

*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  
June 1958

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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  $\text{UO}_2\text{SO}_4$  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  $85^\circ\text{C}$ .

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