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UC-80 → Reactor Technology

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ARMY REACTORS PROGRAM
ANNUAL PROGRESS REPORT
FOR PERIOD ENDING OCTOBER 31, 1962

OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

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ORNL-3386

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ARMY REACTORS PROGRAM ANNUAL PROGRESS REPORT
for Period Ending October 31, 1962

L. D. Schaffer
Program Coordinator

Date Issued

APR 10 1963

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
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U.S. ATOMIC ENERGY COMMISSION

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SUMMARY

The Laboratory's participation in the Army Nuclear Power Program continued to include review, inspection, and support in various areas of reactor technology. In addition, an advanced fuel irradiation test program was established that is to be conducted in the pressurized-water loop in the Oak Ridge Research Reactor.

Review of the design of the MH-1A reactor was initiated. This reactor, a pressurized-water system fueled with low-enrichment bulk UO_2 clad with stainless steel, is being designed by the Martin-Marietta Corporation for the U.S. Army Corps of Engineers as a floating plant to furnish electrical energy to shore installations.

Studies of the out-of-pile corrosion resistance of stainless steel brazed joints were completed. T-joint specimens of type 304 stainless steel were brazed together with 18 different alloys. Initial testing resulted in the selection of five of these alloys for extended testing, which was carried out in autoclaves with oxygen or oxygen and hydrogen added to the autoclave water. These alloys, General Electric alloys Nos. 81 and 75, Coast Metals alloy NP, low-melting Microbraz, and a palladium-base alloy, all proved satisfactory. Coast Metals alloy NP was selected as the reference braze material for the SM-1 fuel elements because it was more amenable to the brazing method established for this element.

A method was developed for stabilizing Eu_2O_3 against aqueous corrosion in the event a control rod containing this material became defective in service. The method developed involves the addition of MoO_3 to the Eu_2O_3 . When a mixture of Eu_2O_3 -15 wt % MoO_3 is reacted, a single phase, believed to be $\text{Eu}_2\text{MoO}_{12}$, is formed. This was tested for aqueous corrosion resistance and found to be essentially inert.

The irradiation testing that was carried out on nonsintered bulk UO_2 fuel in the ORR pressurized-water loop was completed, and the loop is to be used for irradiation testing of advanced cermet fuel elements, as well as water chemistry experiments in support of the Army pressurized-water reactor program. The design of the cermet fuel assemblies is under way. It is expected that experimental assemblies will be placed in the

loop during the spring of 1963. In the meantime, the water chemistry studies now under way will be continued.

The ORR loop has operated satisfactorily since December 1959 in the ORR. This loop was designed to recirculate water at conditions up to 2500 psig, 650°F, and 80 gpm. A recent test to determine the loop heat exchanger capacity at a nominal operating temperature of 480°F indicates that it is capable of removing 300 kw. The operating experience of the loop has been very good. Water leakage is extremely low, and thus excellent control of the water chemistry is obtained.

While much information on water chemistry is already available, relatively little in the way of a fundamental understanding of observed behavior has been gained, especially in connection with the important unsolved problems of the transport and deposition of corrosion products and their associated radioactivity. At the beginning of the water chemistry study in the ORR pressurized-water loop in early 1961, cooled water samples taken from the loop upstream of the ion exchangers showed relatively low activity levels during normal undisturbed loop operation compared with similar samples taken during loop startup. About 90% of the activity was removed by the purification system ion exchangers, and about two-thirds could be removed by a 0.45- μ millipore filter with a cellulose backing pad. The crud level was estimated to have been <0.05 ppm during normal operation. In contrast, during loop startup, the water-borne activity level was about 50 times higher and the crud level was often several ppm; 98 to 99% of the activity was removed by the ion exchangers, and 95% could be removed by a 0.45- μ filter. After the middle of 1961, radiochemical analysis of the water samples taken upstream of the ion exchangers showed a decrease in the level of water-borne activity. These later samples generally showed a lower percentage of activity removed by a 0.45- μ filter. It appears that the normal crud level in the ORR loop since the latter part of 1961 has been 0.01 ppm or less.

The loosely deposited crud on some of the surfaces of the fuel assemblies in the loop were removed and analyzed. No marked trends were found in the various specific activities with location of the fuel rod or even from one experiment to another.

The crud samples collected on a filter from the loop water during a period of high crud level were studied and found to have a magnetite structure. As the oxide film on the metal surface grows, magnetite presumably is released as crud to the water. Its presence in the crud as well-developed crystallites, however, seems especially significant in view of the nearly complete absence of crud particles in the 0.01- to 0.5- μ range. The implication is that recrystallization of the magnetite occurs, and this implies that appreciable radiochemical exchange can take place between activated and nonactivated crud. In fact, the present results provide evidence that significant radiochemical exchange takes place between the two fractions of water-borne activity — the filterable crud fraction and the nonfilterable, apparently ionic, fraction. It would appear that the dissolved material is the principle agent of activity transport in well-conditioned, neutral-pH, pressurized-water systems during periods of normal operation. The transport process would consist of the liberation of activity from the in-flux-crud deposits as ionic material, which can exchange with crud deposits throughout the system. It would at the same time account for the well-crystallized appearance of the crud. A simple model was formulated that should permit analysis of this transport process.

Since magnetite is a principle constituent of crud, it suggests itself as a possible filter ion-exchange medium. Experiments were performed in which high-temperature loop water was passed through a cartridge filled with magnetite. The results showed quite favorable decontamination from activity that was mostly nonfilterable.

Postirradiation examinations were performed on 34 fuel specimens contained in six BMI-32 NaK-filled capsules that had been irradiated in the ETR. The fuel specimens were in the form of plates with a cermet fuel body of either UO_2 or UN dispersed in type 347 stainless steel along with various additions of burnable poison in the form of B_4C , NbB_2 , and ZrB_2 . These specimens were clad with type 347 stainless steel and a few specimens contained boron in the frame of the specimen or a Eu_2O_3 cermet adjacent to the fuel cermet. The fuel was highly enriched to nominal concentrations of 26 and 40 wt % UO_2 and 34 wt % UN. The UO_2 used was

either of the hydrothermal or the spherical type. Eleven of the specimens failed in degrees varying from complete disintegration to surface blistering. It is felt that these failures were attributable primarily to overheating in the capsule rather than to irradiation-damage effects. From the examination of the 23 specimens that survived the test in good condition, it was found that the 26 wt % UO_2 cermet could sustain a U^{235} depletion of 67% with a surface temperature in the range of 600°F and maintain adequate dimensional stability and structural integrity. The burnable poison or Eu_2O_3 cermet cores did not appear to affect the stability of the specimens.

The examination of a third SM-1 fuel assembly, S-81, is under way. This element remained in the SM-1 for 19.5 Mwy of reactor operation. The two assemblies examined previously, S-72 and S-79, were removed after 10.5 and 16.4 Mwy of reactor operation. The preliminary findings with respect to element S-81 are that (1) the outer fuel plates exhibit greater rippling than noted before, (2) the thickness measurements of individual fuel plates vary in proportion to the axial burnup, with the maximum thickness increase being 10% or 0.003 in. over the nominal dimensions where the U^{235} depletion was found to be 62%, and (3) no matrix cracking is present, but the frequency of cladding cracks appears to be significantly greater than noted previously.

Twelve miniature stainless steel-clad specimens containing 20, 30, and 40 wt % Eu_2O_3 cores were inserted in the ETR for irradiation testing. Eight specimens have acquired their requested exposure and have been subjected to postirradiation examinations. These examinations revealed that the specimens underwent no dimensional or structural changes. Corrosion tests were performed on these specimens by defecting the cladding to expose the Eu_2O_3 and exposing these specimens to high-temperature water. The specimens swelled, and the extent of swelling was a function of the Eu_2O_3 loading. The irradiation exposure had little or no effect on the swelling characteristics. The autoclave water was analyzed after these tests, and the europium content was found to be below spectrographic detection limits.

During an inspection of the SM-2B fuel element at the SM-1, an area suspected of having a crack was noted on one of the welded side plates.

This element was therefore examined, and it was found that the suspect area was not cracked and that the indication noted was composed of a series of fine scratches.

Two pieces of equipment were developed to facilitate the postirradiation examination of fuel components. Plate-spacing measurement equipment was constructed and used, and a fission-gas-sampling apparatus was completed and used for sampling the gas in the BMI-32 capsules.

Two pressure-vessel steel surveillance capsules were removed from the SM-1. One was removed after 16.4 Mwyr of reactor operation and the other was removed after 20.8 Mwyr of reactor operation. These capsules each contained three subsize Izod impact specimens made of ASTM A212, grade B, steel with six notches in each. These impact specimens were tested, and it was concluded that notch No. 2 of the impact specimens best represented the dose of the pressure vessel. The Charpy-V transition temperature increase for notch No. 2 after 20.8 Mwyr reactor exposure was determined to be 92°F.

1. INTRODUCTION

In 1952, the U. S. Army was assigned responsibility for the development of the land-based nuclear power plants required by the military services for heat and power at remote locations. The Army delegated this responsibility to the Corps of Engineers, and the Army reactor program was established as a joint program of the Army and the Atomic Energy Commission; the program is now termed the Army Nuclear Power Program. In 1953, the Oak Ridge National Laboratory was requested to participate in the Army program. The first task assumed by the Laboratory was to design, in a short time, a practical nuclear reactor plant that could furnish electrical power in certain isolated localities at a cost that would be competitive with the cost of power produced by existing power generation facilities in similar locations. Based on these requirements, a pressurized-water package reactor was selected. The conceptual design of this 10-Mw reactor plant, which was called the Army Package Power Reactor (APPR), was completed in the summer of 1954. In late 1954, Alco Products, Inc., was selected as the contractor to build the APPR, based on the ORNL design, at Ft. Belvoir, Virginia, as the first Army reactor. The APPR, since renamed the SM-1 to correspond with present Army nomenclature, was completed and taken to criticality on April 8, 1957. During the detailed design and construction of the APPR, ORNL supported the project by reviewing the design, by performing the critical experiment, by developing and testing the fuel and control materials, and by fabricating the core.

Since the initial effort on the APPR, ORNL has continued to provide similar types of development and consultation support to the Army Nuclear Power Program. This report summarizes the work performed at ORNL during the past year. A major addition to the program during the year was the establishment of an experimental program utilizing the pressurized-water loop in the Oak Ridge Research Reactor. Advanced-fuel development testing and basic pressurized-water system water-chemistry studies will be conducted with this loop (see sec. 6).

2. GENERAL REVIEW AND SUPPORT ACTIVITIES

During the current report period assistance was provided on a variety of problems that required short-term efforts of several members of the Laboratory staff. The more significant activities of this nature are discussed in this section.

SM-1 Support Activities

On October 16, 1961, the SM-1 was shut down for maintenance. During the shutdown, ORNL and Alco Products personnel performed, on October 26, 1961, an inspection of several core components in the SM-1. The results of this inspection are summarized in Table 2.1. Because the line observed on fuel element SM-2B ran through one of the side-plate welds, the Army requested that ORNL examine this assembly more closely in the hot-cell facilities at Oak Ridge to determine whether this mark was a crack. The element was therefore transferred to ORNL and the examination was performed. The results of the examination are reported in Section 7 of this report.

One problem associated with on-site inspection of the fuel and control components at the SM-1 plant is that the equipment available for underwater viewing does not permit magnified viewing of the component surfaces. The Army requested that ORNL study and make recommendations on optical equipment that would not only permit convenient underwater observation of radioactive components but would also include magnification and photographic facilities. In the recommendations submitted it was suggested that the Army consider an optical system that would include equipment comparable to the Omniscope and the Questar telescope. Such a setup was used at ORNL in the inspection of the HRT core tank; however, this inspection, although made remotely, was performed in air rather than under water.¹ If the distance between the objective of the Omniscope and the surface being viewed is small, a magnification of 6 can be obtained.²

¹Oak Ridge National Laboratory, "HRPP Prog. Rep. for Period from August 1 to November 30, 1960," USAEC Report ORNL-3061, Feb. 27, 1961, pp. 1-11.

²Letter to L. D. Schaffer, Oak Ridge National Laboratory, from John L. Maulbetsch, Lerma Engineering Corporation, Jan. 16, 1961.

Table 2.1. Summary of Results of Inspection of SM-1 Core Components

Component	Description	Findings
20V	Core II fuel element	Element appeared to be in good condition; a few scratches were evident
SM-2A	Experimental SM-2 type fuel element	Element appeared to be in good condition; some film discoloration was observed
SM-2B	Experimental SM-2 type fuel element	A questionable area was observed on the dowel-pin side plate; a line was evident in the central region of the side plate; it progressed through one weld and extended to the adjacent welds
S-80	Core I high-burnup fuel element	Element appeared to be in good condition
S-81	Core I high-burnup fuel element	Element appeared to be in good condition, but its surfaces were more discolored than those of element S-80
BG-1	Experimental boron-gradient control rod	Rod appeared to be in good condition; some surface scratches were evident, and some of the weld surfaces appeared to be irregular

Much higher magnifications can be obtained if the Questar telescope is included in the equipment arrangement. Such viewing equipment can be procured with builtin or externally supplied lighting and with various cameras.

The lighting design of the optical system for the SM-1 will, of course, require careful study. Possibly a mockup would be worthwhile, since the lighting is a critical aspect in the use of such equipment, particularly for underwater viewing. An unsheathed Omniscope designed for underwater use is available,² but the scope can also be used within a thin, metal, protective sheath. The use of a sheath would reduce the problem of decontaminating the equipment after use.

Two high-burnup fuel elements, S-80 and S-81, which were part of core I, were retained in the SM-1 after core I had reached its end of

life on April 28, 1960 after 16.4 Mwyr of reactor operation, as reported previously.³ These two fuel elements were subsequently removed from the SM-1 in October 1961 (19.5 Mwyr of reactor operation) at the recommendation of ORNL. This recommendation was made as a result of the postirradiation examination performed by ORNL on core I fuel assembly S-79, which was removed from the SM-1 at the end of core I life. The examination of element S-79 revealed that, in addition to intergranular cladding cracks of the type previously observed in the examination of fuel element S-72, which was exposed for a shorter time (10.5 Mwyr), transgranular cracking was also beginning to occur in the cladding.

It had been hoped that at least one of the two fuel assemblies S-80 and S-81 could be operated in the SM-1 to a very high fuel depletion, but, when the new cracking mechanism was noted, it was felt that further testing should be postponed until more was learned about this mechanism. To study this problem, ORNL requested element S-81 for examination; the companion fuel element S-80 was retained in storage at the SM-1 for future disposition. The examination of element S-81 is under way, and preliminary findings are reported in Section 7 of this report. In addition to the examination of element S-81, which remained in the SM-1 2.1 Mwyr longer than element S-79, waterlogging tests are being planned at ORNL on specimens from fuel elements S-72, S-79, and S-81 in an attempt to ascertain the susceptibility to failure of these cermet fuel elements as a function of burnup. It is hoped that from this work it will be possible to determine the value and the hazards involved in further irradiation testing of element S-80.

Associated with the problem of determining whether or not to continue testing element S-80 is the problem of selecting a place for further testing if such is considered worthwhile as a materials test but uncertain as far as operational hazards to the SM-1. At the request of the Army, ORNL made a survey of the pressurized-water loops in the United States and Canada that would be suitable for testing this SM-1 fuel element. It was determined that there is no loop in the U. S. or Canada

³Oak Ridge National Laboratory, "Army Reactors Program Prog. Rep.," USAEC Report ORNL-3231, pp. 1 and 2, Jan. 31, 1962.

large enough in diameter to contain this fuel element, whose cross-sectional diagonal length is $4 \frac{1}{32}$ in. If a loop test is selected, it appears therefore that it could best be accomplished by construction of a "loopsule" of the type proposed by the General Electric Company for use in the GETR.

Code Development Program Review

Consulting service continued to be provided for the Army's Code Development Program until July 1962, when the program was terminated. In this program, the Martin Company had the responsibility for developing an integrated system of reactor calculation programs that would provide in a single turn at the computer the analysis data required from the initial geometry determination to evaluation of a final spent core. A code was completed in one-dimensional form, but a period of "debugging" will probably be required. Plans to develop a multidimensional code were abandoned, at least temporarily, following a program review by the AEC Advisory Committee on Reactor Physics in December 1961.

Review of Military Compact Reactor (MCR) Design

The Army requested that ORNL review the Military Compact Reactor (MCR) design and the proposed program associated with the design, as prepared by the Nuclear Development Corporation of America (now merged with the United Nuclear Corporation) and the General Motors Corporation. The MCR system provides a light-weight, extremely compact nuclear power plant with an electrical output of 3 Mw; it includes a high-temperature liquid-metal-cooled reactor. The details of the design and review are classified. The review was completed and recommendations of the ORNL review committee were transmitted to the Army on February 15, 1962.

General Support Activities

A review was made and comments were transmitted to the Army on December 14, 1961 with regard to Alco Products' specification for

the procurement of the third core for the SM-1, SM-1A, and PM-2A reactors.⁴

In anticipation of future core procurement, ORNL personnel prepared recommendations for the Army on the preparation of so-called "pedigreed" fuel elements and control rods. Since the time between fabrication and postirradiation examination of reactor core components is usually long, the preirradiation measurements and design variations of such components are often difficult to obtain and to interpret for comparison with the postirradiation measurements. In addition, the preirradiation information, if available, is not always as complete as is desirable. Therefore it was suggested that representative core components be selected during fabrication, that they be carefully measured and marked, and that this preoperational information be documented for future reference to facilitate the postirradiation examination and to permit close comparison of pre- and postoperational measurement data. In addition, it was suggested that these pedigreed components be located in the reactor so that they would be exposed to the desired operational conditions, such as highest burnup, steepest neutron flux gradient, etc.

There are many problems associated with obtaining precise and meaningful pre- and postirradiation measurements of core components. It is usually a difficult task to measure the plate spacing variations by hand between all the fuel plates of a plate-type assembly. An automated method developed⁵ and currently being used at ORNL in the postirradiation examination of Army fuel elements involves the use of Dermatron⁶ and a strip-chart recorder. This equipment gives significantly better results than the hand-operated elliptical probe equipment used in the past. The new technique is being considered by the Army as a possible method for

⁴E. C. Edgar and H. R. Clayton, "Specification and Fabrication Procedures for Type 3 Fuel Elements," USAEC Report APAE Memo-311, rev. 1, Alco Products, Inc., April 22, 1962.

⁵C. V. Dodd and R. W. McClung, "Fuel Element Coolant Channel and Other Spacing Measurements by Eddy-Current Technique," USAEC Report ORNL-TM-129, Oak Ridge National Laboratory, March 20, 1962.

⁶The Dermatron, a unit produced by Unit Process Assemblies, Inc., is a device that measures the change of impedance in an electrically energized coil moved in close proximity along a surface.

preirradiation measurements of plate spacing of the flat-plate assemblies manufactured for Army reactors. The measurement equipment used at ORNL is discussed further in Section 8 of this report.

On the basis of an inspection made by ORNL personnel at Ft. Greeley, Alaska, of the fuel elements comprising core I of the SM-1A, seven fuel elements were rejected. These seven fuel elements were shipped to ORNL for further examination. A summary of the findings and recommendations is presented in Table 2.2. The elements are now in storage at ORNL, and their disposition is under study by the Army.

Consultation was provided on the Army advanced fuel-element development program for pressurized-water reactors. Both flat-plate and tubular cermet fuel elements are being considered. In addition, ORNL personnel participated in the review required for selecting one of the two basic pressurized-water reactor designs employed in the existing Army pressurized-water reactor plants for the first standard plant concept. This review began as a comparison of the merits of the core designs of the two types of plant, one of which utilizes flat-plate cermet fuel elements and the other tubular cermet fuel elements; however, it became evident that general aspects of the primary system designs were of major importance in such a selection. Therefore the review was expanded to include a comparison of the primary system designs of the two plants. The result of the over-all review was that the Army selected as their first standard plant the primary system that employs tubular fuel elements. The fuel-testing program for this plant is just getting under way and is briefly discussed in Section 6 of this report.

Consulting services were provided on the Nuclear Engineering Test Reactor which is a 10-Mw pool-type military test reactor being constructed at Dayton, Ohio. The consulting work included making recommendations on equipment and procedures for measuring the energy distribution of the neutrons and gamma rays in the test positions of the facility, making suggestions on operational procedures, and presenting ideas on how to establish a procedure for the hazards review of experiments to be performed in the test facility.

Table 2.2. Findings and Recommendations Concerning Rejected SM-1A Fuel Elements

Element Identification	Findings	Recommendations
2S	A superficial transverse scratch less than 0.005 in. deep is evident approximately halfway down the length of the bottom plate; braze run out and braze reaction on top plate appear to be excessive; in several areas the brazed joint on the top plate, as viewed from the top, is not visible	Reject because of poor-quality brazed joints on top plate; this assembly could be used in the SM-1 if necessary
8S	Braze fillets on bottom plate are negligible, but there is braze metal in the joints	Can be used in the reactor; although the fillet in the brazed joints on the bottom plate is quite small, rejection is not warranted
29S	Surface quality is satisfactory; top plate at location 26 extends 0.002 in. above the side plate because of rippling	The top fuel plate distortion is such that the reactor designer should determine whether or not this fuel assembly is suitable for reactor use; it might be possible to repair this element by locally deforming the area of deviation
34S	There are several small gouges 0.015 in. deep on the top plate 4.5 in. from the top end fitting	Can be used in the reactor, but it might be better to hold it as a spare
36S	There are several small gouges 0.0175 in. deep on the top plate 0.5 in. down the plate and near the edge of the side plate	Can be used in the reactor, but it might be better to hold it as a spare
40S	There are superficial scratches on the plates, slight braze-metal splatter on the top plate, and a pit less than 0.005 in. deep that appears to contain some braze metal	Reject because of braze-metal splatter over the fuel section; this element could be used in the SM-1 if necessary
47S	There is a slight scratch less than 0.005 in. deep on the bottom plate approximately 7 in. from the top end fitting	Can be used in the reactor

The Army requested information about the Personal Radiation Monitor (PRM) developed at ORNL.⁷ The PRM is the size of a fountain pen, and may be worn in the pocket of the user. The device provides an immediate audible and visual indication of the gamma dose rate. The audible warning produced is a chirping sound, and simultaneously a visual indication is given by the flash of a neon lamp at the top of the meter. The rate of either of these signals is proportional to the gamma dose rate. The saturation point of the meter, that is, the point where the chirp becomes a continuous sound, can be a preselected value in the range 1 to 100 r/hr. Since the device is not an integrating meter, it is used in conjunction with a dose-integrating device. This information, along with information on cost and availability of the PRM, was transmitted to the Army. In addition, a PRM was loaned to the Army for demonstration and evaluation at the SM-1.

Radioactive carriers were provided during this period for transfer of radioactive material between the SM-1 and ORNL. Arrangements are also being made to provide an SM-1 fuel element fabrication display for the proposed APPR (SM-1) reactor plant display at the Smithsonian Institution.

⁷R. H. Dilworth and C. J. Borkowski, "Personal Radiation Monitor," USAEC Report ORNL-3058, Oak Ridge National Laboratory, Aug. 28, 1961.

3. MH-1A REACTOR DESIGN REVIEW

The MH-1A reactor is being designed by the Martin Marietta Corporation for the U. S. Army Corps of Engineers. It is to be a floating nuclear power plant to furnish electrical energy to shore installations. The letter designation of the plant means that the reactor is mobile (M), that its power level is high (H), and that it is the first (1) field plant (A) of this type. The reactor will be contained within a converted "Liberty" ship, the SS Walter F. Perry. This ship will not have propulsion equipment and will serve only as a floating mount for the reactor plant; however, it will be suitable for ocean towing.

The reactor will be a pressurized-water system operating under the design conditions summarized below:

Reactor thermal power, Mw	45
Coolant flow rate, gpm	8,000
Operating system pressure, psia	1,600
Average coolant temperature, °F	450
U ²³⁵ loading, kg	151.7
Fuel enrichment, % U ²³⁵	
Outer zone	5.4
Inner zone	5.0
Linear heat rate, kw/ft	
Average	4.6
Hot spot based on hot-spot factors	15.8
Fuel burnup, Mwd/MT of UO ₂	
Average	9,924
Maximum	22,500

The fuel will consist of high-density UO₂ pellets clad with type 304 stainless steel tubing. The core will utilize 32 fuel assemblies and 12 boronated stainless steel control rods. Additional reactor control will be achieved by placing 600 ppm of boron in the fuel cladding.

The reactor and all equipment that contains high-pressure primary system water will be enclosed within a vapor-containment vessel. Grounding and collision barriers are being planned to protect the containment vessel in the event of a ship accident. The containment vessel is also

being designed to withstand the external hydrostatic force that would result from submergence to a depth of 150 ft. Pressure-relieving devices are being considered to relieve external pressures on the containment compartment if submergence is greater than 150 ft.

The design is being reviewed by ORNL for the U. S. Army Corps of Engineers. Review studies of the reactor core analysis, materials selection, system analysis, thermal analysis, and plant safety are planned.

It is expected that construction of the MH-1A will be completed early in 1965. Present plans are to perform the postconstruction testing on this floating power plant at Ft. Belvoir, Virginia.

4. FUEL ELEMENT DEVELOPMENT STUDIES

R. J. Beaver C. F. Leitten

The results of studies that led to the selection of Coast Metals alloy NP as the brazing metal for the SM-1 fuel elements were recently published.¹ In these studies, T-joints of type 304L stainless steel brazed with selected alloys were tested in quiescent, degassed and deionized water in autoclaves at 565°F under a pressure of 1200 psi. A typical specimen is shown in Fig. 4.1. In the initial phase of the investigation, tests were limited in duration to 1000 hr in order to quickly screen 18 potential alloys and thus select those worthy of longer testing. Based on weight-change data and metallographic examinations, 5 of the 18 alloys exhibited sufficient corrosion resistance to warrant further investigation. These alloys, identified as General Electric alloys Nos. 81 and 75, Coast Metals alloy NP, low-melting Microbraz, and a palladium-base alloy containing 37 wt % Ni and 3 wt % Si, were then subjected to autoclave tests of 12 and 16 months duration. In these extended tests, 1 cm³ of O₂ or a mixture of 1 cm³ of O₂ and 50 cm³ of H₂ was added per liter of water to simulate SM-1 reactor water conditions and to evaluate the effect of the gases on the corrosion behavior of the alloys.

On the basis of weight-change data and metallographic examination after long-term exposure of T-joint specimens, General Electric alloys Nos. 81 and 75, Coast Metals alloy NP, low-melting Microbraz, and the palladium-base alloy were considered to have acceptable corrosion resistance. As may be seen by the comparative illustrations in Figs. 4.2 through 4.6, no significant differences in the corrosion behavior of these alloys were noted. Coast Metals alloy NP was finally selected as the reference brazing alloy for the SM-1 fuel elements, because it was the alloy most amenable to the brazing method established for this fuel element.

¹R. J. Beaver et al., "An Investigation of the Corrosion Resistance of Brazing Alloys for Austenitic Stainless Steel Fuel Elements for Service in 565°F Pressurized Water," USAEC Report ORNL-2834, Oak Ridge National Laboratory, March 29, 1962.

Fig. 4.1. Braided Stainless Steel Corrosion Test Specimen.

Fig. 4.2. Type 304L Stainless Steel Joint Brazed with Coast Metals Alloy NP and Autoclave Tested at 565°F and 1200 psi for 12 Months in Distilled Water Containing 50 cm³ of H₂ and 1 cm³ of O₂ per liter. Etchant: 10% oxalic acid.

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Fig. 4.3. Type 304L Stainless Steel Joint Brazed with General Electric Alloy No. 81 and Autoclave Tested at 565°F and 1200 psi for 16 Months in Distilled Water Containing 1 cm³ of O₂ per liter. Etchant: 10% oxalic acid.

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Fig. 4.4. Type 304L Stainless Steel Joint Brazed with General Electric Alloy No. 75 and Autoclave Tested at 565°F and 1200 psi for 16 Months in Distilled Water Containing 1 cm³ of O₂ per liter. Etchant: 10% oxalic acid.

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Fig. 4.5. Type 304L Stainless Steel Joint Brazed with Low-Melting Microbraz and Autoclave Tested at 565°F and 1200 psi for 16 Months in Distilled Water Containing 1 cm³ of O₂ per liter. Etchant: 10% oxalic acid.

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Fig. 4.6. Type 304L Stainless Steel Joint Brazed with 60% Pd-37% Ni-3% Si Alloy and Autoclave Tested at 565°F and 1200 psi for 16 Months in Distilled Water Containing 1 cm³ of O₂ per liter. Etchant: 10% oxalic acid.

5. DEVELOPMENT OF EUROPIUM OXIDE AS CONTROL MATERIAL

R. A. Potter

Investigations of the aqueous corrosion resistance of Eu_2O_3 as used in Eu_2O_3 -bearing stainless steel absorber plates were continued,¹ and a method was developed for stabilizing the oxide against corrosion in the event a plate becomes defective in service. During the course of investigations of the effects of additives on the aqueous corrosion resistance of Eu_2O_3 , it was found that additions of MoO_3 made the material inert.

Mixtures of Eu_2O_3 and 15 wt % MoO_3 heated in air reacted to form a single phase, which was identified by x-ray diffraction² as a face-centered-cubic structure with a cell constant of 5.43 Å. The material was found to be optically isotropic, with a refractive index greater than 1.88. The normal body-centered-cubic form of Eu_2O_3 has a cell constant of 10.86 Å and a refractive index of 1.87. The chemical formula of the compound, based on wet chemical analysis, was tentatively established as $\text{Eu}_5\text{MoO}_{12}$ or, typically, an MO_2 -type compound (metal dioxide compound). It is obvious, however, that this molecular formula does not have a balanced valence number. Since the chemical analysis data show the europium to be actually $\text{Eu}_{5.3}\text{MoO}_{12}$, a valence number balance can be obtained if the molecular form of this compound is assigned as $\text{Eu}_{5.3}\text{MoO}_{11}$. The pycnometric density was 7.21 g/cm³, and this value, when used with the x-ray diffraction data, leads to the conclusion that the unit cell of the single phase contains two-thirds of a molecule of $\text{Eu}_{5.3}\text{MoO}_{11}$.

Thermogravimetric analysis of the Eu_2O_3 -15 wt % MoO_3 mixture was performed in air at a pressure of 660 mm Hg. A total loss in weight of 1.05% was noted as the mixture, not previously reacted, was heated from 20 to 1300°C. Subsequent x-ray diffraction analysis of the sample showed that the mixture had reacted to form the cubic compound. Assuming that the

¹R. A. McNees and R. A. Potter, Europium Oxide Studies, pp. 21 and 22, "Army Reactors Program Prog. Rep.," USAEC Report ORNL-3231, Oak Ridge National Laboratory, Jan. 31, 1962.

²X-ray crystallography performed by L. A. Harris, Metals and Ceramics Division, Oak Ridge National Laboratory.

weight loss was due to the evolution of MoO_3 , the calculated composition was found to be in reasonable agreement with the chemical formula $\text{Eu}_{5.3}\text{MoO}_{11}$.

Differential thermal analysis of an unreacted mixture of Eu_2O_3 -15 wt % MoO_3 was performed in ambient air. No reaction peaks were observed as the mixture was heated from 20 to 1450°C at a rate of 10°C/min; however, an x-ray diffraction analysis of the sample showed that the mixture had reacted to form $\text{Eu}_{5.3}\text{MoO}_{11}$.

Pellets made from the compound were tested for aqueous corrosion resistance in 100°C water for periods up to 1250 hr and found to be essentially inert. In 250°C water a weight gain of only 4% was observed after 850 hr. The pellets remained physically intact, and there were no visible signs of attack.

The chief advantage of $\text{Eu}_{5.3}\text{MoO}_{11}$, besides being a stabilized, hydration resistant form of Eu_2O_3 , is that the amount of stabilizer required is small in comparison with most of the other stabilizing compounds investigated. In comparing the molecules of Eu_2O_3 and $\text{Eu}_{5.3}\text{MoO}_{11}$, it is seen that the europium content in the stabilized molecule is 11% less and the density of the MoO_3 stabilized Eu_2O_3 is comparable to Eu_2O_3 fired in air between 1600 and 1700°C.³ Therefore the penalty paid in the reduction of the volumetric concentration of europium in $\text{Eu}_{5.3}\text{MoO}_{11}$ over Eu_2O_3 is relatively small, particularly in comparison with the dilution caused by other stabilizing compounds, such as TiO_2 . Because the cores of compact reactors usually require control rods of high reactivity worth and therefore a high concentration of neutron-absorbing material, and because powder metallurgy considerations limit the amount of europium compound that can effectively be dispersed in stainless steel, the molybdenum-stabilized europium oxide appears to be quite attractive as a long-lived control rod material for compact, pressurized-water reactors.

It was found that WO_3 could be substituted for MoO_3 with the resultant formation of a face-centered-cubic compound similar to $\text{Eu}_{5.3}\text{MoO}_{11}$.

³"Neutron Absorber Materials for Reactor Control," pp. 547-548, U.S. Atomic Energy Commission, U.S. Government Printing Office, Washington, D.C., 1962.

It was also determined that the rare earth oxides Sm_2O_3 , Gd_2O_3 , and Dy_2O_3 react with MoO_3 to form face-centered-cubic compounds.

6. PRESSURIZED-WATER IN-PILE LOOP

Specimen Irradiations

V. O. Haynes

The ORR pressurized-water loop was completed in late 1959 and has been utilized in the ORNL Maritime Reactor Program. The major purpose of the specimen irradiations conducted for this program was to investigate the characteristics of nonpelletized low-enrichment UO_2 for potential use in future N.S. SAVANNAH cores. The irradiation testing of these specimens is now completed, and the loop will be utilized in the ORNL Army Reactors Program.

Although the various Maritime program specimens differed in some features, in general they were UO_2 clad with 0.500-in.-OD type 304 stainless steel tubing with a nominal wall thickness of 0.035 in. They varied in length from 18 to 19 in. The tubes were fueled for 16.25 in. with fused and ground UO_2 having U^{235} enrichments as low as 0.80% and as high as 5%. The fueled rods were furnace brazed into clusters of three on a 0.612-in. 60° triangular pitch. Typical specimen assemblies are shown in Fig. 6.1. Further details of these experiments may be found in the ORNL Maritime Reactor Program annual reports.¹⁻³

In the Army program, the ORR pressurized-water loop will be used for irradiation testing of advanced cermet fuel elements and to carry out water chemistry experiments in support of the advanced pressurized-water reactor program. The first fuel irradiation assembly planned for testing in the loop will include various cermet fuel specimens prepared by the Martin Marietta Corporation. Although the details have not been finalized,

¹W. C. Thurber et al., Fuel Development, pp. 79-92, "Maritime Reactor Program Ann. Prog. Rep. Nov. 30, 1961," USAEC Report ORNL-3238, Oak Ridge National Laboratory.

²V. O. Haynes and W. C. Thurber, Fuel Irradiation Tests, pp. 100-105, "Maritime Reactor Program Ann. Prog. Rep. Nov. 30, 1961," USAEC Report ORNL-3238, Oak Ridge National Laboratory.

³E. L. Long, Jr., Postirradiation Metallographic Examination of Experimental Assembly No. 1, pp. 106-112, "Maritime Reactor Program Ann. Prog. Rep. Nov. 30, 1961," USAEC Report ORNL-3238, Oak Ridge National Laboratory.

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Fig. 6.1. Specimen Assemblies for Experiments 4 and 5.

the specimens will be stainless steel-clad highly-enriched UO_2 -stainless steel cermet in the form of tubes and flat plates, and they will be tested under conditions that will provide performance data beyond the range of current experience. The present plans call for irradiation of nine tubes and up to 24 plates as coupons. The tubes will be 0.506 or 0.540 in. OD and will have a 9-in. fueled length; they will be arranged on a 60° triangular pitch. The cladding will be 0.0075-in.-wall type 347 stainless steel tubing, and the cermet thickness will be 0.030 or 0.090 in. Several loadings ranging from 28 to 45 wt % UO_2 will be tested. The coupon specimens are envisioned as 1.5×1 in. with several thicknesses and UO_2 loadings. The cladding will be the same as for the tubes, that is, 0.0075-in.-thick type 347 stainless steel.

The specimens will be arranged to locate six tubes in the A-2 leg of the loop, and three tubes and the coupons will be placed in the A-1 leg of the loop. The specimen holder is being designed to permit interim examinations and partial specimen replacement if desirable.

The heat flux and total heat generation in the presently planned tests of highly enriched fuel will be much higher than in previous experiments performed in the loop. This situation has required that a special study be performed in order to evaluate the maximum thermal performance of an experimental assembly that can be permitted without loop modifications. This work is under way and is discussed in the following subsection.

It is expected that the fuel specimens being prepared by the Martin Marietta Corporation will be ready for insertion into the loop during the spring of 1963. In the meantime, the water chemistry studies now under way will continue.

Loop Operation

J. A. Conlin J. K. Franzreb
I. T. Dudley D. E. Tidwell
D. B. Trauger

The pressurized-water loop at the ORR has operated satisfactorily since December 1959, with only routine maintenance being performed. The

arrangement of equipment in this facility has contributed greatly to its successful operation. The use of three pumps in a parallel system, with only one operating at a given time, makes possible continuous operation of the loop over most of the combined operating lifetime of the pumps. Since two are in standby at all times, the full lifetime of each pump can be utilized without hazard.

Two test spaces are accessible in the loop, in addition to the in-pile section. These include an out-of-pile simulation of the in-pile section in the equipment room and an extended region above the irradiation test space in which samples can be located by attachment to the specimen support rod.

Numerous sampling tubes are provided that are brought to a common station, including one system for removal of water at the loop operating temperature. The sampling station provides for attachment of the filters and bombs needed for water chemistry studies.

Removal of fuel (and any associated metal test pieces) can be conducted underwater in the ORR pool, and the fuel can be transferred directly to the ORR hot cell. Thus the irradiated specimens can be examined soon after reactor shutdown. In addition, since it is possible to return specimens to the loop for further exposure, interim examinations can be performed and followed by continued testing.

The loop is constructed entirely of austenitic stainless steel. Type 347 stainless steel was used for most parts of the loop, except the in-pile section, which was constructed of type 316 stainless steel because of its higher strength properties. Some valves, instrumentation, and other minor components were available only in type 304 stainless steel.

The test loop was designed to recirculate water at pressures up to 2500 psig, temperatures up to 650°F, and flow rates up to 80 gpm. It has a heat-removal capacity in excess of 150 kw. The piping within the reactor is arranged as a U-tube, with straight vertical access through the reactor vessel cover. Each leg provides a space 1.5 in. in diameter and approximately 20 in. long for irradiation of specimens. The maximum perturbed thermal-neutron flux is 7×10^{13} neutrons/cm².sec, and the peak gamma heat is approximately 5 w/g. The legs of the U-tube are enclosed

in a stainless steel vacuum jacket to provide thermal insulation and auxiliary containment. The specimen access hole is closed with an O-ring seal above the reactor vessel for ease in insertion and removal of test assemblies.

A simplified flow diagram of the loop is presented in Fig. 6.2, and Fig. 6.3 is a cutaway view of the pressurized-water loop installed in the ORR. The equipment arrangement in the basement of the ORR is shown in Figs. 6.4 and 6.5, and a view of the operating area is presented in Fig. 6.6. The loop makeup system is shown in Fig. 6.7, and Fig. 6.8 is a view of the ORR pool showing the upper end of the in-pile section and the in-pool piping vacuum jacket. The three hermetically sealed Westinghouse pumps are shown in Fig. 6.9. Additional details are presented in the design report, which was originally published in February 1961 as an internal report and was recently issued for external distribution.⁴

⁴I. T. Dudley, D. E. Tidwell, and D. E. Trauger, "Design Report for NMSR Pressurized-Water Loop at ORR," USAEC Report ORNL-3188, Oak Ridge National Laboratory, Dec. 19, 1962.

Fig. 6.2. Simplified Flow Diagram of the ORR Pressurized-Water Loop.

Fig. 6.3. Cutaway Drawing of the ORR Pressurized-Water Loop.

In order to assure that the loop would have a heat capacity in excess of 150 kw, the heat exchanger was designed to remove 150 kw at a primary coolant temperature of 300°F. A recent test to determine the heat exchanger capacity at a nominal operating temperature of 480°F indicated that under this operating condition it was capable of removing 300 kw of heat.

There are features of the loop that make it uniquely suitable for certain pressurized-water system tests. Water leakage is extremely low; in fact, it is below detectable levels, and no routine water makeup is required. Thus, the loop affords a facility for excellent control over

Fig. 6.4. Elevation Drawing of the ORR Pressurized-Water Loop Equipment.

water chemistry. The long operating periods that can be achieved without maintenance shutdowns or drainage of the system also contribute to chemical stability of the loop.

The loop has been operated primarily with a water flow rate of 40 gpm at an in-pile inlet temperature of 500°F or at 50 gpm and 480°F. Other operating parameters of importance have been the following:

Pressure, psi	1750
pH	6.5 to 8.5
Hydrogen content, ppm	1.8 to 3.6
Total maximum solids, ppm	1
Oxygen content, ppm	<0.05
Chloride content, ppm	<0.10

Fig. 6.5. Plan Drawing of the ORR Pressurized-Water Loop Equipment.

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Fig. 6.6. Operating Area for the ORR Pressurized-Water Loop.

Fig. 6.7. Water Makeup System for the ORR Pressurized-Water Loop.

Fig. 6.8. View of ORR Pool Showing Upper End of In-Pile Section of the ORR Pressurized-Water Loop and In-Pool Piping Vacuum Jacket.

Fig. 6.9. Primary System Pumps of the ORR Pressurized-Water Loop System.

Water Chemistry Studies

C. F. Baes, Jr. T. H. Handley

Early in 1961 a study of the water chemistry of the ORR in-pile pressurized-water loop was initiated as part of the Maritime Reactor Program at ORNL.⁵ Since July 1962, this study has been supported by the ORNL Army Reactors Program. In order to describe the present status, the results of work performed for the previous, as well as the present, program are presented here.⁶

⁵C. F. Baes, Jr., and T. H. Handley, Water Chemistry Studies, pp. 43-50, "Maritime Reactor Program Ann. Prog. Rep. Nov. 30, 1961," USAEC Report ORNL-3238, Oak Ridge National Laboratory.

⁶This work was discussed at the Third Conference on Nuclear Reactor Chemistry, Gatlinburg, Tenn., Oct. 9-11, 1962.

As mentioned above, the loop is especially suitable for water chemistry studies. It is well instrumented; its operation is relatively trouble free; it is exceptionally leaktight; and it is relatively free of fission-product contamination. One purpose of the studies has been to provide chemical and radiochemical data on water conditions pertinent to loop operation, but the principle objective has been to increase the basic understanding of water chemistry as an approach to more economical operation of pressurized-water reactors.

While much information is already available, there is relatively little that provides a basis for a fundamental understanding of the observed behavior, especially in connection with the important unsolved problems of the transport and deposition of corrosion products (crud) and the associated radioactivity. The deposition of crud can foul heat transfer surfaces, clog flow channels, and impair the operating reliability of a mechanical system. The transport and deposition of radioactivity can cause activity buildup throughout a pressurized-water system and thus increase the cost of routine maintenance. The activity buildup may eventually necessitate decontamination, which would create the associated problems of radioactive waste disposal. There is accordingly a strong incentive to learn more of the mechanisms of these processes of crud and activity transport and deposition in the hope that new methods for their control will be suggested.

The operating parameters of the ORR pressurized-water loop pertinent to the water chemistry studies are summarized in Table 6.1. A general description of the fuel rod assemblies that have been tested in this loop is given above in the subsection on Specimen Irradiation, and the irradiation history of the assemblies is summarized in Table 6.2.

Observed Activity Behavior

Water-Borne Activity and Crud Levels. At the beginning of this study, cooled water samples taken upstream of the ion exchangers showed relatively low activity levels during normal undisturbed loop operation compared with similar samples taken during a loop startup (Table 6.3). During normal operation about 90% of the water-borne activity was removed

Table 6.1. ORR Pressurized-Water In-Pile Loop Parameters
Pertinent to Water Chemistry Studies

Material	
Out of flux	Type 347 stainless steel
In flux	Type 316 stainless steel
Fuel rods	Type 304 stainless steel
Peak neutron flux ^a	
A-2 leg	$\sim 6 \times 10^{13}$ neutrons/cm ² ·sec
A-1 leg	$\sim 3 \times 10^{13}$ neutrons/cm ² ·sec
Nominal water temperature	490°F
Water flow rate	50 gpm
Volume	
Out of flux ^b	1.1×10^5 cm ³
In flux ^c	1.37×10^3 cm ³
Two fuel rod assemblies	3.7×10^2 cm ³
Area	
Out of flux ^b	1.38×10^5 cm ²
In flux ^c	1.44×10^3 cm ²
Two fuel rod assemblies	1.17×10^3 cm ²
Water conditions	
Purification rate ^d	3.7×10^{-4}
pH	7
Hydrogen content	1.8 to 3.6 ppm
Conductivity	~ 0.1 μ mho/cm

^a2200 meter/sec flux measured with a stainless steel monitor wire in the facility during specimen irradiation.

^bExcludes pressurizer.

^cWith test section empty.

^dFraction of loop volume passed to ion-exchangers per second, based on 3/4 gpm (45 cm³/sec).

by the purification system ion exchangers. Other tests showed that about two-thirds of the activity could be removed by a 0.45- μ millipore filter with a cellulose backing pad, as indicated in Table 6.4.

The small volume of the loop system limits the size of the sample that can be taken and has prevented direct chemical analysis of the high-purity water, which has, typically, a specific resistance of 10^7 ohms.

Table 6.2. Irradiation History of Experimental Fuel Rod Assemblies

Experimental Assembly No.	Irradiation Dates		Location ^a in Loop	Reactor Cycles	Exposure (Mwd of reactor operation)
	Start	Finish			
1	3-26-60	5-14-60	A-1, A-2	2	2100
	10-4-60 ^b	11-20-60		1	
3	5-21-60	7-9-60	A-1, A-2	2	6000
	12-1-60 ^b	7-4-61		4	
4	7-12-61	10-22-61	A-2	2	2600
5	7-12-61	7-29-62	A-1	7	9400
6	10-31-61	2-11-62	A-2	2	2800
7	2-19-62	6-3-62	A-2	2	2600

^aLegs of U-shaped test section occupy lattice positions A-1 and A-2 in the ORR core.

^bInterrupted irradiation.

Table 6.3. Results of Radiochemical Analyses of Water from the ORR Pressurized-Water Loop

Nuclide	Activity (dps/liter)			
	Sample Taken with Loop Operating ^a		Sample Taken During Loop Startup ^b	
	Sampled Ahead of Ion Exchanger	Sampled After Ion Exchanger	Sampled Ahead of Ion Exchanger	Sampled After Ion Exchanger
A ⁴¹	8 × 10 ⁵			
Mn ⁵⁶	9 × 10 ⁴	2 × 10 ³		
Fe ⁵⁹	8 × 10 ³	8 × 10 ²	3 × 10 ⁵	3 × 10 ³
Co ⁵⁸	6 × 10 ³	5 × 10 ²	2 × 10 ⁵	3 × 10 ³
Co ⁶⁰	5 × 10 ³	4 × 10 ²	2 × 10 ⁵	2 × 10 ³
Mo ⁹⁹	3 × 10 ³	1 × 10 ²		
Mn ⁵⁴	2 × 10 ³	1 × 10 ²	3 × 10 ⁴	6 × 10 ²
Cr ⁵¹	2 × 10 ³	2 × 10 ²	1.3 × 10 ⁵	3 × 10 ³

^aSamples taken on March 6, 1961.

^bSamples taken on March 17, 1961.

Table 6.4. Activity Removed by Filtering Samples of Loop Water

Filter Pore Size (μ)	Activity Removed by Filtration (%)			
	With Loop Operating ^a		During Loop Startup ^b	
	Filter Ahead of Ion Exchanger	Filter After Ion Exchanger	Filter Ahead of Ion Exchanger	Filter After Ion Exchanger
0.45 ^c	65	58	95	93
0.05 ^d	<0.01		0.1	
0.01 ^d	<0.05			

^aSamples taken on March 6, 1961.

^bSamples taken on March 17, 1961.

^cGross activity (less A⁴¹) removed by filter.

^dActivity subsequently removed by filter.

Moreover, the concentration of solids (crud) that can be removed by filtration is too low during normal operation to be determined directly by weighing; however, from the activity retained by a filter and from the specific activity of the crud (Table 6.5) measured during a period of disturbed loop operation when weighable samples of crud could be collected on a filter, the crud level was estimated to have been <0.05 ppm during normal operation. During loop startup, the water-borne activity level is about 50 times higher than during normal operation, and the crud content is often several ppm; 98 to 99% of the activity can be removed by the ion-exchangers; and 95% of the activity can be removed by a 0.45- μ filter.

Until the middle of 1961, further evidence for the presence of appreciable transportable crud in the ORR loop was provided by occasional increases in the pressure drop across the in-pile test section of the loop. Such pressure drop increases were presumably the result of clogging in the flow channels in the test section. Temperature and flow cycling were performed to overcome this effect. In addition, the fuel rod test assemblies removed from the loop at the conclusion of the early exposure tests (assemblies 1 and 3, Table 6.2) were found to be well coated with loosely adhering deposits of black crud.

Table 6.5. Radiochemical Analyses of Various Crud Samples

Date ^a	Sample ^b	Specific Activity (dps/g)							Activity Ratio	
		Fe ⁵⁹ (45.1 d)	Fe ⁵⁵ (2.94 y)	Co ⁵⁸ (72 d)	Co ⁶⁰ (5.3 y)	Ta ¹⁸² (112 d)	Ce ⁵¹ (27.8 d)	Mn ⁵⁴ (300 d)	Fe ⁵⁵ /Fe ⁵⁹	Co ⁶⁰ /Co ⁵⁸
Fuel Rod Crud Deposits										
		× 10 ⁷	× 10 ⁷	× 10 ⁷	× 10 ⁷	× 10 ⁷	× 10 ⁷	× 10 ⁷		
1-21-61	1-(A-2)	22		13	6.7		20	1.2		
	1-(A-1)	19		11	4.6		13	1.0		0.48 ± 0.05
7-4-61	3-(A-2)-1	12	12	10	4.1	0.29	25	1.3		
	3-(A-2)-2	19	17	14	4.9	1.6	23	2.7		
	3-(A-2)-3	21	18	19	6.4	0.31	26	2.7	1.0 ± 0.1	0.39 ± 0.05
	3-(A-1)-3	11	14	10	4.1	0.84	13	1.6		
	3-(A-1)-2	11	12	11	3.9	1.4	10	1.6		
	3-(A-1)-1	9	9	7	3.5	0.88	6.5	0.9		
10-22-61	4-(A-2)-1	16	13	25	5.4	0.50	180	3.2		
	4-(A-2)-2	18	20	23	6.3	0.29	200	4.1	0.8 ± 0.2	0.27 ± 0.06
	4-(A-2)-3	17	10	18	6.9	0.43	260	3.1		
3-11-62	6-(A-2)-1	10	13	12	5.3	0.08	290	3.0		
	6-(A-2)-2	11	14	18	7.2	0.11	180	2.8	1.4 ± 0.1	0.40 ± 0.05
	6-(A-2)-3	10	15	19	6.9	0.11	290	3.0		
Water-Borne Crud										
5-17-61		7.2	17	5.4	3.2					
		8.8	22						2.5 ± 0.2	0.67 ± 0.08
		5.9	16	4.5	3.4					

^aDate fuel rod irradiation was terminated or date water sample was taken.

^bPrefix numbers refer to irradiation assembly (see Table 6.2), A-1 and A-2 refer to the legs of U-shaped test section, and the final numerals are: 1 for top, 2 for middle, and 3 for bottom of fuel rod.

After the middle of 1961, much less difficulty was encountered with the pressure drop across the test section, and loop operation was relatively undisturbed. Similarly, radiochemical analyses of water samples taken upstream of the ion exchangers (Table 6.6, Fig. 6.10) showed a decrease in the level of water-borne activity. The sums of the Fe^{59} , Co^{58} , Co^{60} , and Mn^{54} activities in the samples taken November 9, 1961, January 8, 1962, and March 19, 1962 (all taken during periods of normal, undisturbed loop operation) were 4900, 2300, and 4000 dps per liter of sample, respectively, compared with $\sim 2 \times 10^4$ dps per liter for similar earlier samples (Fig. 6.10). Also, these later samples generally showed a lower percentage of activity removed by a 0.45- μ filter. Judging from these results, the normal crud level in the ORR loop since the latter part of 1961 has been 0.01 ppm or less.

Results from filtration of loop water samples through filters of decreasing porosity (Table 6.4) indicate that there is a discontinuity in crud particle size. Virtually all the activity that can be removed by filtration is caught by a 0.45- μ filter; very little additional material is stopped by a 0.01- μ filter.

These observations are in general agreement with the results of a water chemistry study of the SM-1 pressurized-water reactor, which, like the ORR loop, is operated at neutral pH and with dissolved hydrogen present.⁷ The total coolant activity and the crud level of the SM-1 are similarly affected by reactor operation. At startup, shutdown, and during low-power operation, crud levels are usually >0.5 ppm. At steady-state full-power operation, crud levels are usually <0.1 ppm, and most of the activity is nonfilterable; that is, it is not held by a 0.45- μ filter.

Fuel Rod Crud Deposits. At the termination of irradiation of assemblies 1, 3, 4, and 6, samples of loosely adhering crud were removed from the fuel rod test assemblies for radiochemical analyses. The various samples are listed in Table 6.5. The samples are listed according to the direction of water flow through the test section, the inlet being at (A-2)-1 and the outlet at (A-1)-1.

⁷C. A. Bergmann, "SM-1 Research and Development Program: Long Lived Induced Activity Buildup in SM-1 Core I Lifetime; Task XVIII - Phase I," USAEC Report APAE No. 77, Alco Products, Inc., Nov. 30, 1960.

Table 6.6. Results of Radiochemical Analyses of Loop Water Samples Taken Upstream of the Ion Exchanger

	Sample Taken July 27, 1961		Sample Taken November 9, 1961		Sample Taken January 8, 1962		Sample Taken March 19, 1962	
	In Filtrate	On Filter	In Filtrate	On Filter	In Filtrate	On Filter	In Filtrate	On Filter
Nuclide activity, dps per liter of sample								
Fe ⁵⁹	210		530	1600	530	120	450	115
Fe ⁵⁵	880		1900	5300	210	35	1380	203
Co ⁵⁸	130		780	580	400	45	530	70
Co ⁶⁰	220		450	530	430	83	650	45
Mn ⁵⁴	780		400	530	880	160	1080	150
Fraction of total activity			0.34	0.66	0.85	0.15	0.88	0.12
Nuclide activity ratio								
Fe ⁵⁵ /Fe ⁵⁹	4.2		3.6	3.3	0.4	0.3	3.1	1.8
Co ⁶⁰ /Co ⁵⁸	1.7		0.58	0.91	1.1	1.8	1.2	0.6

Fig. 6.10. Sum of Fe^{59} , Co^{58} , Co^{60} , and Mn^{54} Activity in Loop Water.

There are no marked trends in the various specific activities with location on the fuel rods, or even from one sample to another, in spite of the differences in water exposure times and the duration of radiation exposure (Table 6.2). The variation in the Ta^{182} activities are attributed to analytical difficulties. The high Cr^{51} activities of the samples from assemblies 4 and 6 probably reflect the fact that the crud deposits were thin on these assemblies, and in scraping them off a relatively large amount of Cr^{51} was probably removed from the chromium-rich underlying oxide.

The specific activities in the samples of water-borne crud (Table 6.5), while lower than those in the fuel rod crud deposits, are not greatly different in level or in distribution of activity. Spectrochemical analyses of crud from fuel rods and from coolant samples (Table 6.7) also gave similar results.

Characteristics of Particulate Material. Electron micrographs of crud samples collected on a filter during a period of high crud level are shown in Fig. 6.11. Many of the particles have pronounced geometrical form, suggesting that they are crystallites. Electron diffraction patterns from such fields indicated the presence of the magnetite structure, and the appearance of the diffraction patterns confirmed that magnetite was present as large crystals.⁸

Magnetic mixed oxides of iron and chromium of the magnetite structure have often been reported to be a principle constituent of crud.⁹ Magnetite (Fe_3O_4) is the stable oxide of iron under the operating conditions of such pressurized-water systems as the ORR loop. It is formed at the

⁸T. E. Wilmarth, Analytical Chemistry Division, Oak Ridge National Laboratory, private communication.

⁹D. J. DePaul, "Corrosion and Wear Handbook," USAEC Report TID-7006, Chap. 2, pp. 11-20, March 1957.

Table 6.7. Spectrochemical Analysis of Various Crud Samples

Date ^a	Sample	Crud Weight (g)	Composition (wt %)			
			Fe	Ni	Mo	Cr
			Fuel Rod Crud Deposits			
7-4-61	3-(A-2)-1	0.1941	51.6	11.8		0.23
	3-(A-2)-2	0.0833	74.5	11.2		
	3-(A-2)-3	0.0165	74.2	10.1		
	3-(A-1)-3	0.0717	64.2	11.5		
	3-(A-1)-2	0.1356	51.7	12.9		
	3-(A-1)-1	0.1890	58.2	17.2	0.21	
			Water-Borne Crud			
5-17-61		0.0548	51	8.2		
		0.0074	74	11		
		0.0035	57	8.6		

^aDate fuel rod irradiation was terminated or date water sample was taken.

interface between water and the protective, chromium-rich, oxide film present on the surface of corroding stainless steel. Atoms of iron that have diffused through the oxide film are oxidized by water. As the oxide film grows, magnetite presumably is released as crud to the water. Its presence in crud as well-developed crystallite seems especially significant, however, in view of the apparent nearly complete absence of crud particles in the 0.01- to 0.05- μ range. The implication is that recrystallization of the magnetite occurs, and this implies, in turn, that appreciable radiochemical exchange can take place between activated and nonactivated crud.

Behavior of Dissolved Material. Several observations have been made which suggest strongly that the activity not removed from loop water samples by filtration through a 0.45- μ filter is associated with dissolved ionic material. First, it was found, as indicated in Table 6.8, that appreciable portions of the manganese, iron, and cobalt activity were retained by cellulose backing pads, which initially were used to support

Fig. 6.11. Electron Micrographs of Solids Removed from the ORR Pressurized-Water Loop (Left, May 1961; Right, August 1962).

Table 6.8. Results of Loop Water Filtration Tests Conducted on April 12, 1961

Sample ^a Pretreatment	Distribution of Activity ^b (%)					
	Co ⁵⁸			Mn ⁵⁶		
	Filter	Backing	Filtrate	Filter	Backing	Filtrate
Untreated	21	77	2	3	77	21
0.01 M in HCl	31	<1	64	5	<1	95
0.01 M in NaOH	43	11	43	3	13	83
0.01 M in EDTA and 0.01 M in NaOH	32	<1	69	4	<1	94

^aSample taken ahead of ion exchangers during normal loop operation.

^bValues are percentages of the introduced activity found at the position indicated.

the millipore filters. Mild pretreatment of the sample with dilute acid, dilute base, or half-neutralized EDTA (a strong ion-complexing agent) reduced this retention. This behavior suggests that the activity was retained by an ion-exchange process.

In paper electromigration tests, activity which had passed through a 0.45- μ filter and had been absorbed on a backing pad was found to move toward the cathode, indicating the activity to be associated with cationic material. On comparing the velocity of migration with that observed for dissolved metal salts, it further appeared that the activity was present as the simple metal cation and not as colloidal particles.

The conductivity of loop water in the cooled side-stream purification system, upstream of the ion exchangers, was typically 0.2 μ mho/cm (at 37°C) during normal loop operation. This corresponds to a dissolved solids level of 0.027 ppm as Fe(OH)₂. If it is assumed that all the activity which cannot be removed by filtration is associated with such ionic material, the specific activity is found to be approximately that of the water-borne particulate material (Table 6.5). This is consistent with radiochemical exchange between these two forms of water-borne material.

Radiochemical Exchange. More recent results have provided further evidence that significant radiochemical exchange takes place between the two fractions of water-borne activity, that is, the filterable crud fraction and the nonfilterable, apparently ionic, fraction. This is indicated by the similarity of the $\text{Fe}^{55}/\text{Fe}^{59}$ and the $\text{Co}^{60}/\text{Co}^{58}$ activity ratios for the two fractions (Fig. 6.12) and also by the similarity in other nuclide ratios (Table 6.6). In particular, if it is assumed that complete radiochemical exchange has occurred, the distribution ratio, X_i , of a given nuclide specific activity, A_i , between the filterable fraction, s , and the nonfilterable fraction, w , should be a constant, that is,

$$X_i = A_i^s / A_i^w .$$

Fig. 6.12. Nuclide Activity Ratios in Loop Coolant and Fuel Rod Deposits.

Unfortunately such X_i ratios cannot be determined directly from the present results because concentrations in the nonfilterable fraction (in grams per liter) have been too low for direct measurement. The constancy of X_i can, however, be tested by considering the ratio of two X_i values:

$$\frac{X_1}{X_2} = \frac{A_1^S/A_1^W}{A_2^S/A_2^W} = \frac{A_1^S/A_2^S}{A_1^W/A_2^W}$$

It may be seen that the ratio X_1/X_2 is equivalent to a ratio of known quantities, that is, the ratio of the two different nuclide specific activities in each of the water fractions. Such ratios, listed in Table 6.9, were found to be reasonably constant in those cases for which sufficient data were available for their calculation.

Table 6.9. Relative Distribution of Activities Between Filterable and Nonfilterable Water Fractions^a

Sampling Date	Activity Ratios			
	$X_{Fe^{55}}/X_{Fe^{59}}$	$X_{Co^{60}}/X_{Co^{58}}$	$X_{Fe^{59}}/X_{Co^{58}}$	$X_{Mn^{54}}/X_{Co^{58}}$
11-9-61	0.92	1.6	4.4	
1-8-62	0.72	1.7	2.0	1.6
3-19-62	0.6	0.5	1.9	1.0

^aBased on data of Table 6.6.

Activity Transport Mechanism

The small amount of particulate crud in the coolant represents only a small fraction of the coolant activity, and thus is not expected to play as great a role in the transport of activity in the system during normal operation as does the nonfilterable, presumably ionic, material. Indeed, from the present results, it appears that this dissolved material is the principal agent of activity transport in well-conditioned, neutral-pH, pressurized-water reactor systems during periods of normal, undisturbed operation. This transport process would consist of the liberation of activity from the in-flux crud deposits as ionic material, which then would

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Fig. 6.13. Assumed Mechanism of Activity Transport in ORR Loop.

The following rate processes occur, all in atoms of nuclide per second:

Gain by activation

Loss by decay

Loss by purification (assumes absolute removal by purification system)

exchange with crud deposits throughout the system. It would at the same time account for the well-crystallized appearance of the crud.

The simple model shown in Fig. 6.13 was developed to permit an analysis of this transport process. A system of three regions is assumed: (1) the volume V^c (cm^3) of crud deposited on in-flux surfaces, (2) the

$$\sigma_a N_a^c \phi V^c$$

$$\lambda_b N_b^c V^c$$

$$\lambda_b N_b^w V^w$$

$$\lambda_b N_b^s V^s$$

$$P N_b^w V^w$$

$$\begin{array}{ll}
 \text{Release to coolant from} & \left\{ \begin{array}{l} RN_b^C V^C \\ RN_b^S V^S \end{array} \right. \\
 \text{surface} & \\
 \\
 \text{Gain to surface from} & \left\{ \begin{array}{l} DN_b^W V^C \\ DN_b^W V^S \end{array} \right. \\
 \text{coolant} &
 \end{array}$$

The values of N_b^C and N_b^W are known from measurements. Since N_b^S is not known directly, it is taken to be equal to the corresponding quantity in the water-borne particulate material; that is, it is assumed that the water-borne crud is typical of out-of-flux crud deposits. This seems reasonable, since the out-of-flux surface area is about 50 times the in-flux surface area.

Differential equations describing the change with time of N_b^C , N_b^W , and N_b^S can be written; however, the solution of such simultaneous equations is quite involved, and the resulting expressions are complex. Culver¹⁰ has presented a complete solution for a somewhat simpler model, and Bergmann⁷ has solved a set of differential equations for a more complex model, using an analog computer.

For the present purpose, it is sufficient to examine the steady-state condition that has been reached in the loop, that is, the condition wherein the concentration terms N_b^C , N_b^W , and, presumably, N_b^S are approximately constant. This permits a simple balancing of the gain and loss processes associated with each of the three regions:

$$\sigma_a N_a^C \phi V^C + DV_b^C N_b^W = RV_b^C N_b^C + \lambda_b V^C N_b^C, \quad (1)$$

$$RV_b^C N_b^C + RV_b^S N_b^S = DV_b^C N_b^W + DV_b^S N_b^W + \lambda_b V^W N_b^W + PV_b^W N_b^W, \quad (2)$$

$$DV_b^S N_b^W = RV_b^S N_b^S + \lambda_b V^S N_b^S. \quad (3)$$

In order to consider first the ratio N_b^S/N_b^W , the distribution of a nuclide

¹⁰H. N. Culver, "Cooling Water and Demineralizer Activities in NMSR," USAEC Report ORNL CF 58-5-83, Oak Ridge National Laboratory, May 14, 1958.

between the coolant and the crud on the out-of-flux surface, Eq. (3) can be reduced to

$$\frac{N_b^S}{N_b^W} = \frac{D}{\lambda_b + R} \sim \frac{D}{R}. \quad (4)$$

It appears that R is considerably greater than λ_b , which has a maximum value of 10^{-7} sec^{-1} for the nuclides being considered, since the data indicate that A_b^S/A_b^W and hence N_b^S/N_b^W are about the same for both long-lived and short-lived nuclides of the same element (Table 6.9). Therefore the ratio is not sensitive to the value of λ in the region of interest.

In order to consider next the ratio N_b^C/N_b^S , Eqs. (2) and (4) yield

$$\frac{N_b^C}{N_b^S} = 1 + \left(1 + \frac{V^S}{V^C}\right) \frac{\lambda_b}{R} + \frac{V^W(\lambda_b + P)(R + \lambda_b)}{DRV^C}. \quad (5)$$

Since $P \gg \lambda_b$ for the nuclides being considered, and $R > \lambda_b$, it follows that

$$\frac{N_b^C}{N_b^S} \sim 1 + \left(1 + \frac{V^S}{V^C}\right) \frac{\lambda_b}{R} + \frac{V^WP}{V^CD}. \quad (6)$$

The ratio V^S/V^C may be taken as the ratio of the area out of flux to that in flux, or ~ 50 . It may be seen then from Eq. (6) that if λ_b/R for shorter lived activities ($\text{Fe}^{59}, \text{Co}^{58}$) is greater than ~ 0.01 , the ratio of each short-lived activity in the fuel rod deposits to that in the out-of-flux surfaces will be significantly greater than for the longer lived activities ($\text{Fe}^{55}, \text{Co}^{60}$). As noted previously, this is indeed the case, and hence $10^{-5} > R > 10^{-7}$; however, R was estimated more closely by comparing activity ratios for each of the four nuclides Fe^{59} , Co^{58} , Fe^{55} , and Co^{60} in the two regions. The latter estimate gave $R \sim 5 \times 10^{-6}$. From the

ratio $N_b^S/N_b^W \sim D/R \sim 6.3 \times 10^8$, this gives $D \sim 3100$. By considering only the long-lived Fe^{55} and Co^{60} activities, for which $N_b^C/N_b^S \sim 1$, Eq. (5) reduces to

$$\frac{N_b^C}{N_b^S} = 1 + \frac{V^W P}{V^C D} \sim 1 + \frac{0.013}{V^C}, \quad (7)$$

since $V^W = 1.1 \times 10^5$ and $P = 3.7 \times 10^{-4}$ for the ORR loop. Thus V^C is evidently considerably greater than 0.01, perhaps 0.1 cm^3 or more. Since the in-flux area of the loop is $2.5 \times 10^3 \text{ cm}^2$, it follows that the average thickness of the layer of exchangeable crud is $\sim 4 \times 10^{-5} \text{ cm}$ (0.4 μ) or more, which is a reasonable lower limit, in view of the observed size of the crud crystals.

To sum up these considerations, it can be said that the observed specific activities and nuclide ratios in the flux, in the soluble material, and in the water-borne crud are consistent with the simple model depicted in Fig. 6.13. The transport of activity associated with dissolved material in the coolant, corresponding to total concentrations of the order of 10^{-7} molar, yields exchange-rate constants of the order of $R = 5 \times 10^{-6} \text{ sec}^{-1}$ for release and $D = 3.1 \times 10^3 \text{ sec}^{-1}$ for deposition. With an estimated crud layer thickness of 0.4 μ or more, this value of R corresponds to a dissolution rate ($R \times$ thickness of crud layer) of $2 \times 10^{-10} \text{ cm/sec}$ or more. This is about three orders of magnitude greater than the expected limiting corrosion rate for conditioned stainless steel (1 mg of stainless steel per cm^2 per month or approximately $7.5 \times 10^{-13} \text{ cm}$ of crud per second). It is not unreasonable, however, to expect the "crud release rates" to be much higher than the net corrosion rates.

The ratio D/R , here determined to be $\sim 6 \times 10^8$, is a constant that is expected to be applicable to other similar pressurized-water systems which use neutral pH water at the same temperature. The significance of the individual values of D and R , in units of sec^{-1} , is less clear. In general, the dissolution rate (atoms/ $\text{cm}^2 \cdot \text{sec}$, or cm/sec) should be a constant that is characteristic of a given crystalline solid at a given temperature; however, in a crud layer the effective area for dissolution

could be the total area of the crystallites. This might be more nearly proportional to the product of the area of the deposit times its thickness than equal to the area alone. Then the significant rate constant would not be in cm/sec but, rather, cm/sec·cm or just sec⁻¹. Thus it is possible that R and, by the same argument, D are the significant constants at a given temperature for pressurized-water systems containing dispersed crud deposits.

Crud Balance. As noted above, the rate at which material is released from crud deposits appears much greater than the rate at which the system metal corrodes. For this reason the production rate of crud and of transportable activity by corrosion has thus far been neglected. It is, nonetheless, of considerable interest to know whether over a long period of time the total amount of crud in the system will increase, decrease, or remain nearly constant at some steady-state level.

Crud is produced continuously in the system, the amount being given in cm³/sec by considering the product of the corrosion rate constant, C (cm of crud per second), and the system area, A (cm²). This crud is transported about the system by the ionic dissolution and deposition exchange process, as has been assumed. Since it is assumed to be lost from the system only by passage to the purification system, it follows that in order for a steady-state crud level to exist, that is, for the total amount of crud in the system to remain constant,

$$CA = \frac{PV^w}{D/R} . \quad (8)$$

Here, D/R, as before, may be assumed to represent the ratio of concentration in crud to that in the coolant. From this it is seen that a constant amount of crud will be present in the system only when

$$P \approx 6.3 \times 10^8 \frac{CA}{V^w} .$$

If P is greater than this, crud is removed by the purification system faster than it is produced; if P is less, crud is formed by corrosion

faster than it is removed. In the present case, assuming a corrosion rate of 1 mg of stainless steel per dm^2 per month ($\sim 7.5 \times 10^{-13}$ cm of crud per second), a purification rate of $> 6 \times 10^{-4} \text{ sec}^{-1}$ is required to assure that the crud can be removed faster than it is produced. The actual purification rate ($3.7 \times 10^{-4} \text{ sec}^{-1}$) is close to this value, suggesting that an approximately steady-state crud level does exist in this system.

Decontamination by Magnetite. The evidence that ionic material is present in a pressurized-water system of neutral pH and the suggestion that it can be responsible for significant activity transport emphasizes the potential value of a high-temperature filter and ion-exchange medium that would perform the same function as the low-temperature, organic, ion exchangers presently used in the side-stream purification system of the ORR loop (and used generally in pressurized-water reactors). If a suitable inorganic ion-exchange material could be found for use in such a purification unit, it might be very effective in removing the ionic activity, which, presumably, is supplied to the loop water by dissolution and ion exchange processes involving crud and corrosion products throughout the system.

Since magnetite (Fe_3O_4) is the stable oxide of iron in such systems, and since it is a principle constituent of crud, it is a possible filter and ion exchange medium. If, as is suggested here, this constituent of crud both supplies and exchanges with the ionic material in the high-temperature water, the introduction of a bed of nonactivated magnetite into the system might well, by the same process which now contributes to the transport of activity, accumulate the dissolved activity without otherwise disturbing the chemical or nuclear behavior of the system.

In terms of the present assumed model of the activity transport process, the presence of a bed of natural magnetite in the system would greatly increase the value of V^S in Eq. (5). The resulting new steady state, when reached, would produce an increase in the ratio N_b^C/N_b^S . In particular, since N_b^C is also related to N_b^S by

$$N_b^c = \frac{\sigma_a N_a^c \phi}{R + \lambda_b} + N_b^s \quad (9)$$

(from a combination of Eq. 1 and 3), then by combination with equation (6), N_b^s is given by

$$N_b^s = \frac{\sigma_a N_a^c \phi}{\left(1 + \frac{V^s}{V^c}\right) \lambda_b + \frac{RPV^w}{DV^c}} \sim \frac{\sigma_a N_a^c \phi V^c}{V^s \lambda_b + (R/D)PV^w} \quad (10)$$

The activity of a given nuclide in out-of-flux deposits (dps/cm³) is thus

$$N_b^s \lambda_b \sim \frac{\sigma_a N_a^c \phi V^c}{V^s + \frac{(R/D)PV^w}{\lambda_b}} \quad (11)$$

It is clear that the activity per unit volume of system crud, $N_b^s \lambda_b$, is decreased by increasing V^s , as well as by increasing P , the relative effectiveness of the two depending on λ_b . In the present system

$$(R/D)PV^w = 6.5 \times 10^{-8}.$$

For Co⁶⁰ ($\lambda = 4 \times 10^{-9} \text{ sec}^{-1}$), an increase in V^s of $\sim 15 \text{ cm}^3$ (as inactive exchangeable magnetite introduced into the system) would thus reduce the activity of deposited crud as effectively as would a doubled purification rate.

It might be argued that such a bed of magnetite, since it represents an increase in the amount of crud in the system, would increase water-borne crud levels and thus produce greater fouling and activity transport. Such is not expected to be the case provided the magnetite is of suitably large particle size and adequately contained, since the present arguments

would suggest that the levels of ionic material in the coolant would remain unchanged (fixed by R/D).

Engineering tests at the Westinghouse Bettis Plant have already shown that magnetite beds are practical and effective as mechanical filters in removing water-borne crud at the operating temperatures of a pressurized-water loop.¹¹ In addition, magnetite is ferromagnetic, with a curie temperature (585°C) well above the operating temperature of pressurized-water reactor primary systems. This suggests the possibility of designing a remotely operated disposable-renewable filter in which the filter bed is supported by a magnetic field, similar to the unit described by McNamee and Shappert.¹² The presence of a magnetic field might also improve the retention of crud, which is magnetic.

To explore further the possibilities of magnetite as a decontaminant, experiments were performed in which a high-temperature loop water bleed stream was passed through a cartridge containing ~100 g of magnetite contained in a bomb. Results for two kinds of magnetite (Table 6.10) showed quite favorable decontamination factors for loop water activity, most of which was nonfilterable.

¹¹C. P. Paulson, "Performance of Magnetite Bed Filters," Chem. Eng. Prog., 56(3): 64 (1960).

¹²R. J. McNamee and L. B. Shappert, "An Engineering Evaluation of a Magnetite Filter as a Means of Remote Control Filter," USAEC Report ORNL CF 58-5-32, Oak Ridge National Laboratory, May 1958.

Table 6.10. Decontamination of Loop Water by Magnetite in High-Temperature (425°F) Filter Unit

Date	Magnetite	Flow Rate (ml/min)	Activity Removed (%)		
			Mn ⁵⁶	Co ⁵⁸ + Mn ⁵⁴	Co ⁶⁰ + Fe ⁵⁹
7-5-62	Prince Mfg. Co., +80 mesh	60	>99	>99	95
9-11-62	Mapico Black, sintered	80	>99	97	98

Conclusions

The observations reported here suggest that, in pressurized-water reactor systems of neutral pH, dissolved ionic material present at very low concentrations is responsible for appreciable chemical and radio-chemical exchange that results in the recrystallization of crud and in the transport of activity from in-flux to out-of-flux crud deposits. A simplified treatment of this process, assuming steady-state conditions, permits estimates of the release and deposition rates. This treatment further allows estimation of the purification rates required to remove crud as rapidly or more rapidly than it is formed. It also indicates that magnetite could be used effectively as a high-temperature decontaminating agent in such systems, and this indication is supported by initial test results.

It should be emphasized that the assumed kinetic model is simple. The release and deposition rate processes have been assumed to be unaffected by the reactor flux, the system has been assumed to be isothermal, and the effects of irreversible deposition of activity on system surfaces have not been considered. The extent to which these effects can be neglected cannot be judged without further investigation; it can be said that, in the present system, temperature variation is not great and the exchange rates involving the dissolved material are probably rapid compared with the rates of irreversible deposition. In applying an analogous treatment to pressurized-water reactors, effects of varying temperature and, in particular, higher pH on release and deposition processes involving dissolved material should be examined.

7. IRRADIATION TESTING AND POSTIRRADIATION EXAMINATION OF FUEL AND ABSORBER MATERIALS

Postirradiation Examination of Specimens from BMI-32 High-Burnup Capsules

A. E. Richt L. D. Schaffer

The SM-2 reactor was designed by Alco Products, Inc., for the Division of Reactor Development, Army Reactors Branch, U.S. Atomic Energy Commission. The materials development required was initiated by Alco Products and contracted in part to Battelle Memorial Institute. The BMI work included fabrication development and irradiation testing of UO_2 fuel dispersed in type 347 stainless steel plates clad with type 347 stainless steel.

Ten NaK-filled capsules that contained a total of 62 fuel specimens were irradiation tested. The material variables of the specimens were:¹

1. fuel concentration — nominally 26 and 40 wt % UO_2 and 34 wt % UN,
2. type of fuel — hydrothermal UO_2 and spherical UO_2 and UN,
3. concentration of dispersed neutron poisons, such as B_4C , NbB_2 , and ZrB_2 ,
4. stainless steel core frame with and without alloyed B^{10} ,
5. fuel core preparation, that is, sintered or green pressed.

The capsules were inserted into the MTR and ETR for testing. Four capsules, BMI-32-1, -2, -3, and -4, were discharged from the reactor in the spring of 1960. BMI performed the postirradiation examinations, and the results were reported.¹ The samples were irradiated at a surface temperature near 700°F and accumulated total U^{235} depletion that ranged from 20 to 45%. Of the 28 specimens in the four capsules, three failed to survive in good physical condition. It should be noted that each of the three specimens which failed was located at an end position in a capsule and that each exhibited signs of overheating, which appears to be the primary reason for failure.

¹S. J. Paprocki et al., "Fabrication and Irradiation of SM-2 Core Materials," USAEC Report BMI-1528, Battelle Memorial Institute, July 12, 1961.

The remaining six of the ten capsules were retained in the ETR until the fall of 1961, and the last capsule was discharged from the reactor during November 1961. The capsules arrived at ORNL in late December 1961 for postirradiation examination, and since this work intimately involved personnel of the Army reactors group, Alco Products, Inc., and Battelle Memorial Institute, the examinations of the capsules were carried out in close cooperation with these groups.

Capsule Design

The capsule employed in this irradiation test program was designed to maintain a specimen surface temperature of 600°F for the design specimens,² which contained 26 wt % UO₂ highly enriched (93%) in U²³⁵. As shown in Fig. 7.1, the capsule was composed of two separate compartments. The upper compartment contained four fuel specimens, and the lower compartment contained either two fuel specimens or a single specimen that was a composite of a fuel specimen and a Eu₂O₃ neutron-flux-suppressor section. The suppressor sections were included to establish the compatibility and general behavior of a UO₂ specimen and a Eu₂O₃ dispersion adjacent to each other.

Both compartments contained split cylindrical copper blocks. These blocks were positioned on either side of the specimens so as not to touch them. Both compartments were filled with NaK, except for a space at the top, which was filled with helium. The specimens were supported in their compartments between the copper blocks by wire hangers. Attached to the hangers were 18-mil nickel-cobalt dosimeter wires (0.5 wt % cobalt). These wires were centered vertically on both sides of each specimen and extended over the entire length of the specimen. The bottom cap of each compartment was countersunk to facilitate puncturing the capsules for fission-gas release.

The lower capsule compartment contained no thermocouples, but the upper compartment contained four thermocouples. These thermocouples were

²D. B. Hamilton et al., "Irradiation-Capsule Study for SM-2 Reference and Alternate Dispersion Fuels," USAEC Report BMI-1916, Battelle Memorial Institute, May 25, 1961.

Fig. 7.1. Simplified Sketch of a BMI-32 Capsule Showing Fuel Specimen Location.

embedded in the copper block adjacent to the fuel specimens. The thermocouples were placed in the capsule from the top, with each lead terminating at the centerline of one of the four specimens.

The readings of these thermocouples in each of the top compartments of the six capsules that remained in the ETR until the fall of 1961 were monitored during the entire irradiation testing period.² Since these capsules were to be exposed for a very long period of time, auxiliary electrical heaters were wound around the outside of the copper blocks in the upper sections. When the fission-heat generation in the instrumented section of the capsule was below that required to maintain the fuel specimen surface at 600°F, the heaters were switched on.

Fission-Gas Sampling

When the capsules were received at ORNL, they were first examined visually and photographed in cell 3 in Building 3025. They were then transferred to the "D" cells in Building 3026, where the upper and lower compartments were cut apart. Each of these fuel compartments was then inverted and placed in the fission-gas-sampling rig. When a given compartment was secure in the rig, it was punctured by drilling at the point which was countersunk in the bottom cap, as shown in Fig. 7.1, and any gas in the compartment was removed into an evacuated flask. Samples that contained approximately 0.3% of the original volume collected were then transferred to glass vials and analyzed for Kr^{85} by gamma-ray spectrographic techniques.

The fission-gas collection rig was made up of a valved piping system attached to a vacuum pump and a puncturing stand. The gas-collection flask was attached to the piping system, isolated by valves, and then evacuated. Next, the capsule compartment was placed in the puncturing stand and a pressure seal was made on the top of the capsule section. This portion of the system was then evacuated. At this point, the puncturing-stand system was opened to the collection system, and a drill rotating through a seal in the puncturing stand was applied to the cap of the capsule compartment. When the drill penetrated the cap, the gas in the compartment was released to the evacuated system. When the pressure buildup of the system leveled off, the collection flask was valved off and removed for further sampling.

The results of the gas analyses are summarized in Table 7.1. Since partial loss of vacuum was detected during the drilling operation on several of the capsule sections, it is felt that these results are not precise and that they should be considered as only qualitative.

The measurements reported in Table 7.2 correlate with the loss of cladding integrity of the fuel specimens in all cases except the bottom of capsule 6 and the top and bottom of capsule 7. In the case of the bottom section of capsule 6 and the top section of capsule 7, heavy crud buildup was found at the bottom of the capsule section. Since the capsule compartments were designed to be punctured from the bottom, the

Table 7.1. Analysis for Kr^{85} in the Gas Released from Capsules BMI-32-5, -6, -7, -8, -9, and -10

Capsule No.	Compartment	Kr^{85} Activity (dps) ^{a, b}	Decay Factor ^c	Atoms of Kr^{85} Present When Capsule Was Removed from the Reactor
5	Top	2.7×10^6	1.025	4.4×10^{17}
	Bottom	$<4 \times 10^3$ (d)	1.025	$<6.5 \times 10^{14}$
6	Top	$<4 \times 10^3$ (d)	1.049	$<6.6 \times 10^{14}$
	Bottom	$\sim 4 \times 10^3$	1.049	6.6×10^{14}
7	Top	$<4 \times 10^3$ (d)	1.025	$<6.5 \times 10^{14}$
	Bottom	$<4 \times 10^3$ (d)	1.025	$<6.5 \times 10^{14}$
8	Top	$<4 \times 10^3$ (d)	1.049	$<6.6 \times 10^{14}$
	Bottom	$<4 \times 10^3$ (d)	1.049	$<6.6 \times 10^{14}$
9	Top	1.7×10^4	1.031	2.8×10^{15}
	Bottom	(e)	1.031	
10	Top	$<4 \times 10^3$ (d)	1.025	$<6.5 \times 10^{14}$
	Bottom	$<4 \times 10^3$ (d)	1.025	$<6.5 \times 10^{14}$

^aSamples counted on March 29, 1962.

^bSample analyzed represented approximately 0.3% of the volume of gas withdrawn from the capsule.

^cThis factor accounts for the decay of the Kr^{85} atoms from the time the capsules were removed from the reactor (see Table 7.5) until the gas samples were counted.

^dBecause of the high-energy beta emission of Ar^{39} and the branching ratio of Kr^{85} , the Kr^{85} detection limit was 4×10^3 dps.

^eThis sample was contaminated with what appeared to be highly activated stainless steel. This material probably came from some drilling chips blown into the flask by the initial release of the gas from the capsule compartment.

samples were punctured in a position inverted to their normal operating position. It is postulated that the low gas release obtained from the two compartments occurred because the drill, although it penetrated the end cap, did not completely penetrate the highly dense crud packed in the bottom of these two capsules. This crud buildup was found to be associated with compartments that contained disintegrated or burnt-through samples. The fact that no fission gas was detected in the lower compartment of

capsule 7, even though the sample was blistered, was subsequently explained when no cracks or penetrations were found in the cladding.

Capsule Opening and Removal of Specimens

After the gas in each capsule compartment was sampled, the compartment was opened and the fuel specimens were removed. Initially, after the end caps were cut off and the NaK was reacted with a mixture of alcohol and oil, an attempt was made to push the copper blocks and fuel specimens from the capsule compartment. This operation proved to be quite difficult, and thereafter the capsule side walls were slit following the removal of the ends caps. With the wall slit, the capsule sections could be pulled away from the copper blocks. The copper blocks from the lower compartment were separated easily by cutting the two wires wrapped around the blocks. Separating the copper blocks from the upper compartment proved to be a more difficult task. These blocks were finned and had heater wires wrapped and brazed into the fin recesses. The location of the wires made cutting difficult. Since the wires were quite brittle, they would break easily and could not be pulled loose. These copper blocks were finally separated by partially slitting them to cut all the wires. With this accomplished, the blocks could be separated to expose the test specimens.

The specimens were visually inspected when first removed from the capsules. Capsules 8 and 10 were the only ones in which all the specimens survived in good condition. The upper section of capsule 9 as it appeared when the copper blocks were first separated is shown in Fig. 7.2. The upper two samples had partially disintegrated; the third specimen was blistered and highly discolored; and the lower specimen appeared only to have suffered surface discoloration. All the specimens removed from the capsules were found in a recognizable form, except the specimens in the bottom compartment of capsule 6. The inside surface of a copper block where the fuel samples should have been located is shown in Fig. 7.3. Apparently this portion of the capsule had been severely overheated, possibly to the melting point of the samples. There was no NaK evident in the capsule compartment; rather, there was a dark-red coarse-grained mud-like deposit. Since the upper section of the capsule did not show such

Fig. 7.2. Specimens in Top Compartment of Capsule 9.

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R-9034

Fig. 7.3. Inner Face of One of the Copper Blocks from the Lower Compartment of Capsule 6.

effects, it is believed that this bottom section experienced a partial or complete loss of its NaK sometime after its manufacture. No evidence exists, however, to indicate when such loss occurred.

At this point, the plan of examination of the individual specimens was determined, as described in Table 7.2.

Gamma Scanning

Because some fuel samples had nearly a 50% greater fuel loading than others, there was interest in determining whether significantly greater self-shielding occurred in the specimens with the higher fuel loading. This was determined by measuring the relative gamma activity of each fuel specimen during the postirradiation examination. Since the fuel specimens were small, no scanning was attempted; rather, each specimen was placed in the gamma-scanning device and the relative total energy of the gamma rays being emitted from each specimen was measured. These measurements, along with the fuel loading in atoms of U^{235} per cm^3 , and the fuel burnup in fissions per cm^3 , computed from dosimetry measurements, are included in Table 7.3. For convenience of comparison, the three groups of values are normalized for each capsule. For the most part the variation in fission density of the specimens within a given capsule follows the fuel concentration, and perhaps to a less quantitative degree the gamma intensity varies with the fission density. At these fuel loadings it appears that the gamma intensities which are indicative of the power generation of the specimen near the end of the test are only a rough quantitative measure of fission density. Further, in these cases it appears that the fission density is essentially determined by the fuel loading and that self-shielding is not particularly apparent, except possibly for the heavier fuel loadings in capsule 5.

Measurements of Physical Dimensions and Density

The physical dimensions and density of each fuel specimen were determined, with the exception of the two specimens that completely disintegrated in the bottom compartment of capsule 6 and the two top specimens in the upper compartment of capsule 9, which partially disintegrated. The density determination of each measurable specimen was made by determining

Table 7.2. Plan for Examination of Specimens from Capsules BMI-32-5, -6, -7, -8, -9, and -10 Following Irradiation in ETR

Capsule No.	Specimen Position	Specimen No.	Examination Plan
5 ^a	A	2-13	Perform metallographic examination, fuel burnup analysis, and hardness test
	B	2-15B	Same as above; store one-half of specimen, if possible, for future study
	C	2-6A	Store entire specimen
	D	5-17B	Same as for specimen 2-15B
	E	4-5B	Perform bend test, metallographic examination, and fuel burnup analysis
	F	5-11	Same as for specimen 2-13
6	A	3-1A	Store one-half of specimen, if possible, for future study; perform metallographic examination, fuel burnup analysis, and hardness test
	B	2-4A	Perform annealing test, metallographic examination, and fuel burnup analysis
	C	3-9A	Same as above
	D	4-13A	Same as above
	E	3-15B	Specimen completely disintegrated
	F	1-16B	Specimen completely disintegrated
7	A	4-1A	Store one-half of specimen, if possible, for future study; perform metallographic examination, fuel burnup analysis, and hardness test
	B	3-4A	Perform metallographic examination, fuel burnup analysis, and hardness test
	C	3-5C	Same as above plus annealing test
	D	3-6V	Store one-half of specimen, if possible, for future study; perform metallographic examination, fuel burnup analysis, and hardness test
	E	2-16B	Same as above
	F	3-17B	Same as above
8	A	1-1A	Store one-half of specimen, if possible, for future study; perform metallographic examination, fuel burnup analysis, hardness test, and waterlogging test
	B	1-4A	Perform metallographic examination, fuel burnup analysis, and hardness test
	C	7-13	Same as above
	D		Same as above plus bend test on frame, which contains B ¹⁰
	E		Store one-half of specimen, if possible, for future study; double specimen containing fuel in upper section and a Eu ₂ O ₃ suppressor below; perform metallographic examination, fuel burnup analysis, hardness test, and waterlogging test
9 ^a	A	2-1A	Store one-half of specimen, if possible, for future study; perform metallographic examination
	B	5-5B	Perform metallographic examination
	C	2-1A	Store one-half of specimen, if possible, for future study; perform metallographic examination, fuel burnup analysis, and hardness test
	D	3-13A	Perform metallographic examination, fuel burnup analysis, and hardness test
	E	3-8A	Store one-half of specimen, if possible, for future study; double specimen containing fuel in upper section and a Eu ₂ O ₃ suppressor below; perform metallographic examination, fuel burnup analysis, hardness test, and waterlogging test
10	A	4-3	Perform metallographic examination, fuel burnup analysis, and hardness test
	B	4-5C	Same as above plus waterlogging test
	C	1-6A	Same as above; store one-half of specimen, if possible, for future study
	D	2-9A	Same as for specimen 4-3
	E	1-2C	Same as for specimen 4-3; store one-half of specimen, if possible, for future study
	F	3-14A	Same as for specimen 4-3 plus bend test on frame, which contains B ¹⁰

^aAn Ni⁵⁸(n,p)Co⁵⁸ reaction analysis was planned to obtain a fast-flux determination.

Table 7.3. Gamma-Activity Measurements of Specimens from Capsules BMI-32-5, -6, -7, -8, -9, and -10

Capsule No.	Specimen Position	Specimen No.	Initial U ²³⁵ Concentration (atoms/cm ³)	Normalized U ²³⁵ Concentration	Relative Gamma Activity	Normalized Gamma Activity	Fuel Burnup (fissions/cm ³)	Normalized Fuel Burnup
			$\times 10^{21}$				$\times 10^{21}$	
5	A	2-13	4.57	1.00	53	1.00	2.51	1.01
	B	2-15B	6.64	1.45	107	1.84	3.57	1.33
	C	2-6A	4.57	1.00	69	1.19	2.48	1.00
	D	5-17B	7.02	1.54	102	1.76	3.71	1.50
	E	4-5B	4.57	1.00	69	1.19	2.53	1.02
	F	5-11	4.57	1.00	71	1.22	2.83	1.14
6	A	3-1A	4.22	1.00	14	1.00	1.91	1.00
	B	2-4A	4.57	1.08	16	1.14	2.16	1.13
	C	3-9A	4.57	1.08	18	1.29	2.17	1.14
	D	4-13A	4.57	1.08	18	1.29	2.23	1.17
	E	3-15B	6.64	1.57				
	F	1-16B	7.02	1.66				
7	A	4-1A	4.22	1.00	54	1.00	2.24	1.00
	B	3-4A	4.57	1.08	54	1.00	2.49	1.11
	C	3-5C	4.57	1.08	57	1.05	2.55	1.14
	D	3-6V	4.57	1.08	59	1.09	2.42	1.08
	E	2-16B	7.02	1.66	88	1.63	3.92	1.75
	F	2-17B	7.02	1.66	88	1.63	3.64	1.63
8	A	1-1A	4.22	1.00	17	1.00	2.10	1.00
	B	1-4A	4.57	1.08	19	1.12	2.29	1.09
	C	7-13	4.57	1.08	18	1.06	2.37	1.13
	D	2-14A	4.57	1.08	21	1.23		
	E	1-8A	4.57	1.08	18	1.06	2.49	1.19
	E'	Eu ₂ O ₃			62	3.65		
9	A	2-1A	4.22	1.00			2.18	1.00
	B	5-5B	4.57	1.08			2.39	1.10
	C	2-7A	4.57	1.08	25	1.00	2.39	1.10
	D	3-13A	4.57	1.08	29	1.16	2.84	1.30
	E	3-8A	4.57	1.08	25	1.00	3.07	1.41
	E'	Eu ₂ O ₃			50	2.00		
10	A	4-3A	4.57	1.00	60	1.03	2.32	1.00
	B	4-5C	4.57	1.00	62	1.07	2.41	1.04
	C	1-6A	4.57	1.00	66	1.14	2.32	1.00
	D	2-9A	4.57	1.00	62	1.07	2.44	1.05
	E	1-12C	4.57	1.00	58	1.00	2.54	1.09
	F	3-14A	4.57	1.00	61	1.05	2.52	1.09

the difference of the specimen weight in air as compared with its weight when immersed in carbon tetrachloride at room temperature. The results of these measurements are listed in Table 7.4.

The postirradiation dry weights of these specimens checked satisfactorily with the preirradiation weights, except for specimen 5B, for which the postirradiation weight was much greater than the preirradiation weight. Since this specimen had blistered significantly, it was concluded that the cladding had lost its integrity and had allowed NaK to flow into the cavity formed by the blister. This conclusion was further verified when the specimen was immersed in carbon tetrachloride. Each time the specimen was immersed, the subsequent dry weight was different. Therefore the density change listed for specimen 5B in Table 7.4 is only an approximation.

During the capsule opening operation, the bottom ends of samples 9D and 10D were damaged with the cutting saw. The damage did not extend into the fueled region of the specimens but did result in the loss of some of the dead-end stainless steel. This mishap is not considered to have significantly affected the density determination or the density changes observed.

Visual Examination

After removal from the sample holders, the individual fuel specimens were cleaned in alcohol and acetone and subjected to thorough visual examination. Most of the specimens appeared to be in excellent condition, but nine of them were damaged in various degrees. Examples of an undamaged specimen and typical damaged specimens are shown in Fig. 7.4. All six specimens with high fuel loadings failed. The four (2-15B, 2-16B, 3-17B, and 5-17B) that contained 38 wt % UO_2 or 34 wt % UN were badly swollen (Fig. 7.4b), and the other two (3-15B and 1-16B), which were in the uninstrumented compartment of capsule 6, had completely disintegrated. Specimen 4-1A, from the top position in capsule 7, had two holes that completely penetrated both core and cladding (Fig. 7.4c). The three specimens (2-1A, 5-5B, and 2-7A) from the upper positions of capsule 9 were severely damaged (Fig. 7.4d). Also, specimen 3-8A, from the

Table 7.4. Results of Measurements of Physical Dimensions and Density of Specimens from Capsules BMI-32-5, -6, -7, -8, -9, and -10

Capsule No.	Specimen Position	Specimen No.	Specimen Condition	Density Change (%)	Thickness Change (%)	Width Change (%)	Length Change (%)
5	A	2-13	Good	-4.45	+7.77	+0.39	+0.19
	B	2-15B	Blistered	-23.00	+65.90	-0.28	-0.10
	C	2-6A	Good	-4.15	+7.51	+0.10	-0.17
	D	5-17B	Blistered	-30.90	+160.00	-0.10	-0.05
	E	4-5B	Good	-4.94	+8.89	+0.07	-0.01
	F	5-11	Good	-5.04	+9.42	+0.50	-0.05
6	A	3-1A	Good	-2.91	+4.15	-0.05	-0.09
	B	2-4A	Good	-3.31	-5.14	+0.14	+0.07
	C	3-9A	Good	-3.58	+6.44	+0.05	-0.03
	D	4-13A	Good	-3.90	+6.68	+0.37	-0.01
	E	3-15B	Completely disintegrated				
	F	1-16B	Completely disintegrated				
7	A	4-1A	Burnt	-6.36	+20.50	-0.37	-0.09
	B	3-4A	Good	-3.68	+6.70	+0.36	+0.06
	C	3-5C	Good	-4.44	+7.54	+0.47	+0.05
	D	3-6V	Good	-4.28	+7.54	+0.77	-0.14
	E	2-16B	Blistered	-45.90	+273.60	-6.24	-0.03
	F	3-17B	Blistered	-37.70	+201.70	-1.61	-0.09
8	A	1-1A	Good	-3.64	+6.58	-0.00	+0.05
	B	1-4A	Good	-3.60	+5.42	+0.16	+0.10
	C	7-13	Good	-4.04	+6.75	+0.14	+0.13
	D	2-14A	Good	-4.35	+6.80	+0.23	+0.11
	E	1-8A	Good		+2.43	+0.03	(a)
	E'	1-8A	Good	-2.62	-0.49	0.00	(a)
9	A	2-1A	Partially disintegrated				
	B	5-5B	Partially disintegrated				
	C	2-7A	Blistered	-18.60	+72.20	+0.05	-0.00
	D	3-13A	Good	-5.82	+11.50	+0.05	(b)
	E	3-8A	Blistered		+58.20	-0.17	(a)
	E'	3-8A	Good	-15.30	+0.98	0.00	(a)
10	A	4-3	Good	-3.65	+5.50	-0.10	-0.03
	B	4-5C	Good	-3.60	+5.75	+0.19	+0.15
	C	1-6C	Good	-4.02	+7.40	+0.10	-0.03
	D	2-9A	Good	-4.34	+7.55	-0.05	(b)
	E	1-12C	Good	-3.84	+6.02	+0.07	-0.03
	F	3-14A	Good	-4.40	+6.50	+0.12	+0.06

^aSpecimens were too long to be measured lengthwise in the in-cell rig.

^bThe sample end was damaged when the capsule was opened.

uninstrumented compartment of capsule 9 had a large blister over the fueled region of the specimen (Fig. 7.4e).

It is obvious from the appearance of specimens 4-1A, 2-1A, 5-5B, and 2-7A that damage to these specimens was a result of very high temperatures during irradiation. This may have resulted either from a partial loss of NaK from capsules 7 and 9 or a not-yet-understood vapor "blanketing" effect during irradiation.

(a) TYPICAL UNDAMAGED SPECIMEN
(SPECIMEN 1-12C)

(b) TYPICAL SWELLING OF A SPECIMEN WITH A
HIGH FUEL LOADING (SPECIMEN 2-16B)

(c) SPECIMEN 4-1A, NOTE HOLES THROUGH
CORE AND CLADDING

(d) TYPICAL ONE OF TOP THREE SPECIMENS IN
CAPSULE NO.9 (SPECIMEN 5-5B)

(e) SPECIMEN 3-8A, NOTE BLISTER OVER FUELED END OF DUPLEX SPECIMEN

Fig. 7.4. Typical Appearance of BMI-32 Irradiation Specimens. Approx. 2X.

The damage of specimens with high fuel loadings (2-15B, 2-16B, 3-17B, and 5-17B) and specimen 3-8A from capsule 9 may also be partially attributed to the irradiation temperature being significantly greater than 600°F. The greater heat generation rate of these highly loaded specimens in capsules designed for specimens with 26 wt % U^{235} resulted in surface temperatures of approximately 1000°F. The initial test surface temperature of all specimens in capsule 9 was also approximately 1000°F. This was a result of the capsule being inadvertently irradiated in a much higher flux position than originally planned. Thus, it is quite evident that all specimens which showed evidence of external damage were irradiated at temperatures significantly greater than the desired 600°F.

Neutron Dosimetry Analysis

The neutron dosimeters were removed from the capsule specimen support wire during the specimen removal operation. These dosimeters, which were 0.018-in.-diam nickel-0.5 wt % cobalt wires, were attached to the specimen support basket so that the wires were approximately 1/16 in. from the surface of the specimen. A dosimeter was placed parallel to the vertical centerline of each specimen and was sized to extend over the entire length of the specimen. As the dosimeters were removed they were placed in labeled sample bottles. All dosimeter wires were recovered except from the bottom of capsule 6, which had apparently experienced a very high temperature. There was no evidence of the dosimeters in the reaction products in the bottom compartment.

The dosimeters were analyzed radiochemically to determine the quantity of Co^{60} formed during the irradiation test period:

During the radiochemical analysis, the dosimeter from sample position 8D was lost. The results of the analyses of all the other dosimeter wires are listed in Table 7.5 as the specific Co^{60} activity in disintegrations per second per gram of wire. The specific activities reported are corrected to the specific activity that each wire would have had at the time of its removal from the reactor.

Table 7.5. Summary of Irradiation History and Burnup Data for Capsules BMI-32-5, -6, -7, -8, -9, and -10

Capsule No.	Irradiation History of Capsule ^a				Specimen Position	Specimen No.	Fuel ^b		Burnable Poison ^b		Estimated ^{a,c} Peak Surface Temperature (°F)	Predicted ^d U ²³⁵ Depletion (at. %)	Total U ²³⁵ Depletion			From Analyses of Fuel Specimens, U ²³⁵ Depletion (at. %)		
	ETR Position	Insertion Date	Discharge Date	Full Power Irradiation Period (days)			Type	Loading (wt %)	Type	Boron Content (wt %)			From Specimen Dosimetry Data			Isotopic Analysis	Radiochemical Analysis	U ²³⁵ Balance
													Specific Co ⁶⁰ Activity (dps per gram of wire)	Integrated Thermal-Neutron Flux (neutrons/cm ²)	U ²³⁵ Depletion (at. %)			
5	0-6	9-19-60	11-13-61	184	A	2-13	Spherical UO ₂	26	NbB ₂	0.203	690 ^e	78	1.66	2.53	64.4	63.0	64.0	63.8
					B	2-15B	UN	34.1	ZrB ₂	0.315	950 ^e		1.62	2.46	63.2	59.4	57.4	57.7
					C	2-6A	Spherical UO ₂	26	ZrB ₂	0.200	700 ^e		1.63	2.48	63.7			
					D	5-17B	Spherical UO ₂	38.4	ZrB ₂	0.350	950 ^e		1.76	2.37	62.1	61.2	62.9	61.8
					E	4-5D	Hydrothermal UO ₂	26	ZrB ₂	0.197	(f)		1.68	2.56	65.0			
					F	5-11	Hydrothermal UO ₂	26	NbB ₂	0.207	(f)		1.85	2.82	68.5	66.5	71.0	71.5
6	0-5	4-26-60	7-10-61	197	A	3-1A	Hydrothermal UO ₂	24.2	B ₄ C	0.048	500	71	1.20	1.86	53.2	52.2	56.4	55.6
					B	2-4A	Spherical UO ₂	26	None		580		1.27	1.97	55.3	55.2	58.0	56.8
					C	3-9A	Hydrothermal UO ₂	26	NbB ₂	0.222	610		1.28	1.99	55.6	56.9	56.2	59.0
					D	4-13A	Spherical UO ₂	26	NbB ₂	0.204	620		1.33	2.07	57.3			
					E	3-15B	UN	34.1	ZrB ₂	0.315	(f)		Sample disintegrated					
					F	1-16B	Hydrothermal UO ₂	38.4	ZrB ₂	0.430	(f)		Sample disintegrated					
7	M-4	7-8-60	11-13-61	211	A	4-1A	Hydrothermal UO ₂	24.2	B ₄ C	0.048	420	80	1.59	2.39	62.3	63.8	65.4	67.3
					B	3-4A	Spherical UO ₂	26	None		~730 ^g		1.66	2.50	64.0	64.0	65.4	66.4
					C	3-5C	Hydrothermal UO ₂	26	ZrB ₂	0.183	610		1.73	2.60	65.6	63.9	61.3	67.2
					D	3-6V	Spherical UO ₂	26	ZrB ₂	0.213	540		1.59	2.39	62.3	64.7	63.9	65.5
					E	2-16B	Hydrothermal UO ₂	38.4	ZrB ₂	0.430	(f)		1.73	2.60	65.6			
					F	2-17B	Spherical UO ₂	38.4	ZrB ₂	0.350	(f)		1.52	2.29	60.9			
8	L-4	4-26-60	7-10-61	197	A	1-1A	Hydrothermal UO ₂	24.2	B ₄ C	0.048	590	73	1.38	2.14	58.5	58.2	63.2	61.9
					B	1-4A	Spherical UO ₂	26	None		580		1.39	2.16	58.8	59.4	60.9	61.0
					C	7-13	Spherical UO ₂	26	NbB ₂	0.202	480		1.48	2.30	60.9	60.6	65.0	63.7
					D	2-14A	Spherical UO ₂	26	B ¹⁰ (h)	0.098	610		Dosimeter lost			67.2	68.8	67.6
					E	1-8A	Spherical UO ₂	26	ZrB ₂	0.222	(f)		1.60	2.49	64.0	61.9	68.0	67.0
					F ⁱ						(f)		1.74	2.70	66.8			
9	0-9	5-24-60	10-14-61	200	A	2-1A	Hydrothermal UO ₂	24.2	B ₄ C	0.048	1290	95	1.44	2.27	60.6			
					B	5-5B	Hydrothermal UO ₂	26	ZrB ₂	0.197	~910 ^g		1.47	2.32	61.4			
					C	2-7A	Spherical UO ₂	26	ZrB ₂	0.093	800		1.79	2.32	61.4	71.1	69.7	75.8
					D	3-13A	Spherical UO ₂	26	NbB ₂	0.204	1120		2.03	3.20	73.0	73.6	76.8	76.9
					E	3-8A	Spherical UO ₂	26	ZrB ₂	0.222	(f)		2.35	3.70	78.0	82.1	81.9	84.0
					F ⁱ						(f)		2.61	4.11	81.4			
10	K-4	9-19-60	11-13-61	184	A	4-3	Hydrothermal UO ₂	26	None		(j)	77	1.46	2.22	59.7	58.0	61.0	62.8
					B	4-5C	Hydrothermal UO ₂	26	ZrB ₂	0.183	450		1.54	2.34	61.8	58.8	63.4	62.6
					C	1-6A	Spherical UO ₂	26	ZrB ₂	0.200	700		1.46	2.22	59.7	59.8	63.6	60.8
					D	2-9A	Hydrothermal UO ₂	26	NbB ₂	0.222	590		1.59	2.42	62.8	60.2	63.1	62.2
					E	1-12C	Spherical UO ₂	26	ZrB ₂	0.168	(f)		1.70	2.59	65.3	65.1	69.7	71.1
					F	3-14A	Spherical UO ₂	26	B ¹⁰ (h)	0.023	(f)		1.67	2.54	64.8	65.0	67.1	65.7

^aJ. F. Lagedrost and J. H. Stang, Irradiation History of Capsules BMI-32-5 Through BMI-32-10, December 14, 1961 (unpublished).

^bS. J. Paprocki et al., "Fabrication of SM-2 Core Materials," USAEC Report BMI-1528, Battelle Memorial Institute, July 12, 1961.

^cD. B. Hamilton et al., "Irradiation-Capsule Study for SM-2 Reference and Alternate Dispersion Fuels," USAEC Report BMI-1516, Battelle Memorial Institute, May 25, 1961.

^dThis was a prediction of the peak U²³⁵ depletion which could be expected somewhere within the fuel in the capsule.

^ePersonal communication from J. F. Lagedrost, Battelle Memorial Institute, to L. D. Schaffer, Oak Ridge National Laboratory, October 2, 1962.

^fNoninstrumented section; contained no thermocouples.

^gInterpolated from data in reference given in footnote c.

^hBoron-10 was alloyed in the stainless steel picture frame.

ⁱBottom half of a double length specimen containing 22.6 wt % Eu₂O₃ in type 347 stainless steel adjacent to the fueled section.

^jThermocouple failed to operate; see reference in footnote c.

From the specific activity data, integrated thermal-neutron flux values were calculated for each dosimeter. These numbers are also listed in Table 7.5. The neutron cross section used in this calculation was the thermal neutron cross section, that is, 36.3 barns. It should be noted here that this calculation would have been more precise if an effective neutron cross section could have been used to account for the interactions of epithermal neutrons and Co^{59} . Since no cadmium-covered dosimeters were included in the capsules, it was difficult to determine what value should be assigned as the effective neutron cross section. Since it was necessary to use the thermal-neutron cross section, the integrated thermal-neutron flux values are too high, possibly by between 10 and 20%.

The U^{235} depletion was calculated from the integrated thermal-neutron flux values. The dosimetry-derived U^{235} depletion values reported in Table 7.5 were obtained by the following equation:

$$D = (1 - e^{-\phi \sigma_a t}) 100, \quad (1)$$

where

D = percentage of original U^{235} depleted by fission and nonfission neutron interaction,

σ_a = thermal-neutron adsorption cross section for $\text{U}^{235} = 6.83 \times 10^{-22} \text{ cm}^2$,

ϕ = thermal neutron flux (neutrons/ $\text{cm}^2 \cdot \text{sec}$),

t = time (sec).

A factor had to be applied to the neutron flux or integrated neutron flux value of the wire dosimeter in order to account for the depression of the flux from the dosimeter to within the fuel region. The U^{235} depletion of the fuel specimens, as determined by radiochemical and isotopic analyses, was used to correct the integrated thermal-neutron flux (ϕt) computed for the wire dosimeters, so the depletions calculated from the dosimetry results correspond to the chemical and isotopic results. This factor can be considered to be the ratio of the effective neutron flux in the fuel (ϕ_{eff}) to the specimen surface or dosimeter neutron

flux (ϕ_s). The factor selected that best fit the burnup analysis data was

$$\frac{\phi_{\text{eff}}}{\phi_s} = 0.6 \quad (2)$$

U²³⁵ Depletion Analysis

With the exception of the specimens that had completely disintegrated in capsule 6 and a few other specimens being held for special tests, all fuel specimens in the six test capsules were analyzed. They were analyzed chemically for uranium, radiochemically for Cs¹³⁷, and mass spectrographically for each uranium isotope, in order to determine the U²³⁵ fuel depletion (total U²³⁵ burnup, including nonfission captures). The samples cut from the fuel specimens amounted to approximately one-half the fuel region of each specimen. If the fuel burnup profile is considered symmetrical along the length of the specimen, the analytical result represents an averaged determination.

In the isotopic analysis of these samples, the atom fractions of U²³⁴, U²³⁵, U²³⁶, and U²³⁸ were determined. The preirradiation isotopic analyses of the uranium used in these samples were furnished by the Battelle Memorial Institute. The pre- and postirradiation isotopic analysis values for the uranium content of each sample were utilized in the following equation:

$$D = f_1 C_1 \times 100, \quad (3)$$

where

$$D = \text{U}^{235} \text{ depleted (at. \%),}$$

$$f_1 = \frac{(E_0 - E_1)(1 + \alpha)}{E_0(1 + \alpha - E_1)},$$

$$C_1 = 1 + \frac{E_1^{25}}{E_0^{25} - E_1^{25}} \frac{\Delta U_{26} + \Delta U_{28}}{U_0},$$

and

E_0^{25} = atom fraction of U^{235} in uranium before irradiation,

E_1^{25} = atom fraction of U^{235} in uranium after irradiation,

U_0 = total concentration of uranium atoms before irradiation (atoms per gram of sample),

U_1 = total concentration of uranium atoms after irradiation (atoms per gram of sample),

ΔU_{26} = number of absorptions in U^{236} (number per gram of sample),

ΔU_{28} = number of absorptions in U^{238} (number per gram of sample),

$\alpha = \sigma_c / \sigma_f$ = capture-to-fission ratio = 0.20.

Because the fuel in these specimens was highly enriched, ΔU_{26} and ΔU_{28} were considered small, and C_1 was set equal to 1. Therefore Eq. (3) simplified to

$$D = f_1 \times 100 . \quad (4)$$

The fuel depletions computed from the mass spectrographic analysis data by utilizing Eq. (4), in which α was assigned the value 0.20, are listed in Table 7.5.

The radiochemical and chemical analyses were performed in order to determine the fuel depletion by an alternate method. The concentration of the fission product Cs^{137} was determined radiochemically. From this determination, the U^{235} fissions per unit volume of fueled core were determined by the following equation:

$$F_{25} = \frac{A_{cs} F_D^{25} \rho_s}{\lambda_{cs} \gamma_{cs}^{25}} , \quad (5)$$

where

F_{25} = total number of U^{235} fissions per cm^3 of sample,

A_{cs} = specific activity of Cs^{137} (dpm per gram of sample),

γ_{cs}^{25} = fission yield of Cs^{137} from U^{235} = 0.0615,

λ_{cs} = decay constant for Cs^{137} = $4.611 \times 10^{-8} \text{ min}^{-1}$,

ρ_s = density of sample (g/cm^3),

$$F_D^{25} = \frac{\sigma_a^{25} \phi - \lambda_{cs}}{\sigma_a^{25} \phi} \frac{e^{\lambda_{cs} T} \left(1 - e^{-\sigma_a^{25} \phi t} \right)}{\left(e^{-\lambda_{cs} t} - e^{-\sigma_a^{25} \phi t} \right)},$$

and

σ_a^{25} = thermal-neutron absorption cross section of U^{235} (cm^2),

ϕ = thermal-neutron flux (neutrons/ $\text{cm}^2 \cdot \text{sec}$),

t = irradiation time (sec),

T = cooling time (sec).

The total uranium remaining in the fueled core was determined chemically as atoms of uranium per unit volume of fueled core. Based on the Cs^{137} and postirradiation total uranium analysis data and the postirradiation atom percentages of U^{235} and U^{236} obtained from the mass spectrographic analysis, the U^{235} depletion could be determined from the following equation:

$$D = \left(\frac{F_{25} + U_1 E_1^{26}}{F_{25} + U_1 E_1^{25} + U_1 E_1^{26}} \right) 100, \quad (6)$$

where

E_1^{25} = atom fraction of U^{235} in uranium after irradiation,

E_1^{26} = atom fraction of U^{236} in uranium after irradiation.

The values computed by this method are reported in Table 7.5 under the heading radiochemical analysis.

A third method was employed to calculate the U^{235} depletion. This method is referred to as the U^{235} balance. The information required for this computation was the preirradiation and postirradiation U^{235} contents of the fueled core. The postirradiation value was obtained from the post-irradiation isotopic and total-uranium analyses. The U^{235} depletion was computed by utilizing the following equation:

$$D = \left(\frac{U_0^{25} E_0^{25} - U_1^{25} E_1^{25}}{U_0^{25} E_0^{25}} \right) 100. \quad (7)$$

The U^{235} depletion numbers obtained by this method are also listed in Table 7.5.

Metallographic Examination

Metallographic examination revealed no evidence of matrix cracking or blistering in fuel specimens irradiated at temperatures which appeared to have been near 600°F. These specimens, all containing 24.2 or 26 wt % UO_2 , had burnups of 52 to 67 at. % U^{235} (1.9 to 2.6×10^{21} fissions/cm³). No significant difference was noted between specimens fueled with hydrothermal or spherical UO_2 particles or between specimens with the various burnable poisons used. The microstructures of these specimens were quite similar to those found in high-burnup regions of the SM-1 fuel elements.³ As shown in Fig. 7.5, considerable fission-gas porosity was present in the UO_2 particles. The extent of the porosity varied slightly from specimen to specimen, apparently depending on burnup. Two unidentified phases were also found in the microstructure of these samples: a bright, specular material apparently associated with the fission-gas pores, and a gray, oxide-like material predominately around the periphery of the UO_2 particles. Both these phases have been observed in the examination of irradiated SM-1 reactor fuel elements.

Examination of specimen 3-13A from capsule 9 revealed what appeared to be the initiation of matrix cracking (Fig. 7.6). This specimen, with a 26 wt % UO_2 fuel loading, had been subjected to 74% depletion of the U^{235} (2.9×10^{21} fissions/cm³) at approximately 1000°F. Specimen 3-8A, also with 26 wt % UO_2 , was severely blistered after 82% depletion of the U^{235} (3.2×10^{21} fissions/cm³) at approximately the same irradiation temperature. These failures appear to establish a burnup limitation of 74% (depletion) for the 26 wt % UO_2 fuels at temperatures of approximately 1000°F. This limitation should not, however, be applied to these fuels at SM-2 operating conditions (600°F).

Metallographic examination of the specimens with high fuel loadings (38.4 wt % UO_2 and 34.1 wt % UN) revealed severe matrix cracking

³A. E. Richt, Postirradiation Examination of SM-1 Fuel Elements, pp. 33-38 "Army Reactors Program Prog. Rep.," USAEC Report ORNL-3231, Oak Ridge National Laboratory, Jan. 31, 1962.

Fig. 7.5. Typical Microstructure After High Burnup of Specimens Containing 26 wt % UO_2 . As polished. 250X.

(Fig. 7.7). The fission-gas pores appeared to be larger and more spherical in the UO_2 particles than in the UN dispersion. The microstructure of the UO_2 dispersion specimen is typical (Fig. 7.7a) of high-burnup UO_2 dispersions, with the exception of the matrix cracking. As shown in Fig. 7.8, however, UN particles reacted with the stainless steel matrix in specimen 2-15B. Such reaction may be a result of nitriding of the stainless steel by the excess nitrogen released from the highly fissioned UN particles.

Examination of the UO_2 - Eu_2O_3 duplex specimens 1-8A and 3-8A revealed no evidence of structural damage (Fig. 7.9). The Eu_2O_3 particles appeared to be unaffected by irradiation, whereas the UO_2 particles showed significant fission-gas porosity. No evidence of a reaction between the two regions was apparent.

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Fig. 7.6. Microstructure of Specimen 3-13A After Irradiation to 74% Depletion of U^{235} at 1000°F. Note swelling of specimen and matrix cracking in central region. As polished. 100X.

Fig. 7.7. Matrix Cracks in Specimens with High Fuel Loading. (a) Specimen 5-17B, 38 wt % UO_2 , (b) Specimen 2-15B, 34 wt % UN. As polished. 100X.

Fig. 7.8. Evidence of Reaction Between UN and Stainless Steel Matrix.
(a) As polished. (b) Etched with glycerol regia. 250X.

Fig. 7.9. Longitudinal Section Through Fuel and Suppressor Region of Specimen 3-8A. Note difference in porosity of Eu₂O₃ and UO₂ particles. As polished. 100X.

Attempts to prepare the highly damaged specimens in capsule 9 for metallographic examination were unsuccessful; however, a section was made through one of the holes found in specimen 4-1A. This section is shown in Fig. 7.10. It is quite evident that the damage to this specimen was a result of localized surface temperatures high enough to cause melting of the stainless steel.

One perplexing effect noted in the examination of these specimens is shown in Fig. 7.11. This section, taken from the upper end of specimen 3-8A, shows fracturing of the ZrB_2 particles and slight crack propagation into the core matrix. Note that no matrix cracking from the UO_2 particles is apparent. This is contrary to the results of the examination of specimen 3-13A, where matrix cracking apparently originated from

Fig. 7.10. Metallographic Section Through Edge of Hole Found in Specimen 4-1A. As polished. 100X.

Fig. 7.11. Metallographic Section of Specimen 3-8A. Etchant glyceria regia. 100X.

the UO_2 particles and not the burnable poison. The reason for this behavior is not known.

Special Tests

A number of other tests are planned to obtain additional information on the effects of irradiation on these dispersion fuels. Waterlogging tests will be made on a number of specimens to determine whether the

waterlogging susceptibility of the fuel dispersion is significantly affected by high burnup. Specimens containing both hydrothermal and spherical UO_2 fuel particles will be tested. Also, waterlogging tests will be made on the Eu_2O_3 -bearing flux-suppressor regions from specimens 1-8A and 3-8A.

Microhardness measurements will be made on a large number of specimens to determine the effect of burnup upon the hardness of the fuel dispersions. Hardness measurements and bend tests are also planned for the B^{10} -bearing frame material used in specimens 2-14 and 3-14A.

Postirradiation annealing tests were conducted on specimens 2-4A, 4-13A, and 3-5C. No dimensional changes were apparent after heat treatment at 600°F for 24 hr. A few small blisters were noted after subsequently annealing the specimens for 24 hr at 700°F . Annealing at successively higher temperatures resulted in increases in both size and number of blisters. Even after heat treatment at 1000°F for 24 hr, however, no evidence of gross swelling, such as noted in irradiated specimen 3-8A, was found. These specimens are currently being prepared for metallographic examination.

Conclusions

The results of this investigation indicate that dispersion fuels containing 26 wt % UO_2 maintain adequate dimensional stability and structural integrity to burnups in excess of 67% depletion of the U^{235} at temperatures of interest to the SM-2 reactor program. Within this burnup range, there is no significant difference in behavior of specimens fueled with either hydrothermal or spherical UO_2 particles. The type of burnable poison (NbB_2 , ZrB_2 , or B_4C) incorporated in the core matrix also has little or no effect on the irradiation stability of these specimens.

At irradiation temperatures of approximately 1000°F , the 26 wt % UO_2 dispersions appear to be limited to burnups of approximately 74% depletion of the U^{235} . This suggests that these fuels should be stable to at least slightly greater burnups at the lower SM-2 irradiation temperature of 600°F .

Dispersion fuels with higher fuel loadings, that is, 38.4 wt % UO_2 or 34.1 wt % UN, are subject to gross swelling when irradiated at approximately 1000°F to burnups of 60% depletion or greater. The data are insufficient, however, to establish a burnup limitation on these fuels either at 1000 or 600°F. At the higher irradiation temperatures, some reaction occurs between the UN particles and the stainless steel matrix. The significance of this reaction is not known.

The incorporation of a Eu_2O_3 -dispersion flux-suppressor section does not significantly affect the irradiation stability of the adjacent fuel section. The Eu_2O_3 section is both dimensionally and structurally stable under these irradiation conditions, and no reaction occurs between the fuel and suppressor regions.

Postirradiation Examination of SM-1 Fuel Elements

A. E. Richt

The effects of irradiation on the dimensional stability and structural integrity of SM-1 reactor fuel elements are being studied. In this work, fuel elements are being subjected to thorough postirradiation examination after significant exposures in the active lattice of the SM-1 reactor. These elements are part of the first core loading for the reactor and consist of 18 composite fuel plates brazed to grooved side plates. The flat, rectangular fuel plates consist of a 0.020-in.-thick core section containing 24 wt % UO_2 and 0.13 wt % B_4C dispersed in a type 302B stainless steel matrix. The fuel section is clad with 0.005 in. of type 304L stainless steel by roll bonding.

As reported previously,^{3,4} two stationary fuel elements, S-72 and S-79, were examined after 10.5 and 16.4 Mwyr of reactor operation, respectively. The examination of a third stationary element (S-81) is now under way. This element was removed after 19.5 Mwyr of reactor operation. Although the examination of this element is not complete, some preliminary results are available.

⁴Oak Ridge National Laboratory, "APPR Ann. Prog. Rep. Jan. 31, 1960," USAEC Report ORNL-2907, pp. 37-41.

Dimensional measurements on element S-81 revealed that rippling of the outer fuel plates of the fuel assembly is greater than noted in the examination of previous elements. This effect is shown in Fig. 7.12. Maximum ripple amplitudes of 0.023, 0.033, and 0.045 in. have been noted for elements S-72, S-79, and S-81, respectively. Thus, rippling of the outer fuel plates appears to be increasing in magnitude with increasing in-pile exposure.

Fig. 7.12. Fuel Plate Rippling Profiles of Fuel Element S-81 After 19.5 M_wyr of Reactor Operation.

Plate-spacing measurements on element S-81 show excellent correlation between the outer fuel-plate rippling profiles and the plate spacing measurements in the water channels between fuel plates 1 and 2 and 17 and 18. No significant changes in plate spacing were found in the other water channels. This indicates that rippling of the inner fuel plates is probably negligible and that the rippling phenomenon is restricted to the outermost fuel plates of the fuel assembly.

Thickness measurements on the individual fuel plates revealed fuel-plate thickness profiles apparently directly proportional to the axial fuel-plate burnup profile. The maximum fuel-plate thickness recorded was 0.033 in. This represents a 0.003-in., or a 10%, increase in fuel-plate thickness for a nominal 0.030-in.-thick fuel plate. Although complete analytical burnup results are not available, preliminary results indicate that the maximum burnup achieved in this element was 62% depletion of the U^{235} (2.2×10^{21} fissions/cm³).

Preliminary metallographic examinations of sections from element S-81 have revealed no evidence of matrix cracking or other gross deterioration of the UO_2 fuel dispersion. The frequency of clad cracking appears to be significantly greater, however, than noted in the examination of previous elements. Both intergranular and transgranular cracks are present. Occasionally the cracks penetrate to the surface of the fuel section; however, in no instance has crack propagation into the core matrix been found. An attempt to quantitatively evaluate cracking of the cladding of all three fuel elements as a function of position along the length of the element is currently under way. Also an analysis of the fuel-plate scale for chlorides is being made to determine whether the transgranular cladding cracks can be attributed to chloride stress corrosion.

Irradiation Testing and Postirradiation Examination of Stainless Steel-Clad Dispersions of Eu_2O_3

A. E. Richt

During early stages of the development of the Eu_2O_3 -stainless steel dispersion control rods for the SM-1 reactor, a program was initiated to

investigate the effects of irradiation on the stability of this absorber material. Miniature stainless steel-clad specimens containing 20, 30, and 40 wt % Eu_2O_3 cores are being irradiated in the ETR at approximately 150°F to integrated thermal-neutron fluxes equivalent to those expected after 1/2, 1, 2, and 3 fuel-core lives in the SM-1 reactor.⁵ To date, eight specimens have acquired the requested in-pile exposures, as shown in Table 7.6. These specimens have been subjected to thorough postirradiation examination. Four other specimens are still being irradiated.

Examinations of the eight specimens removed from the reactor revealed the absorber samples to be in excellent condition. No evidence of dimensional changes was found, and metallographic examinations showed no evidence of structural changes (Fig. 7.13). Thus, it appears that the Eu_2O_3 dispersions are dimensionally and structurally stable throughout a wide range of in-pile exposures.

⁵C. F. Leitten, Jr., "The Irradiation of Miniature Eu_2O_3 -Bearing Absorber Plates - Irradiation Request ORNL-MTR-39," USAEC Report ORNL CF 59-9-25, Oak Ridge National Laboratory, Sept. 14, 1959.

Table 7.6. Irradiation History of Miniature Eu_2O_3 -Dispersion Absorber Plates

Specimen No.	Core Loading (wt % Eu_2O_3)	Irradiation Dose	
		Thermal-Neutron Dose ^a (neutrons/cm ²)	SM-1 Fuel Core Lives ^b
		$\times 10^{20}$	
39-1	20	7.0	0.5
39-2	20	12.1	0.8
39-5	30	7.6	0.5
39-6	30	14.6	1.0
39-7	30	27.9	1.9
39-9	40	7.2	0.5
39-10	40	14.8	1.0
39-11	40	28.7	2.0

^aUnperturbed integrated thermal-neutron flux based on ETR estimates.

^bBased on a specimen perturbation factor of 0.6 and SM-1 fuel core life equivalent to 8.64×10^{20} thermal neutrons/cm².

Fig. 7.13. Comparison of Microstructures of Unirradiated and Irradiated Dispersion of 30 wt % Eu_2O_3 in Stainless Steel After an Estimated Exposure of 9×10^{20} neutrons/cm². (a) Unirradiated. (b) Irradiated. As polished. 100X.

As reported previously,⁶ absorber specimens containing 30 or 40 wt % Eu_2O_3 dispersions are susceptible to gross swelling when intentionally defective specimens are exposed to water at 570°F and a pressure of 1200 psi. As shown in Table 7.7, corrosion tests on both irradiated and control specimens indicate that swelling is predominantly a function of the Eu_2O_3 loading; irradiation has little or no effect on the swelling characteristics of the specimens. As shown in Fig. 7.14, exposure of these specimens to high-temperature water does not result in significant leaching of the Eu_2O_3 particles. This was confirmed when the europium content

⁶A. E. Richt, Irradiation Testing and Postirradiation Examination of Dispersions of Eu_2O_3 in Stainless Steel, pp. 41-43, "Army Reactors Program Prog. Rep.," USAEC Report ORNL-3231, Oak Ridge National Laboratory, Jan. 31, 1962.

Table 7.7. Corrosion Resistance of Eu_2O_3 -Stainless Steel Dispersions in Water at 570°F and a Pressure of 1200 psi

Specimen No.	Unperturbed Integrated Dose (neutrons/cm ²)	Weight Change (%)	Thickness Change (%)
<u>20 wt % Eu_2O_3</u>			
2	Unirradiated	+0.04	+0.9
39-1	7.0×10^{20}	+0.01	+0.8
39-2	12.1×10^{20}	+0.05	+2.5
<u>30 wt % Eu_2O_3</u>			
3	Unirradiated	+0.01	0
39-5	7.6×10^{20}	+0.42	+13.3
39-6	14.6×10^{20}	+0.62	+15.4
39-7	27.9×10^{20}	+0.71	+11.3
<u>40 wt % Eu_2O_3</u>			
4	Unirradiated	+1.53	+27.3
39-9	7.2×10^{20}	+0.24	+34.0
39-10	14.8×10^{20}	+1.58	+30.5
39-12	28.7×10^{20}	+1.51	+31.3

Fig. 7.14. Exposed Core of an Irradiated 40 wt % Eu_2O_3 Absorber Plate After Exposure to 570°F Water for 88 hr. As polished. 500X.

of the autoclave test water was found to be below spectrographic detection limits. Thus, in the unlikely instance of a cladding failure in an SM-1 control rod, essentially no europium should be lost to the reactor coolant system.

Interim Postirradiation Examination of the SM-2B Fuel Element

A. E. Richt L. D. Schaffer

The SM-2B fuel element was one of two experimental fuel elements prepared for testing in the SM-1 by Alco Products, Inc. It is similar in over-all dimensions to the SM-1 core I fuel assembly. The major difference in the two designs is that the SM-2 plates are joined to the side plates by TIG welding, whereas the SM-1 plates are joined to the side plates by brazing. Other differences are listed in Table 7.8.

The SM-2B assembly was made so that each plate varied from the other 17 plates in composition or manufacturing history or both. The plates

Table 7.8. Comparison of SM-1 Core I and the SM-2 Fuel Assembly Designs

	SM-1 Core I	SM-2
Cladding material	Type 304L stain- less steel	Type 347 stain- less steel
Fuel core thickness, in.	0.020	0.030
Fuel core width, in.	2.500	2.650
Assembly U ²³⁵ content, g	515.16	886.55

are described in Table 7.9. The SM-2B assembly was placed into the core of the SM-1 reactor and operated there during the spiked core I run from October 1, 1960 to April 15, 1961. The reactor energy output in that period was 1.6 Mwyr. At the conclusion of this operational period, the two SM-2 fuel assemblies, SM-2A and SM-2B, were inspected visually with the periscope in the shield tank above the pressure vessel of the SM-1 reactor. During this inspection a vertical mark was noted in one of the central welds of fuel element SM-2B. When the inspection was completed, the SM-2B fuel element was placed back into the reactor with the new core II loading along with the five Eu₂O₃ control rods and two high-burnup fuel elements, S-80 and S-81. The reactor then operated from June 2, 1961 until October 16, 1961, during which time the reactor energy release was 1.5 Mwyr.

On October 26 and 27, 1961 another fuel element inspection was performed at the SM-1. During the inspection of the SM-2B assembly marks were noted in and connecting two welds on one of the side plates in the same general area in which the single mark was noted in the previous inspection. As a result of these observations and because third-core procurement for the Army reactors utilizing the flat-plate fuel elements was soon to be initiated and was to be based on the SM-2 design, ORNL was requested to examine this fuel element and, if possible, to determine the nature of the marks.

Table 7.9. SM-2B Fuel Plate Data^a

Plate No.	Serial No.	Remarks ^b	U ²³⁵ Content (g)	Boron Content (g)	Condition
1	EAR-30	SM-2 spherical UO ₂ , ZrB ₂	49.47	0.518	Flat annealed
2	-68	Boron in picture frame only	49.45	1.003	Flat annealed
3	-70	Green core	49.27	0.516	Flat annealed
4	-73	Geneva oxide fuel	49.43	0.518	Flat annealed
5	-75	Columbium boride poison	49.49	0.518	Flat annealed
6	-79	UN fuel, no poison	51.47	None	Flat annealed
7	-60	Coated ZrB ₂ poison	49.45	0.509	Flat annealed
8	-82	UN fuel, coated ZrB ₂	51.79	0.539	Flat annealed
9	-84	SM-1 core II Geneva oxide, B ₄ C	43.82	0.222	Flat annealed
10	-85	SM-1 core II Geneva oxide, B ₄ C	43.24	0.219	Cold rolled, 20% reduction
11	-86	UN fuel, coated ZrB ₂	51.79	0.539	Cold rolled, 20% reduction
12	-61	Coated ZrB ₂ poison	49.14	0.505	Cold rolled, 20% reduction
13	-80	UN fuel, no poison	51.61	None	Cold rolled, 20% reduction
14	-76	Columbium boride poison	49.55	0.518	Cold rolled, 20% reduction
15	-74	Geneva oxide	49.40	0.518	Cold rolled, 20% reduction
16	-71	Green core	49.29	0.516	Cold rolled, 20% reduction
17	-78	Boron in picture frame only	49.40	1.003	Cold rolled, 20% reduction
18	-48	SM-2 spherical UO ₂ , ZrB ₂	49.49	0.518	Cold rolled, 20% reduction
Total			886.55	8.769	

^aH. R. Clayton, Alco Products, Inc., personal communication, December 18, 1961.

^bUnless otherwise indicated, fuel is spherical UO₂ and poison is zirconium diboride.

Examination of Exterior Surfaces

The exterior surfaces of the element were examined first at low magnification to establish the areas that contained marks which could possibly be cracks. These areas were examined at low power with oblique lighting and then at higher magnification using a low-powered, binocular-type microscope with direct lighting. Next, the areas in question were checked by dye-penetrant testing. It was planned that if the marks or lines noted in these areas could not be clearly classed as either scratches or cracks, local polishing and etching would be attempted for higher magnification study.

Visual Inspection. The low-power visual examination was begun December 27, 1961. The oxide film on the element appeared to be darker and more uniform than that noted previously on core I fuel assemblies. The two outside fuel-plate surfaces did not exhibit any significant marks or scratches. The side plate 180° from the dowel pin was scratched to some extent, but there were no marks that appeared to be cracks. The side plate on the dowel pin side appeared to be scratched to a greater

extent, but the only area observed that was of real concern was the one noted during the fuel component examinations at the SM-1.

In order to facilitate description of the examination, a picture of a welded side plate is shown in Fig. 7.15. Sixteen longitudinal rows of welds are made on each of the side plates to hold the fuel plates in place within the assembly. There are nine welds per transverse row, and a given plate is welded along alternate transverse rows. A sketch identifying the area in question is presented in Fig. 7.16, which gives the weld row numbering scheme.

A series of photographs at a magnification of 1.4 were taken of the side plates in question beginning with the spring end and proceeding to the bottom end adapter. The area in question is shown in Fig. 7.17, and the same area is shown in Fig. 7.18 with modified lighting. In the latter photograph the depression of the welds and the embossed effect of the fuel-plate channels on the side plate are evident. It should be noted that these photographs are inverted as a consequence of the optical system in the camera; they have been reproduced as photographed to prevent loss of detail.

Viewing the area in question at low magnification provided no solution to the problem of whether these surface lines were cracks or scratches. Therefore, the fuel assembly was placed under the objective of a low-power, binocular-type microscope. With this equipment the area in question was located and was visually examined at magnifications from 5X through 15X.

A more detailed sketch of the side plate on the dowel-pin side is shown in Fig. 7.19. The two welds, 9'-9 and 10'-10, and the lines connecting them comprised the area in question. The defect observed is sketched in detail A of Fig. 7.19, and in this detail the areas which were photographed with the stereo camera are shown with their figure numbers (Figs. 7.20 through 7.25) listed. Note that photographs of Figs. 7.20 through 7.25 are inverted. These photographs were left in this position in order to avoid any loss in distinctness that would come with printing away from the emulsion side of the negative.

Figure 7.20 and a higher magnification of the same area, Fig. 7.21, show the suspect mark along the lower weld and toward the upper weld.

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Fig. 7.15. SM-2 Fuel Element Showing Welded Side Plate. (From Alco Products report ALCO-F-31, Oct. 18, 1960, p. 97.)

Fig. 7.16. Sketch Showing the Area in Question and the Weld Location Scheme Used in the Examination of SM-2B Fuel Assembly.

With these higher magnification photographs, this mark is resolved sufficiently so that it can be seen that it is not a crack but is, rather, a series of fine-line scratches.

Figure 7.22 shows the bottom end of the lower weld. In this photograph, the weld crater can be seen, as well as some scratches at the tip of the weld running longitudinally on the embossed surface of the side plate below the weld.

Figure 7.23 shows the mark as it exists between the two welds. Although the photograph does not clearly show the nature of the mark in this area, careful visual examination indicated that this portion of the mark was merely a continuation of the scratches noted on the lower weld.

Figures 7.24 and 7.25 show the upper weld, 10'-10. The mark in this region again appears to be scratches along the edge of the weld depression. At the top end of the weld shown in Fig. 7.25, several heavy scratches can be seen. Thus the visual examinations indicated that there were no cracks in the area involving welds 9'-9 and 10'-10.

Fig. 7.17. Side Plate of the SM-2B Fuel Assembly Showing Weld Rows 9' and 10'. 1.4X.

Fig. 7.18. Same Area as in Fig. 7.17 Showing Weld Rows 9' and 10' Under Oblique Lighting. 1.4X.

Fig. 7.19. Sketches of Area in Question. Detail A shows weld nuggets 10'-10 and 9'-9, the suspect lines, and the areas for further study.

Visual Dye-Penetrant Inspection. In order to confirm the conclusion drawn from the visual examination, a visual dye-penetrant inspection was performed.⁷ The visual dye-penetrant method was used because it is more convenient in a hot cell than the fluorescent-penetrant method and because visibility in the cell is very good.

The fuel assembly was positioned with the dowel-pin side up. The area involving welds 9'-9 and 10'-10 was located with the Kollmorgan visual equipment. Since the fuel element had operated in hot water for some time, no degreasing was performed. The red dye was applied to the general area of welds 9'-9 and 10'-10. In observing the application of the penetrant through the viewing equipment, it was noticed that it was

⁷K. K. Klindt of the ORNL Inspection Engineering Department supervised this operation.

Fig. 7.20. Upper End of Weld 10'-10 on One of the SM-2B Fuel Assembly Side Plates. 5X

Fig. 7.21. Upper End of Weld 10'-10 on One of the SM-2B Fuel Assembly Side Plates, at a Higher Magnification. 15X.

Fig. 7.22. Lower End of Weld 10'-10 on One of the SM-2B Fuel Assembly Side Plates. 5X.

Fig. 7.23. Area Between Welds 9'-9 and 10'-10 on One of the SM-2B Fuel Assembly Side Plates. 5X.

Fig. 7.24. Lower End of Weld 9'-9 on One of the SM-2B Fuel Assembly Side Plates. 5X.

Fig. 7.25. Upper End of Weld 9'-9 on One of the SM-2B Fuel Assembly Side Plates. 5X.

smoking. Apparently the surface of the side plate was sufficiently hot to cause the penetrant fluid to vaporize rapidly enough to be noticeable. Therefore, the red dye was applied as often as necessary to prevent the fluid from drying. A liquid dye film was maintained on the area of study for 20 min. After this period, the excess fluid was wiped off with paper towels dampened with carbon tetrachloride. Next, the developer was applied to the test area; and, because the metal surface was above room temperature, the developer dried rapidly.

The test area was inspected visually through the hot cell optical equipment for about an hour, and no sign of red or pin colorations appeared in the white developer. This observation was repeated 18 hr later, and still no indications of red dye were noted.

The conclusion that normally would be made from this test is that there were no cracks in the test area, and all marks in the area must be shallow imperfections, such as scratches or abrasions. The fact that the metal surface was above normal room temperature cast doubt on the sensitivity of the test, however, and therefore a test was undertaken to determine whether the visual-penetrant method was reliable in this situation.

An aluminum test block having cracks perceptible to the naked eye and some fine-line cracks capable of visual detection at a magnification of 5X was heated with an air heater. The surface temperature of the SM-2B side plates was estimated to be below 212°F, probably about 180 to 190°F.⁸ Therefore, two Tempilstik marks were placed on the aluminum plate; one was an indicator for 175°F and the other was a 275°F indicator. From the temperature indications, the aluminum block was heated to above 175°F but below 275°F. This set-up was maintained while a visual dye-

⁸This approximation was later shown to be relatively close. When the SM-2B assembly was placed back into the SM-1 carrier, its temperature was measured with a thermocouple. The thermocouple was placed in one of the central water channels of the assembly and was moved into the center portion of the element. A spring clip was placed on the thermocouple to keep it against the fuel-plate surface. The assembly was then placed in the carrier and the carrier drawer was shut tight except to leave space for the thermocouple leads entering the carrier cavity. The temperature stabilized within a few hours and varied from 150 to 155°F over a 24-hr period. This measurement was performed on January 5, 1962.

penetrant test of the type made on the SM-2B element was performed on this test block. The test was then repeated on the block at room temperature, and the results were compared. It was found that at room temperature all cracks showed up immediately, and at the higher temperature the larger cracks showed up just as rapidly, but the smaller cracks were not in evidence until several hours later.

On the basis of this qualification test, it appears that, when the material being inspected is above room temperature, the sensitivity of the visual dye-penetrant inspection method is reduced; however, by waiting several hours for the final inspection, the sensitivity was still sufficient to pick up cracks that were not visible to the naked eye but were visible at a magnification of 5X. Therefore, it was concluded that the visual dye-penetrant test as applied to the SM-2B side plates was a useful test. Since the results from this test showed that there were no cracks in the suspect area, this latter inspection can be considered as a substantiation of the visual examination findings.

Conclusions and Recommendations

From the visual examination performed on the SM-2B element and, in particular, from the detailed examination of the area in question on the one side plate, it was concluded that the marks noted in association with weld 9'-9 and 10'-10 are not cracks but are merely a series of fine scratches and that the fuel element is in good condition.

It was recommended that the fuel element be reinserted into the SM-1 for further testing, since no signs of significant radiation, corrosion, or operational damage which would preclude continued use were found. This recommendation was accepted and the SM-2B fuel assembly was reinserted into the SM-1.

Measurements for Later Comparisons

Since it was concluded that no further examination of element SM-2B was required, some measurements of the assembly were made that might provide a useful basis for future comparison. The gamma activity was measured and is compared with the activity of SM-1 fuel element S-79 in Fig. 7.26.

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Fig. 7.26. Biological Dose Rate as a Function of Distance from SM-Type Fuel Elements. Fuel element SM-2B discharged after exposure of approximately 3 Mwyr. Fuel element S-79 discharged on April 28, 1960 after exposure of 16.4 Mwyr.

In addition, the dimensional profile of the assembly and the top and bottom fuel plate rippling were measured. These data are presented in Figs. 7.27, 7.28, and 7.29 and compared with the Alco Products, Inc., preirradiation measurements. These two sets of measurements provide initial and interim operational data with which the final postirradiation measurements can be compared. The three sets of measurements should indicate whether there are progressive rippling or assembly distortion effects on a welded assembly operated at SM-1 reactor conditions.

Fig. 7.27. Center Line Fuel-Plate Rippling Profiles on Fuel Element SM-2B.

Fig. 7.28. Dimensional Profiles Across Fuel Plates of Fuel Element SM-2B.

Fig. 7.29. Dimensional Profiles Across Side Plates of Fuel Element SM-2B.

8. DEVELOPMENT OF EQUIPMENT FOR POSTIRRADIATION EXAMINATION

S. E. Dismuke

In the course of postirradiation examination of various fuel and control materials, special equipment is often needed to better accomplish the work or to improve the quality of the data being collected. The equipment described below was developed recently for measuring fuel-plate spacing and for sampling fission gas.

Equipment for Measuring Plate Spacing

The measurement of plate spacing in the SM-type flat-plate fuel elements is accomplished by utilizing eddy-current techniques. The probe developed for these measurements was based on work done by Dodd and McClung,¹ and details of the fabrication of the probe have been reported.²

Plate-spacing measurements are made on a fuel element with the use of the milling-machine table shown in Fig. 8.1. The probe shown in Fig. 8.2 is inserted by a master-slave manipulator into the channel being measured. The milling-machine table is then positioned so that the probe handle can be clamped to a stationary support from which the table is driven at a constant rate to pull the fuel element from the probe while the spacing is continuously recorded. Equipment improvements have been made since the SM-2B fuel element was measured that include provisions for servo tracking of the milling-machine table and for linearizing the probe output.

Fission-Gas Sampling Apparatus

Fission-gas sampling of the BMI-32 capsules was attempted by using a drill to penetrate the capsule and then letting the gas expand into an

¹C. V. Dodd and R. W. McClung, "Fuel-Element Coolant Channel and Other Spacing Measurements," Trans. Am. Nucl. Soc., 4(1): 136-137 (June 1961).

²S. E. Dismuke, "Directions for the Fabrication of a Probe for the Measuring of Plate Spacing in Plate-Type Fuel Elements," USAEC Report ORNL-TM-158, Oak Ridge National Laboratory, April 11, 1962.

Fig. 8.1. Milling Machine Used for Plate-Spacing Measurements of Irradiated SM-1 Fuel Elements.

evacuated chamber that was sealed off and removed from the cell. A schematic drawing of the system is shown in Fig. 8.3, and the assembled apparatus is shown in Fig. 8.4. The gas was transferred from its collection chamber to sampling bottles for radiochemical analysis.

Many difficulties were encountered with the system, and some of them have been eliminated. It is recommended that future capsules include a

thin-walled section at the upper end for penetration for gas sampling. Such a design should facilitate the fission-gas sampling and avoid crud buildup that would prevent gas removal, as was experienced when the sampling was performed on the bottom of capsules containing failed specimens.

Fig. 8.2. Probe for Measuring Plate Spacing of Flat-Plate Fuel Elements.

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Fig. 8.3. Diagram of Fission-Gas Sampling Apparatus.

Fig. 8.4. Fission-Gas Sampling Apparatus.

9. IMPACT TESTS ON IRRADIATED SM-1 PRESSURE VESSEL STEEL

R. G. Berggren	M. S. Wechsler
L. D. Schaffer	T. N. Jones

Surveillance samples of ASTM A212, grade B, steel from the pressure vessel of the SM-1 reactor at Fort Belvoir, Virginia were placed in the reactor at the time of initial startup in April 1957. Eight capsules were located on the core support plate 15.0 in. from the vertical center-line of the reactor, with the bottom ends of the impact bars 15.0 in. from the core horizontal mid-plane. Each capsule contained three sub-size Izod impact specimens with six notches each. Since there is a steep fast-neutron flux gradient where the samples were placed, each notch was exposed to a different fast-neutron flux. The exposure temperature of the capsules was about that of the exit coolant, that is, 450°F.

Thus far, two capsules have been removed; the first after 16.4 Mwy of operation and the second after 20.8 Mwy of operation. The impact tests were conducted at ORNL in hot cell 4 of Building 3025. Unirradiated specimens from the same steel plate were tested in the hot cell along with the irradiated surveillance specimens. The results of the impact tests are presented in Fig. 9.1. Since only three tests were possible for each notch location and exposure, the impact curves are not very well defined. Therefore, an estimate of the shift in the brittle-to-ductile transition temperature is rather difficult; however, the following procedure was adopted. From the scatter band of the points for the unirradiated samples shown in Fig. 9.1, a transparent overlay was constructed. The overlay was translated along the temperature axis until the three points for a given notch were best fitted inside the scatter band. The amount of translation (in °F) thus determined was taken to be the increase in the transition temperature. The results of this analysis for the two capsules are given in Table 9.1.

It is current practice, particularly with regard to design and operating criteria for reactor pressure vessels, to use the Charpy V-notch impact sample in the determination of transition temperatures. It has been found that the irradiation-induced increase in transition temperature for the Charpy-V notch sample is roughly 25°F higher than for the

Fig. 9.1. Impact Energy Versus Test Temperature for Surveillance Specimens of ASTM A212, Grade B, Steel from SM-1 Pressure Vessel Material in Capsule No. I After 16.4 Mwyr and in Capsule No. II After 20.8 Mwyr of Reactor Operation.

subsize Izod sample.¹ On this basis, the corresponding increases in Charpy V-notch transition temperature are also given in Table 9.1.

A study of the neutron flux distribution has been reported² for a mockup of the SM-1 containing mockups of the thermal shield and pressure

¹R. G. Berggren, p. 91 in "USAEC Conference on the Status of Radiation Effects Research on Structural Materials and the Implications to Reactor Design," USAEC Report TID-7588, October 1960.

²K. C. Sontheimer et al., "Fast Neutron Flux Measurements and Analysis in SM-1 and PM-2A Core and Vessel Mock-ups," USAEC Report APAE-95, Alco Products, Inc., March 31, 1962.

Table 9.1. Increase in Brittle-to-Ductile Transition Temperature for the SM-1 Surveillance Samples

Notch No. ^a	Subsize Izod Transition Temperature Increase (°F)		Charpy-V Transition Temperature Increase (°F) ^b	
	Capsule I, 16.4 Mwyr	Capsule II, 20.8 Mwyr	Capsule I, 16.4 Mwyr	Capsule II, 20.8 Mwyr
1	45	51	70	76
2	58	67	83	92
3	69	76	94	101
4	88	109	113	134
5	120	141	145	166
6	130	150	155	175

^aNotches numbered in order of decreasing height, that is, decreasing distance from horizontal core mid-plane.

^bCalculated according to $\Delta TT \text{ (Charpy-V)} = \Delta TT \text{ (Subsize Izod)} + 25^\circ\text{F}$. (See Ref. 1)

vessel. The neutron flux measured with sulphur detectors was found to be 4.8×10^2 neutrons/cm².sec.watt at the pressure vessel and 13.9×10^2 neutrons/cm².sec.watt at a position corresponding to notch 5 of the surveillance samples. The increase in neutron flux at either of these positions in going from room temperature (the temperature at which the mockup flux measurements were made) to the reactor operating temperature was determined to be a factor of 3.50. Thus, for notch 5 of capsule I (16.4 Mwyr) and capsule II (20.8 Mwyr), the calculated sulphur doses were 2.5×10^{18} and 3.2×10^{18} neutrons/cm², respectively. A formula relating transition temperature increase, ΔTT , and neutron dose has recently been given by Nichols and Harries:³

$$\Delta TT = A [\phi(\text{Ni})]^{1/2}, \quad (1)$$

³R. W. Nichols and D. R. Harries, "Brittle Fracture and Irradiation Effects in Ferritic Pressure Vessel Steels," Symposium on Radiation Effects on Metals and Neutron Dosimetry, ASTM, Los Angeles, Oct. 1-5, 1962.

where

ΔT_T = increase in the transition temperature, °F,

A = constant = 49,

$\phi(\text{Ni})$ = integrated neutron flux obtained with a nickel threshold detector (threshold energy = 2.9 Mev) in units of 10^{18} neutrons/cm².

The sulphur flux should be about the same as the nickel flux, since the threshold energy for sulphur (3.0 Mev) is almost the same as for nickel (2.9 Mev).⁴ Equation 1 was determined for Charpy V-notch samples irradiated at 300°F at much higher fluxes than the SM-1 surveillance samples. By substituting the integrated fluxes into Eq. (1), that is, 2.5×10^{18} and 3.2×10^{18} neutrons/cm² as given above for the surveillance samples, it is found that the predicted ΔT_T values are 78 and 88°F for notch 5 of the capsules I and II, respectively, as compared with the values 145 and 166°F (Table 9.1) obtained from the impact tests on the surveillance samples.

These discrepancies between the observed and predicted values may be due to lack of precision in the impact tests, errors in neutron dosimetry, differences in neutron spectrum, and the higher exposure temperature of the surveillance specimens (450°F) than that of the samples on which the values for Eq. (1) were based (300°F). The exposure temperature may be a significant factor since some steels have shown a maximum between 300 and 500°F in the curve relating embrittlement to irradiation temperature.³ Another factor may be the higher flux, by a factor of about 10, for the accelerated-irradiation samples as compared with that for the surveillance samples. It has been pointed out^{5,6} that, if the changes in properties of pressure vessel steels upon irradiation are associated with radiation-enhanced diffusion, the larger effects are to be expected in certain cases when a given dose is received at a lower flux.

⁴T. O. Passel and R. L. Heath, Nucl. Sci. and Eng., 10: 308 (1961).

⁵M. S. Wechsler and R. G. Berggren, "The Radiation Embrittlement of Reactor Pressure Vessels," Nucl. Safety, 4(1): 42-52 (September 1962).

⁶M. S. Wechsler, "Fundamental Aspects of Radiation Effects on Diffusion-Controlled Reactions in Alloys," Symposium on Radiation Effects on Metals and Neutron Dosimetry, ASTM, Los Angeles, Oct. 1-5, 1962.

Besides the flux information obtained from the mockup experiments, dosimetry results have been obtained by the Naval Research Laboratory from a dosimeter capsule that was placed in the SM-1 in a surveillance capsule position.⁷ These data are presently being analyzed. In addition, further mockup experiments are being planned to determine the neutron flux levels at all the impact bar notch positions and to relate these values to the maximum pressure vessel flux.

Because of the errors inherent in the measurement of the brittle-to-ductile transition temperature of the surveillance samples and in the determination of the neutron flux exposure, it is difficult to assess the present condition of the SM-1 pressure vessel. However, an estimate can be made by assuming that the form of Eq. (1) is valid and evaluating the constant A on the basis of the shifts in transition temperature for notch 5 after 16.4 and 20.8 Mwyr of operation. If $\Delta TT = 145^\circ\text{F}$ and 166°F (Table 9.1) and $\Phi (>3 \text{ Mev}) = 2.5 \times 10^{18}$ and 3.2×10^{18} neutrons/cm² for 16.4 and 20.8 Mwyr of operation, respectively, are substituted into Eq. (1), the constant A is found to be approximately 90 in both cases. The flux measurements on the SM-1 mockup² indicated that the ratio of the flux at the pressure vessel belt-line to that at notch No. 5 was 0.345, from which it is calculated that

$$\Phi (>3 \text{ Mev}) = 1.1 \times 10^{18} \text{ neutrons/cm}^2$$

for the pressure vessel belt-line after 20.8 Mwyr. The substitution of this flux and A = 90 into Eq. (1) gives $\Delta TT = 95^\circ\text{F}$. Since the transition temperature of the SM-1 pressure vessel steel in the unirradiated condition was determined to be about 40°F , the value after 20.8 Mwyr would be set at $40 + 95 = 135^\circ\text{F}$.

An extrapolation can be made to estimate the transition temperature of the steel at the pressure vessel belt-line at the end of 20 years of

⁷L. E. Steele and J. R. Hawthorne, Irradiation Effects on Reactor Structural Materials, p. 8.8 in "Quarterly Progress Report: Irradiation Effects on Reactor Structural Materials," USAEC Report HW-74679, Hanford Atomic Products Operation, Aug. 15, 1962.

operation (that is, by 1977), at which time the reactor will have accumulated approximately 83 Mwyr of operation. In this case,

$$\Phi (>3 \text{ Mev}) = 4.4 \times 10^{18} \text{ neutrons/cm}^2 ,$$

and, when substituted in Eq. (1) with $A = 90$, gives $\Delta TT = 189^\circ\text{F}$. The brittle-ductile transition temperature of the steel in the pressure vessel belt-line is then estimated to be $40 + 189 = 229^\circ\text{F}$.

It should be emphasized that the considerations which led to the above predicted transition temperature after 20 years of operation incorporates many uncertainties and approximations. Aside from lack of precision in the impact and flux measurements, there is the question of the applicability of the form of Eq. (1) to the steel used in the pressure vessel of the SM-1 reactor under the particular flux and temperature conditions at the critical regions of the pressure vessel. Furthermore, the basic issue arises as to the extent to which the results of impact tests on unstressed surveillance samples can be correlated with the susceptibility of the operating pressure vessel to brittle failure. The assessment of the condition of the SM-1 pressure vessel should increase in accuracy, however, as additional surveillance capsules are examined in the course of the operating life of the reactor. Also, it is hoped that the analysis of the results will improve by virtue of increased understanding of the nature of the phenomenon of radiation-embrittlement of pressure vessel steels.

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