrication step and simplifies the reprocessing of depleted fuel.

(5) *Continuous addition or removal of fuel.*  Charging and discharging fuel can be accomplished without shutting down the reactor and without the use of solid-fuel charging machines.

(6) *High neutron economy.*  Neutron economy is improved by eliminating absorption of neutrons by cladding and structural material within the reactor core.  Also, there is the possibility of continuously removing $Xe^{135}$ and other fission-product poisons.  In addition, an aqueous fuel system lends itself readily to a spherical core geometry, which minimizes neutron leakage.

(7) *Simple control system.*  Density changes in the moderator create a sensitive, negative temperature coefficient of reactivity which makes this system self-stabilizing.  This eliminates the need for mechanically driven regulating rods.  In addition, shim control can be achieved by changing the fuel concentration.

(8) *Wide range of core sizes.*  Depending on concentration and enrichment, critical $H_2O$ and $D_2O$ homogeneous reactors range from $1\frac{1}{2}$ ft to as large as is practicable.  Correspondingly, there is a wide range of application for these reactor systems.

The principal problems of aqueous fuel systems are:

(1) *Corrosion or erosion of equipment.*  The acidity of fuel solutions and abrasiveness of slurries at high flow rates creates corrosion and erosion

problems in the reactor and its associated equipment. Special provisions must therefore be made for maintaining equipment.

(2) *Radiation-induced corrosion.* The presence of fission radiation increases the rate of corrosion of exposed metal surfaces. This limits the permissible wall power density, which in turn restricts the average power density within the reactor.

(3) *External circulation of fuel solution.* Removal of the heat from the reactor core by circulating fuel solution, rather than coolant only, through external heat exchangers increases the total amount of fuel in the system and greatly complicates the problems of containment of radioactivity and accountability of fissionable material. The release of delayed neutrons in the fuel solution outside of the reactor core reduces the neutron economy of the reactor and causes induced radioactivity in the external equipment, resulting in the need for remote maintenance.

(4) *Nuclear safety.* The safety of homogeneous reactors is associated with the negative density coefficient of reactivity in such systems; however, by virtue of this coefficient, relatively large reactivity additions are possible through heat-exchanger mishaps and abrupt changes in fuel circulation rate. In boiling reactors changes in the volume of vapor within the reactor core may lead to excessive reactivity changes.

(5) *Limited uranium concentration.* In solution reactors, uranium concentration is limited by solubility or corrosion effects, and in slurries, by the effective viscosity and settling characteristics. In $H_2O$-moderated reactors, in particular, a high uranium or thorium concentration is necessary for a high conversion ratio. Concentrations up to 1000 g/liter, however, may be considered for solutions and up to 4000 g/liter for fluidized beds.

(6) *Limited operating temperatures.* At the present time the operating temperatures of aqueous solution systems appear limited because of corrosion problems at $\sim$225°C and phase stability problems above 300°C. Pressures encountered at higher temperatures are also a problem.

(7) *Explosive decomposition product.* Radiation-induced decomposition of the moderator can produce an explosive mixture of hydrogen and oxygen in the reactor system. This hazard means that special precautionary design measures must be taken. To prevent excessive gas formation and reduce the requirement for large recombiners, a recombination catalyst such as cupric ion may be added. Disadvantages associated with this addition are the neutron poisoning effects and changes in chemical equilibria which occur.

A comparison of the advantages and disadvantages of specific homogeneous reactors is given in Table 1–4.

TABLE 1–4

COMPARISON OF HOMOGENEOUS REACTOR TYPES

| Reactor types | Advantages | Disadvantages |
|---|---|---|
| One-region U$^{235}$ burner, H$_2$O or D$_2$O moderator | Possible elimination of chemical processing plant<br>Elimination of D$_2$O requirement (H$_2$O moderator)<br>Low fissile-material inventory (D$_2$O moderator) | Relatively high fuel costs (due to burning of enriched uranium with no regeneration) compared to homogeneous breeders and converters |
| Two-region breeder, solution or slurry core | High neutron economy and low fuel costs<br>Low fissile-material inventory<br>Possible fission-product removal from core solution<br>High neutron economy and low fuel costs | Radiation corrosion of zirconium core tank limits power density (may be more serious with solution core compared with slurry core)<br>Slurry handling problems |
| One-region ThO$_2$ slurry | Elimination of zirconium as a construction material | Startup and shutdown of reactor may be difficult<br>Slurry handling problems |
| One-region UO$_3$ slurry | Relatively low fissile- and fertile-material inventory | Startup and shutdown problems<br>Slurry handling problems |
| One-region UO$_2$SO$_4$ solution | Elimination of zirconium problems<br>Elimination of slurry handling problems | May require all-titanium system<br>Plutonium does not stay in solution and may deposit on walls of equipment |

## 1–3. U$^{235}$ BURNER REACTORS

**1–3.1 Dilute solution systems and their applications.**  One-region re-actors fueled with a dilute solution of highly enriched uranium or "burner reactors" are ideal as a concentrated source of neutrons, since the critical mass and size of the core of this type of reactor can be very small.  Many low-power research reactors are in operation which use this fuel system, and very-high-flux research reactors of this type are being considered [24]. The principal advantages of solution reactors for this latter application are the small amount of U$^{235}$ required for criticality and the ability to add fuel continually.

One-region burner reactors are applicable for both small- and large-scale nuclear power plants.  Such plants can operate for very long periods of time (20 years or more) without necessity for removal of all the fission products.  Corrosion product buildup, however, must be limited to prevent uranium precipitation.  The fuel concentration would be dilute, increasing with time of reactor operation if no fuel processing is carried out.  Either light or heavy water can be used as the moderator-coolant; the fuel con-centrations would always be higher for the light-water-moderated reactors. An advantage of these systems is that they utilize fuel in the concentration range which has been studied most extensively.  Experience in circulating such solutions, however, indicates that careful control of operating condi-tions and the concentrations of the various fuel constituents, such as $H_2SO_4$, $CuSO_4$, $NiSO_4$, $H_2O_2$, $O_2$, etc., is necessary to avoid problems of two-phase separation, uranium hydrolysis, and oxygen-depletion precipi-tation of uranium.

For power production, homogeneous burner reactors can be considered as possible competitors to the highly enriched solid-fuel reactors, such as the Submarine Thermal Reactor and the Army Package Power Reactor. By eliminating fuel-element fabrication, fuel costs in homogeneous burners with either $D_2O$ or $H_2O$ as the coolant-moderator are in the range of 4 mills/kwh at present Atomic Energy Commission prices for enriched uranium [25].

Possible fuel systems for the dilute, highly enriched burner-type reactors are $UO_2SO_4$ in $H_2SO_4$, $UO_2(NO_3)_2$ in $HNO_3$, $UO_2F_2$ in HF, and $UO_3$-alkali metal oxide-$CO_2$ in $H_2O$.  These fuel systems are compared in Chapter 3.

**1–3.2 High-temperature systems.**  Fuel systems of enriched uranium dissolved in highly concentrated phosphoric acid have been suggested for homogeneous power reactors because of the high thermal stability and low vapor pressure of such systems.  This permits operation at higher tempera-tures than is possible with dilute acids, with accompanying higher thermal efficiencies.  Fuel systems of this type include $UO_3$ in 30 to 60 w/o (weight

percent) phosphoric acid, $UO_2$ in 90 to 100 w/o phosphoric acid, and $UO_3$ in concentrated chromic acid. The $UO_3$–$H_3PO_4$ system, used in the Los Alamos Power Reactor Experiment No. 1 (LAPRE–1), must be pressurized with oxygen to prevent uranium reduction. Solutions containing phosphate-to-uranium ratios of 4/1 to 10/1 are stable up to 450°C. However, the neutron economy is poor and these solutions are corrosive to all metals except platinum and gold. The $UO_2$–$H_3PO_4$ systems, pressurized with hydrogen, have somewhat better corrosion characteristics and copper may be used at least in regions which are kept below 250°C.

## 1–4. Converter Reactors

**1–4.1 Purpose of converters.** In converter reactors, $U^{235}$ is burned to produce $U^{233}$ or $Pu^{239}$ by absorption of excess neutrons in fertile material. Thus the purpose of converter reactors is the production of power, fissionable material, or both. Since homogeneous reactors have to operate at temperatures above 225°C and pressures above 1000 psi because of problems of corrosion and gas production, homogeneous converters are thought of as dual-purpose reactors for the production of power and fissionable material or power-only reactors. Such reactors are also considered mainly in connection with the $U^{235}$–$U^{238}$–$Pu^{239}$ fuel cycle, whereas the homogeneous breeder reactors are associated with the thorium fuel cycle.

**1–4.2 One-region converters.** One-region converter reactors may be fueled with a relatively concentrated solution (100 to 300 g/liter $D_2O$) of slightly enriched uranium for plutonium and power production or with a suspension of slightly enriched uranium oxide for power production only.* The principal advantage of the solution-type converter for plutonium production is the insolubility of plutonium in the high-temperature uranium sulfate system (see Chapter 6). This opens the possibility of separating the plutonium by centrifugation rather than by a solvent extraction or absorption process. The costs of this method of recovering the plutonium, which contains only small amounts of $Pu^{240}$, should be considerably less than is possible with solid-fuel reactors and conventional processing techniques. Indications are, however, that the plutonium formed in the fuel solution is preferentially adsorbed on hot metal surfaces in contact with the solution and is difficult to remove (see Chapter 6). Other problems with the solution-type converter are the highly corrosive nature of concentrated uranyl sulfate solutions and the lower temperature at which the two liquid phases separate. An all-titanium high-pressure system may be

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*Early work at Columbia and Chicago was aimed at a low-temperature version of such a reactor for plutonium production only; however, present-day considerations are limited to high-temperature systems.

necessary to contain these solutions, which will lead to considerably higher equipment and piping costs. The addition of lithium sulfate to the solution would reduce corrosion and raise the phase-separation temperature so that it might be possible to use stainless steel; however, the neutron economy with normal lithium is poorer and separated $Li^7$ would be costly.

A single-region converter fueled with natural or slightly enriched uranium oxide as a suspension avoids the problems of plutonium precipitation, phase separation, and corrosion mentioned above. The advantage of such a converter reactor for power production is the elimination of radiation damage and fuel burnup problems encountered with solid-fuel elements; however, the problem of radiation damage to the reactor pressure vessel must be considered.

**1–4.3 Two-region converters.** Two-region homogeneous converters may also be fueled with either $D_2O$ solutions or slurries; in these reactors, however, the $U^{235}$ is in the core and the fertile material in the blanket. Converters of this type become breeders if the bred fuel is subsequently burned in the core and there is a net gain in the production of fuel. A two-region converter with a dilute enriched-uranium core solution and a concentrated depleted-uranium blanket solution shows promise of producing more economical power and plutonium than the one-region converter reactors mentioned previously [26] because of the lower inventory charges and the better neutron economy. Although the power density at the wall of the titanium-lined pressure vessel is lower in the case of the two-region machine, which minimizes the possibility of accelerated corrosion rates, there is some evidence [27] that titanium corrosion will not be severe in any case. The major materials problem in the dilute-solution core converter will be that of zirconium corrosion, which may be above 30 mils/year at power densities necessary for economic production of power and fissionable material.

Two-region converters fueled with a uranium oxide slurry in the core may be a possibility as an alternative to the solution-slurry system; however, not much is known about the corrosion resistance of zirconium in contact with fissioning uranium oxide or about the engineering behavior of such a slurry.

## 1–5. BREEDER REACTORS

**1–5.1 The importance of breeding.** If present projections [28] for the growth of the nuclear power industry in the United States are correct, the installed capacity of nuclear electric plants in 1980 may be as much as 227 million kilowatts and may be increasing by 37 million kilowatts annually. Even assuming optimistic figures for fuel burned in then-existing plants and fuel plus fertile material for inventories in new plants [29],

the annual requirement of fissionable material will be approximately 420,000 kg in 1980. This fissionable material will have to come from natural sources (i.e., uranium mined from the ground) or be produced from neutrons absorbed in fertile material in a reactor (i.e., breeding or conversion). Since presently known reserves of high-grade ores of uranium and thorium in the United States [30] contain 148,000 tons of uranium and 60,000 tons of thorium, respectively, and these in turn contain only $10^6$ kg of fissionable material, it is obvious that conversion of a significant fraction of the fertile material contained in the reserves will be necessary.

Although such a conversion will not reduce the inventory requirement of fuel and fertile material for new plants starting up, this amounts to only about 25% of the burnup requirement. On this basis, the goal of nuclear industry should be to develop reactor designs and associated fuel systems which achieve a consumption of at least 5% to 10% of the total fertile material, as well as the initial fissionable material. At this point the annual burnup requirement would become small compared with the inventory requirement. This corresponds to a total burnup of about 50,000 Mwd/ton. Although such a fuel consumption might be obtained in high-neutron-economy converter reactors through recycling of the fuel, it seems likely that even the best such reactor may fall short of this goal and that both fast and thermal breeders will be needed.

In the long term, therefore, the development of breeding systems is a must. In the short term, where emphasis is on fuel costs rather than on neutron economy and fertile-material utilization, converters rather than breeders may predominate.

**1–5.2 One-region thorium breeders.** Since $U^{233}$ does not occur in nature, homogeneous thorium breeder reactors will probably start out as converters, with $U^{235}$ as the fuel and thorium as the fertile material. One-region reactors of this type utilize a suspension of 100 to 300 g per liter of thorium oxide plus enriched $U^{235}$ as oxide and $D_2O$ as the moderator. In order to maintain a breeding ratio greater than 1, fuel processing is necessary to remove fission-product poisons. Also, to reduce losses due to neutron leakage, the diameter of the reactor should be at least 12 ft.

**1–5.3 Two-region breeder reactors.** Two-region breeder reactors would have thorium oxide suspensions (500 to 1500 g/liter) in the blanket region and could have either a highly enriched uranyl sulfate solution (1 to 10 g/liter) or a thorium oxide-uranium oxide slurry (200 g $ThO_2$/liter and 10 g $UO_3$/liter) in the core region. Use of a solution-type core permits the continuous removal of insoluble fission-product poisons by means of hydroclones, while a slurry-type core leads to higher breeding ratios. Because of these compensating factors, estimated fuel costs are

approximately the same in both types of reactors (see Chapter 10). While the use of a suspension in the core may minimize the problem of radiation-induced corrosion of the zirconium, not much is yet known about the behavior of zirconium in a thorium-uranium slurry-fueled reactor. Calculations summarized in Chapter 10 show that both solution- and slurry-fueled two-region breeders have higher breeding ratios and lower fuel costs than one-region breeders.

Numerous studies of large-scale two-region breeder reactors have been carried out [20,26,31–42], some of which are described in detail in Chapter 9.

## 1-6. MISCELLANEOUS HOMOGENEOUS TYPES

**1-6.1 Boiling reactors.** In May 1951, following completion of the construction of HRE-1, a group at the Oak Ridge National Laboratory focused its attention on the possibility of removing heat from a homogeneous reactor by boiling, rather than by circulating the fuel solution, in recognition of the advantages of a boiling reactor. These are: (a) more rapid response to sudden reactivity increases, minimizing power excursions, (b) elimination of fuel circulating pumps, (c) increase in the temperature of steam delivered to the turbine for a given reactor operating pressure, and (d) reduction or elimination of problems of corrosion and induced radioactivity associated with the circulation of fuel and fertile material through an external heat-removal system. However, at that time, questions of the nuclear stability of a boiling, liquid-fuel reactor and the maximum specific power, in terms of kilowatts per liter, that could be extracted from a given size core remained to be answered.

Experiments on bulk boiling at atmospheric pressures in a 1-ft-diameter cylindrical tank indicated that power densities up to 5 kw/liter might be achieved. It soon became apparent, however, that high-pressure power-density measurements would be required, and the design of a boiling reactor experiment (BRE) called the "Teapot" was initiated. To answer the question of the nuclear stability of such a reactor, a combined group from the Oak Ridge National Laboratory and Los Alamos operated the SUPO under boiling conditions in October 1951. The reactor was operated at a total power of 6 kw and solution power densities of 0.5 kw/liter were obtained.

This removed one of the important obstacles to the construction of an experimental boiling reactor, and in January 1952, the Oak Ridge National Laboratory made a proposal to the Atomic Energy Commission to construct the Boiling Reactor Experiment (BRE) to answer the question of maximum specific power at higher pressures and to investigate the operating characteristics of boiling reactors. The proposed reactor was to operate at a power level of 250 kw of heat and pressures up to 150 psi. The re-

actor was estimated to cost approximately \$300,000, including the building to house it. A one-year effort involving nuclear and engineering calculations, completion of the BRE conceptual design, and experiments on bubble nucleation in the presence of radiation resulted from the proposal. In January 1953, however, the problem of maintaining sufficient oxidizing conditions to prevent reduction and precipitation of the uranium in a boiling uranyl sulfate solution became apparent, and construction of the reactor was deferred pending outcome of solution-stability experiments. These experiments, completed in October 1953, indicated that at oxygen concentrations likely to be encountered in a boiling reactor (6 to 7 ppm), reduction of the uranium would occur in solutions in contact with stainless steel.* Since the metallurgy of titanium or zirconium was not sufficiently advanced to construct a reactor using these alternate metals, it was decided to abandon the BRE itself and continue experimental work on the problems of solution stability, steam separation, and power densities at high pressure. Work on steam separators and experimental measurements of the movement of air and steam through heated solutions at high pressures were carried out under contract by the Babcock & Wilcox Company [44]. These results and theoretical calculations of the power removal from boiling reactors [45,46] provide a basis for estimating the obtainable power density of such reactors under varying core heights, operating pressures, and moderator density decreases. Values range from 10 to 40 kw/liter with an average of 18.5 kw/liter for a 15-ft-high core, operating at 2000 psi, and a density decrease due to steam of 0.4. Although the effect of such a void fraction on nuclear stability is not known, if tolerable, boiling reactors may be able to achieve average power densities comparable to those estimated for large-scale nonboiling circulating-fuel reactors operating under a similar pressure [47]. In this latter case, the holdup of solution in the external circulating system lowers the power density of the core only, to an average of about 10 to 20 kw/liter. The two systems are comparable, therefore, in terms of obtainable power densities, and boiling reactors cannot be excluded on this basis.

The various applications of boiling as a method of heat removal from homogeneous reactors include a one-region boiling solution or slurry reactor, a two-region reactor with a nonboiling core and a boiling blanket, and a two-region reactor with a boiling core and a boiling blanket. The problem of maintaining a sufficiently oxidizing solution in a boiling uranyl sulfate–$D_2O$ reactor, although serious in a stainless-steel system, can be eliminated if all surfaces in contact with the solution are made of titanium and oxygen is supplied continuously. Solutions containing 10 g of uranium

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*In more recent tests [43] with nonboiling solutions, in which oxygen concentrations were held at 2 to 3 ppm, no reduction and precipitation of uranium occurred.

per liter have been successfully boiled at 325°C in titanium-lined pipe [48]. Experiments have not yet been carried out with higher uranium concentrations in titanium. Continued interest in boiling homogeneous reactors has led to a number of studies of large-scale reactors of this type [33,36,37,49–51]. The actual construction of a boiling slurry reactor is under way in the USSR [52].

Use of boiling as a method for removal of power from the ThO₂ slurry blanket of large two-region homogeneous reactors appears to present no major difficulties [53]. The apparent advantages are that no circulating pump would be required to handle slurry, containment of the highly active slurry in the reactor vessel, and the possibility of operating the blanket at the core pressure and using the blanket power for heating the secondary steam [54]. One major problem is that of keeping the slurry suspended during startup.

**1–6.2 Gaseous homogeneous reactors.** Although this book deals primarily with aqueous systems, some mention should be made of other types of fluid-fuel homogeneous reactors in which the fuel and moderator are mixed and can be circulated. The existence of $UF_6$, which has a low parasitic capture cross section and is a gas at ordinary temperatures, makes possible the consideration of gaseous reactors. $UF_6$ boils at 56.4°C at atmospheric pressure and has a critical temperature of 252°C at 720 psi. Although $UF_6$ is corrosive to most metals, it can be contained in nickel and monel. However, the effect of radiation on the integrity of the protective film has not been studied. Considerable experience has been gained in the handling of $UF_6$ in metal containers at high temperatures and pressures.

Pure $UF_6$ is not a practical possibility for a gaseous homogeneous reactor because fluorine is not a good enough moderator. Addition of helium makes such a reactor possible, and calculations by D. E. Hull in a report, "Possible Application of $UF_6$ in Piles" [55], show that the critical mass of a graphite-reflected, He + $UF_6$, reactor is 84 kg of $U^{235}$. About 15 tons of helium in a 60-ft-diameter core would be required. In a recent investigation [56] of reflector-moderated gaseous reactors (Plasma Fission Reactor), the critical mass of gaseous $U^{235}$ in a 2-ft-diameter cavity surrounded by $D_2O$ was calculated to be less than 1 kg. Such reactors would have to operate at extremely high temperatures (3000°K) which many feel are beyond the realm of present technology.

Mixed $UF_6$ gas and dispersions of solid moderators such as beryllium or graphite have been suggested, as well as beds of moderator particles fluidized with circulating $UF_6$ [55]. However, these proposals have no apparent advantages compared with gas- or liquid-cooled fluidized systems described in the following section.

**1–6.3 Fluidized systems.** A variant of the fixed-bed or solid-moderator homogeneous reactors consists of subdividing the fuel and/or moderator to the point where the particles can be fluidized by the flowing gas or liquid. Gas-cooled reactors of this type have received considerable attention because of the higher heat-transfer rates obtainable compared with fixed-bed reactors. A number of studies of gas-fluidized reactors have been carried out by ORSORT groups at the Oak Ridge National Laboratory [57,58] and by other groups [59,60].

In an Oak Ridge study [61] various types of fluidized-bed reactors were compared. Systems investigated were (a) a sodium-cooled fast reactor, (b) a gas-cooled system, (c) an organic-moderated reactor, (d) a heavy-water-moderated reactor, and (e) a light-water system. A detailed study of this latter system was carried out to compare its characteristics and performance with solid-fuel heterogeneous pressurized-water reactors. The results indicated that both the light-water-moderated and organic-moderated fluidized reactors showed promise, while the gas-cooled, the $D_2O$-cooled, and the fast (unmoderated) reactors were found to be less satisfactory for application of the fluidized-bed technique.

Systems using $ThO_2$, fluidized by gas or $D_2O$, were described by the Dutch at the Geneva Conference on Atomic Energy in 1955 [62,63].

Calculations show that a typical, one-region, 400 thermal Mw reactor having a core diameter of 15 ft and a temperature rise of 50°C would require particles in the 40- to 60-micron range [64], whereas a two-region reactor with a liquid-fluidized blanket would require particles in the 200- to 600-micron range [65], and if the particles were confined to a 6-in. annulus next to the core the particle size required would be in the 0.10- to 1.5-cm range [65].

The disadvantages that may be observed with fluidized suspension systems include the possibility of particle attrition [65], and instabilities due to channeling during steady-state operation and due to settling if a circulating pump failed.

Room-temperature attrition tests using 0.1-in.-diameter × 0.1-in.-long $ThO_2$ and $ThO_2 + 0.5\%$ CaO cylinders (cylinders prepared by calcination at temperatures of both 1650 and 1800°C) fluidized in water gave an attrition rate of 12 to 15% weight loss per week [66]. However, circulation tests using 10- to 20-micron $ThO_2$ spheres (calcined at 1600°C) in toroids at superficial velocities up to 26 ft/sec and water temperatures of 250°C showed essentially no attrition for periods up to 200 hr [67].

This appears to indicate that attrition rate is at least a function of particle size, without giving any indication as to the effect of void fraction, slip velocity, particle shape and density.

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