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OF THE UNIVERSITY OF CALIFORNIA • LOS ALAMOS NEW MEXICO

U
REACTOR IRRADIATION OF URANIUM-IMPREGNATED
GRAPHITE AT 1500°C TO 10% BURNUP

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LOS ALAMOS SCIENTIFIC LABORATORY
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REACTOR IRRADIATION OF URANIUM-IMPREGNATED
GRAPHITE AT 1500°C TO 10% BURNUP

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ABSTRACT

Two type-AUC graphite fuel elements loaded by solution impregnation to an average concentration of 0.115 g/cc of 93.13% enriched U converted to UC and UC₂ were irradiated at temperatures about 1500°C to a 10.2% maximum burnup, corresponding to an irradiation level of 219 KWH/cc or 2.45×10^{19} fissions/cc of fuel element.

Post-irradiation measurements of the elements showed dimensional changes of -4.3 and -4.8% with the grain, and -0.8 to -2.5% across the grain. Weight losses were 3.2 and 5.1% for the individual elements with approximately 11% of the total U²³⁵ being lost from the elements. With-the-grain thermal conductivity at nominal room temperature was reduced by a factor of ~7 and electrical conductivities by factors of 3.4 to 6.3, also at room temperature.

Impact strength appeared to be somewhat improved by irradiation. Uranium migration within the element was detected by radiographic density observations but not evaluated quantitatively. As anticipated, fission product release was high.

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TABLE OF CONTENTS

	<u>Page</u>
Abstract	3
Acknowledgements	5
1. Introduction	9
2. Design and Description of Experiment	10
2.1 Fuel Elements under Test	10
2.2 Final Irradiation Capsule Design	12
2.3 Three Initial Irradiation Capsule Designs and Their Patterns of Failure	16
2.4 Irradiation History of Capsule No. 4	19
3. Presentation and Discussion of Results	21
3.1 Visual Examination	21
3.2 Dimensional Changes and Weight Losses	24
3.3 Thermal and Electrical Conductivity	31
3.4 Impact Strength	36
3.5 Uranium and Fission Product Migration	36
4. Conclusions	40
References	41

LIST OF TABLES

1. Radiation Exposure of Elements as Determined by Ce ¹⁴⁴ Analyses	21
2. Dimensional, Weight, and Density Changes of Irradiated Elements	25
3. Measured Thermal Conductivity of Various Materials at Room Temperature Compared with Handbook Values	32
4. Average of Electrical Conductivity Measurements at Room Temperature of Various Type AUC Graphite Materials	34

LIST OF TABLES (continued)

	<u>Page</u>
5. Retention of Sr^{90} , Y^{91} , $\text{Cd}^{115\text{m}}$ and Cs^{137} within the Elements	37
6. U^{235} Concentration in Elements and Percent Burnup	39

LIST OF FIGURES

1. Unirradiated UHTREX Element	11
2. Radiographs of UHTREX Elements QR-45 and QR-42 before Irradiation	13
3. Final Design of Irradiation Capsule No. 4	14
4. UHTREX Element QR-42 after Irradiation	22
5. UHTREX Element QR-45 after Irradiation	23
6. Radiographs of Cross-Sections of Typical UHTREX Elements Showing U Distribution	28
7. Density vs. Sample Position after Irradiation of UHTREX Elements	29
8. Electrical Conductivity vs. Sample Position after Irradiation of UHTREX Elements	35
9. Radiographs of QR-42 and QR-45 Taken after Irradiation	38

1. INTRODUCTION

In the original proposal for the Turret Reactor¹ (since renamed Ultra High Temperature Reactor Experiment, or UHTREX) a minimum burnup of 10% was anticipated for fuel elements consisting of U in graphite at a fuel temperature of about 1650°C. Loading of the U was to be done by solution impregnation, and substantial release of fission products was expected. At the time this proposal was to be implemented, no experimental data were available on radiation damage at this temperature and irradiation level in such fuel elements. Thus, a capsule irradiation of fuel elements typical of the proposal was initiated to obtain experimental data for this set of conditions.

A capsule was designed for use in the core of the Omega West Reactor (OWR) in which the operating temperature was predetermined by calculation of fission heating rates and introduction of heat transfer barriers in the form of He-filled gaps between the elements and the capsule walls. One initial calibration experiment and two subsequent irradiations were made, all being withdrawn due to thermocouple or thermocouple well failures, before a capsule design was derived that permitted a full term irradiation of 1300 hr.

At the conclusion of the irradiation, and after several weeks cooling in an essentially zero flux position in the reactor, the capsule was withdrawn and removed to the DP-West Plutonium Hot Cells for examination.

Comparative tests were run on the fuel elements to detect changes in dimensions, weight, impact strength, and electrical and thermal conductivity. Other post-irradiation work at other facilities included U^{235} burnup determinations, radiographic examination, and gamma scanning of the elements.

2. DESIGN AND DESCRIPTION OF EXPERIMENT

2.1 Fuel Elements under Test

An unirradiated example of the element tested is shown in Fig. 1. The dimensions were 5.875 in. long by 1.000 in. diameter with a 0.500 in. diameter hole running axially through the center. The edges were rounded to a $1/8$ in. radius. Identification numbers were engraved in the ends and sides. Fabrication was accomplished by machining the element to size from type AUC extruded graphite rod stock supplied in 1-1/2 in. diameter.

The loading of the element with 93.13% enriched U was done by evacuating a vessel holding the element and then introducing a solution of $2M\ UO_2(NO_3)_2$ which was absorbed into the graphite. The element was then heated to $300^\circ C$ in air to convert the $UO_2(NO_3)_2$ to UO_x and was held at temperature until a constant weight was achieved. The element was

Fig. 1. Unirradiated UHTREX Element

then heated to 2200°C in a He atmosphere and held for about 1 hr to convert the UO_x to UC and UC_2 .² The purpose of the conversion to the carbide was to forestall any such reaction taking place during the irradiation when the product gases generated in the sealed capsule could have a deleterious effect on the experiment. X-ray analysis of samples taken from the high-fired elements showed less than 5% UO_2 remaining. The elements used in the irradiation were numbers QR-42 and QR-45, which were loaded to an average of 0.107 g U^{235} /cc of graphite matrix, or a total of 12.96 g of U^{235} for the two elements. Figure 2 shows a radiograph of the loaded elements and illustrates the distribution of the U along their length.

2.2 Final Irradiation Capsule Design

Using standard calculational methods for heat and hydraulic flow, well discussed in other reports^{3,4,5} and texts,^{6,7} a capsule was designed to fit a 2-in. diameter hole in the core of the OWR and is shown in Fig. 3. This represents the final design and incorporates changes found necessary as a result of failures in the first three capsules, which will be discussed subsequently. The design of Capsule No. 4 features two UHTREX type elements in a stainless steel tube with a uniform gap at room temperature of 0.037 in. between tube and element which is maintained by Ta spacers. Unloaded graphite holders to keep the element from contacting the Ta spacers were also used to prevent possible extraneous reactions between the Ta and the fuel elements. Insulators of CaO were placed at both ends of the fuel element-spacer assembly to minimize axial heat flow and keep capsule ends cooler.

QR 45

QR 42

Fig. 2. Radiographs of UHTREX Elements QR-45 and QR-42 before Irradiation

Fig. 3. Final Design of Irradiation Capsule No. 4

Thermocouple wells of Ta containing W-W 26% Re thermocouples with high purity Al_2O_3 insulators were used to measure temperatures at the midpoints of the two elements. The wells were affixed to the capsule top by vacuum brazing in some cases and welding in others, both giving satisfactory results.

To protect against capsule failure and release of fission products to the top of the reactor through the stainless steel tube housing the thermocouple extension lead wires, a second sealed volume was incorporated above the capsule proper. This seal was made by using ceramic-to-metal insulating seals which passed the extension lead wires and was located outside the core in a low flux-low temperature region. Both the capsule and the secondary protective volume were back filled with He to atmospheric pressure on assembly. Final sealing of both volumes was accomplished by pinching and melting through the He fill lines of 3/16 in. stainless steel tubing. The entire assembly, including the extension lead wire housing, was checked for integrity with a mass spectrometer He leak detector to ensure that no leaks greater than 10^{-9} std. cc/sec of He existed.

Also included in the housing tube for the extension lead wire was a 1/4 in. diameter tube which brought a He purge down to the top of the ceramic-to-metal insulating seals. Thus, if fission products were released through this barrier they would be carried to the top of the reactor by the purge and detected by monitoring instruments. The top of the housing tube was fitted with a phenolic plug to pass the wire and

tubing and sealed with an epoxy resin. To eliminate any possibility of γ beams from the core to the reactor top, the housing tube was packed with small diameter steel rods for a distance of about 3 ft.

2.3 Three Initial Irradiation Capsule Designs and Their Patterns of Failure

To accurately evaluate the reactor flux and reach the desired temperature of operation by properly sizing the gas gap, the initial capsule was conceived as a calibration experiment.

In this experiment, the capsule design was essentially the same as that shown in Fig. 3 except that CaO spacers were used instead of Ta, and the lead wire housing tube did not have a secondary seal. An estimate was made of the flux in position 4-D of the OWR and a gas gap calculated. Capsule No. 1 was inserted in the reactor and the reactor was brought up in power until the temperature of the capsule reached 1580°C. Failure occurred after 5 min at this temperature, but enough information was gained to evaluate the flux and calculate a proper gas gap that would achieve a temperature of 1550°C at a reactor power of 5.0 MW. A post-irradiation examination of the capsule showed failure occurred when the Pt-Pt 10% Rh thermocouple parted at the hot junction, apparently soldering itself to the bottom of the Ta thermocouple well which it was touching. The bottom also dropped out of the Ta tube, where it was plugged and welded, permitting fission products to be picked up in the He purge and brought to the top of the reactor. Radiation levels there reached a maximum of 13 r/hr at contact with the tube in about 3 hr and then fell off gradually.

The most likely explanation of this failure seems to be that the insulation used on the thermocouple contained impurities (possibly Si) which reacted with the Pt-Pt 10% Rh couple and the Ta at these temperatures.

For Capsule No. 2, the gas gap was set at 0.032 in., based on information gained in the calibration run, and the Ta spacers shown in Fig. 3 were substituted for the CaO spacers. The reason for this change was the fracture of the CaO under the thermal shocks sustained in the capsule. Tantalum-sheathed Pt-Pt 10% Rh thermocouple assemblies of 0.060 in. diameter with MgO insulation were used to measure the temperature in this capsule. The couples were not grounded to the sheathing.

A temperature of 1550°C at 5.0 MW of reactor power was attained at the start of the second irradiation, but this gradually declined to a steady state of 1070°C after ~75 hr of operation. At this time the reactor was shut down, in the course of its normal schedule, and the capsule temperature dropped from 1070°C to 200°C in ~9 min. At this point both thermocouples failed, developing an open circuit with the Pt legs grounded, but there was no release of fission product to the lead wire housing tube. The best explanation for this occurrence seems to be that the differing thermal conductivities and coefficients of thermal expansion for the Ta, MgO, and Pt resulted in excessive stresses in the Pt. No further information was revealed in the hot cell examination of this capsule.

In Capsule No. 3 the gas gap was increased to 0.037 in. The intent

was to operate the reactor at reduced power until a temperature decline similar to that experienced in the prior capsule would permit operation at 5.0 MW without increasing the capsule temperature beyond 1550°C. It was believed that the temperature decline was a result of a change in emissivity of the capsule walls due to condensation of volatile fission products or deposition of carbonaceous material from the element as seen in the autopsies, and/or a change in emissivity of the fuel elements. The increased gas gap would dictate a higher steady-state temperature than 1070°C when the emissivities were stabilized, putting the temperature region up into an area of greater interest. Another change was to oxidize the interior surface of the capsule by heating it to 550°C in air for 65 hr, thus increasing the emissivity prior to irradiation.

A thermocouple assembly such as was used in Capsule No. 1 and is shown in Fig. 3 was incorporated in the belief that it was more stable to thermal stresses and shock than the Ta sheathed assemblies. A high purity Al_2O_3 insulator was used to avoid Si contamination.

This capsule attained a temperature of 1450°C at 4.0 MW of reactor power, after which the reactor power was gradually increased over a 24 hr period until 5.0 MW was reached and the temperature increased to 1550°C. The capsule operated for a period of 75 hr, until fission product activity was discovered in the lead wire housing tube, and was then withdrawn. Both Pt-Pt 10% Rh couples were still operative and showed a temperature of 1530°C at time of failure. During the irradiation, the capsule survived one reactor scram in which the capsule temperature dropped from 1550°C to 850°C in less than 1 min.

Post-irradiation hot cell examination revealed that the fission product leakage had originated from a crack in a Ta thermocouple well. This crack was located about 1/4 in. above the top element in a relatively cooler area where a considerable amount of dark, sooty-appearing material was deposited. Possible explanations for the crack appearing are thermal stressing, fission product corrosion, or oxidation of the exterior by air that may have entered into the He purge lines.

2.4 Irradiation History of Capsule No. 4

Capsule No. 4 was inserted in position 4-D in the core of the OWR on Dec. 11, 1961. The reactor was gradually brought to a power of 4.5 MW, at which time the capsule temperature was 1540°C by the bottom thermocouple and 1300°C by the top. Within the next 24 hr reactor power was gradually increased to its normal level of 5.0 MW and the maximum capsule temperature declined to 1510°C. The difference between thermocouples at this time was 60°C. By Dec. 28 the couples agreed within less than 20°C and so continued for the majority of the irradiation with only occasional divergences as great as those experienced in the first 24 hr.

From the start of the irradiation until withdrawal of the capsule on Mar. 17, 1962, the reactor operated at full power for approximately 1300 hr with an integrated power of 6,578 MWH. During this time the capsule survived 5 scrams from full power in which the temperature dropped from ~1500°C to ~800°C in less than 1 min, and from ~800°C to 50°C in the next 5 min.

The range of the maximum recorded temperature during the irradiation

was 1450 to 1650°C, and at least 90% of the time this range was between 1500 to 1600°C.

With regard to the temperature profile along the axis of the capsule, little can be said quantitatively. An error in the original thermal conductivity values used for the CaO insulators resulted in a larger end loss of heat than was originally anticipated. This is particularly true of the bottom end of QR-45 where the CaO insulator was in contact with the water-cooled end plug of the capsule. This was the path of lowest thermal resistance in the capsule and would result in the lower end of QR-45 being the point of lowest temperature in the elements. Reinforcing the effect of these end losses is the fact that the neutron flux falls off about 20-30% from its peak value at the center of the element assembly to the ends. Another point of relative temperature depression to be anticipated is at the midpoint of the element assembly where the elements are joined by two unloaded graphite holders and one Ta spacer. Fission heat is not produced in these components but is removed through them to the capsule wall by radiation and conduction.

Radiochemical analyses on samples from the ends and center of each element were performed by Bryant and Sattizahn,⁸ and the Y^{91} results confirm this qualitative picture of the axial temperature profile. The Sr^{91} precursor of Y^{91} has a 9.7 hr half-life and is mobile in graphite at these temperatures while Y is relatively immobile. The mobility is a function of the temperature, so the larger the removal of Sr^{91} the

higher the temperature and the less Y^{91} present. The Y^{91} analyses show the bottom end of QR-45 to have been the coolest point and the centers of the elements to have been the hotter points of those analyzed.

Table 1 shows the various measures of element exposure to neutron and fission fragment bombardment.

Table 1

RADIATION EXPOSURE OF ELEMENTS AS DETERMINED
BY Ce^{144} ANALYSES

	<u>U^{235} Burnup by Fission and Capture, %</u>	<u>KWH/cc of Matrix</u>	<u>Fissions/cc of Matrix</u>	<u>KW Generated per Element</u>
Maximum	10.2	219	2.45×10^{19}	-
Average ^(a)	9.2	194	2.17×10^{19}	8.45

(a) Integrated value of three samples from each element.

3. PRESENTATION AND DISCUSSION OF RESULTS

3.1 Visual Examination

Disassembly of the capsule was accomplished by cutting the top off with an abrasive cut-off wheel and sliding the contents out. The interior surface of the capsule opposite the elements was blackened by deposition of fission products and/or graphite dust. A large amount of loose black and silver powder was also removed with the elements. This could have come from the cut-off wheel and the capsule and/or from the interior graphite and Ta components.

The elements themselves (Figs. 4 and 5) were partially covered by a loosely adherent black powder which was easily dusted off with a soft

Fig. 4. UHTREX Element QR-42 after Irradiation

Fig. 5. UHTREX Element QR-45 after Irradiation

paint brush. The surface texture had changed, with respect to the non-loaded graphite components, becoming dull and coarse in appearance. This change was marked at the ends of each element and small at the center. The numbers engraved on the ends and sides were easily visible except at the two extreme ends where axial heat losses had depressed the element temperature. Here there had been visible removal of material resulting in a greater rounding of the element corners and in numbers illegible to the naked eye. Later telescopic examination showed the remnants of the numbers on the cooler end of QR-42 but none on the corresponding end of QR-45. A crack extending through the element wall and $1/8$ in. along the wall in an axial direction from the cooler end of QR-42 was the only defect of this type observed. Exposure of the elements to air and general handling during the succeeding three weeks resulted in a noticeable lessening of the distinguishability of the numbers and in a homogeneous element appearance.

3.2 Dimensional Changes and Weight Losses

A summary of dimensional, weight, and density changes is given in Table 2. The weights were taken on an analytical balance after the previously mentioned loosely adherent powder was lightly brushed from the elements. Dimensions were then taken with a micrometer or vernier caliper and densities calculated from these weights and dimensions. It is of interest to note that in the three weeks subsequent to the taking of the post-irradiation weight tabulated, an additional weight loss of 0.51 g and 0.23 g was measured for QR-42 and QR-45, respectively. This was probably a mechanical loss from handling the elements.

Table 2

DIMENSIONAL, WEIGHT, AND DENSITY CHANGES OF IRRADIATED ELEMENTS

	<u>Length, in.</u>		<u>Outside Diameter, in.</u>		<u>Inside Diameter, in.</u>		<u>Weight, g</u>		<u>Average Density, g/cc</u>	
	<u>QR-42</u>	<u>QR-45</u>	<u>QR-42</u>	<u>QR-45</u>	<u>QR-42</u>	<u>QR-45</u>	<u>QR-42</u>	<u>QR-45</u>	<u>QR-42</u>	<u>QR-45</u>
Pre- Irradiation	5.875	5.875	1.001-0.999	1.001-0.999	0.501-0.499	0.501-0.499	97.91	98.17	1.735	1.740
Post- Irradiation	5.625	5.591	0.985-0.975	0.989-0.975	0.496-0.490	0.495-0.487	94.79	93.15	1.83	1.80
Change, %	-4.3	-4.8	-1.5 to -2.5	-1.1 to -2.3	-0.8 to -2.0	-1.0 to -2.4	-3.2	-5.1	+5.2	+3.4

It is apparent from an examination of the length and diameter changes that the average density increase should be greater than 7 or 8% if the dimensional changes are due to shrinkage from irradiation. Since the average density increase is only 3.4 to 5.2%, the conclusion is that part of the dimensional changes were caused by a release of surface material in one or both dimensions. However, since the numbers on the ends of QR-42 were discernible the amount of material released from the axial surface is limited to less than 0.020-0.040 in. This means that axial (with-the-grain) shrinkage is a minimum of 3.6% and cannot account for the difference between the measured average density and that calculated from dimensional changes. It must, therefore, be that the observed decrease in diameters is partially release of surface material and partially shrinkage. Calculations show that in the case of QR-42 an average diametral (across-the-grain) shrinkage of 0.5 to 1.0% would account for the observed average density increase. This then says that an average of 1.0 to 1.5% of the diameter was lost by physical removal of the material. This is equivalent to 2.0 to 3.0% of the volume and could reasonably account for the larger part of the observed loss of weight of 3.2%. No other explanation was been postulated that could account for a weight loss of this magnitude. Even the visible loss of material at the cooler ends of the elements, generously estimated from the pictures as being no greater than 1%, do not significantly change these considerations. The weight change, therefore, is not merely an end effect but an action that is occurring more or less uniformly along the length of the element.

Using as a model the surface evaporation of carbon atoms caused by deposition of the energy of a fission fragment along its path, Bryant and Sattizahn⁸ predict a loss of 2.8 g from this process, which is in reasonable agreement with the facts considering the uncertainties of such a calculation.

A sample of the dust brushed off the exterior of the element prior to weighing was analyzed for U^{235} and found to contain about 2.8 times the average value of U^{235}/g of sample found in the coherent elements after correction for burnup. This skin concentration effect is generally consistent with the U distribution seen in Fig. 6 showing the radiographs of cross-sections of elements typical of those irradiated, and undoubtedly aggravates the weight loss. It also creates the problem that, while the element weight losses are 3.2 and 5.1%, the percent U^{235} removed from the elements would be 2.8 times as great. The analyses for U^{235} in the capsule show that of the U^{235} originally present, 1.32 g (11%) were represented by dust and 10.86 g were represented by the coherent elements. These analyses give an excellent material balance, agreeing within < 1% with the value of 12.10 g of U^{235} originally present.

At a later stage in the examination of the elements they were cut perpendicular to the axis into $\sim 1/2$ in. thick segments. These segments were weighed and measured and the density calculated. A plot of density as a function of position in the element is shown in Fig. 7. The increase in density from the cooler end toward the hotter end can be attributed to two things. First, the neutron flux (and fission density)

Fig. 6. Radiographs of Cross-Sections of Typical UHTREX Elements
Showing U Distribution

Fig. 7. Density vs. Sample Position after Irradiation of UHTREX Elements

increases in the direction of the hotter ends, and shrinkage is proportional to integrated exposure. Second, the contraction rates for graphite under irradiation are dependent upon and increase with increasing irradiation temperature at these higher temperatures. The preceding two statements have been shown to be probably true for neutron bombardment,^{9,10} and the assumption here is that neutron and fission fragment bombardment act in similar ways to cause shrinkage, but not with the same effectiveness. Comparison of the shrinkage experienced here with that seen in neutron irradiation of unloaded graphite at similar neutron exposures and irradiation temperatures indicates that the integrated neutron flux alone can account for only a small fraction of the observed change.^{10,11} In solution-impregnated graphite almost all fission fragments are attenuated in the graphite matrix because of the small UC and UC₂ particle size, and it is to this fission fragment bombardment that the magnitude of shrinkage is attributed.

Another measurement made was to check the amount of bowing in the elements by inserting them into tubes 6 in. long with known inside diameters. Element QR-42 would slip through such a tube with inside diameters of 1.002, 1.002 and 1.001 at the end, middle and end, respectively. QR-45 would only slip into this tube a distance of 5 in. no matter which end was inserted into either end of the tube. However, QR-45 would easily slip through a tube of approximately 0.010 in. larger inside diameter. This indicates that some bowing took place but it was not great.

3.3 Thermal and Electrical Conductivity

Thermal conductivity measurements of UHTREX elements were made by an apparatus employing a liquid metal heat source at one end of the element and a fixed temperature cold sink at the other. The conductivity relative to that of Al (61-S) was calculated from the amount of energy necessary to maintain the liquid metal at a fixed temperature. Table 3 is a summary of the measurements made compared to handbook values, where available.

While the table shows differences of up to 17.7% between measured and handbook values for the materials listed, the error in the range of the 1300 hr irradiated graphite may be significantly greater. The value of these measurements is to show that a large relative change in thermal conductivity has taken place. Another reason for caution in applying these numbers to other elements is the great variance of properties from piece to piece for a given type graphite. This is illustrated in the values of thermal conductivity obtained for the stock from which QR-42 and QR-45 were fabricated. The numbers were obtained by the measurement of electrical conductivity and relating this to thermal conductivity by the equation

$$K \text{ (cal-cm/sec-cm}^2\text{-}^\circ\text{K)} = 0.00031 \sigma \text{ (mho/cm)}$$

which applies for the room temperature properties of graphite.¹² These measurements showed with-the-grain thermal conductivity to be 85 and 69 Btu/hr-ft²-°F/ft for unirradiated QR-42 and QR-45 graphite stock, respectively, compared with a handbook value of 95 Btu/hr-ft²-°F/ft.¹² Other samples of AUC stock showed variations as great.

Table 3

MEASURED THERMAL CONDUCTIVITY OF VARIOUS MATERIALS AT ROOM TEMPERATURE
COMPARED WITH HANDBOOK VALUES

(Btu/hr-ft²-°F/ft)

<u>Material</u>	Brass ^(a) (65% Cu-35% Zn)	Carbon Steel ^(b) (0.1 to 0.4% C)	Al ^(a) (61-S)	U ²³⁸ AUC ^(c) Loaded Graphite	U ²³⁵ Loaded ^(c) AUC Graphite	
					75 hr ^(d) irradiation	1300 hr irradiation
Measured	56	31	99	99	36	14
Handbook	68	25 - 35	99	--	--	--

(a) T. Lyman, Editor, Metals Handbook, 1948 Edition, American Society for Metals, Cleveland, Ohio.

(b) R. T. Powell and W. A. Blanpied, "Thermal Conductivity of Metals and Alloys at Low Temperatures," NBS Circular 556, Sept. 1, 1954.

(c) Graphite values and measurements are parallel to grain of extruded material.

(d) 75 hr irradiation elements were taken from Capsule No. 2. Irradiation condition same as Capsule No. 4 (1300 hr).

Application of the equation to irradiated graphite is not possible since the irradiation has a greater effect on the thermal conductivity than on electrical conductivity.^{13,14}

Table 4 summarizes some of the electrical conductivity measurements made on various AUC graphites.

The irradiated element holder called out in the table was a graphite spacer used in the capsule but not loaded with U. For this reason it sustained no significant fission fragment bombardment, and radiation damage is primarily from neutrons. The against-the-grain electrical conductivity of this piece is noticeably greater than for the irradiated test elements and illustrates the greater effect of fission fragment bombardment compared to neutrons.

Figure 8 shows a plot of the electrical conductivity with the grain and against the grain as a function of distance from the cooler end of QR-42. Although there is a great deal of scatter in the data, it can be concluded that the with-the-grain conductivity increased with distance from the cooler end, while against-the-grain conductivity showed little change. Since the loss of conductivity is attributed to irradiation damage and the irradiation exposure increases as one moves away from the cooler end, it is concluded that the increase in with-the-grain electrical conductivity is probably related to the increasing temperature. Why it apparently does not also improve the across-the-grain conductivity to some degree is not understood.

Table 4

AVERAGE OF ELECTRICAL CONDUCTIVITY MEASUREMENTS AT ROOM TEMPERATURE
OF VARIOUS TYPE AUC GRAPHITE MATERIALS (mho/cm $\times 10^{-3}$)

	QR-42 <u>Stock</u>	QR-45 <u>Stock</u>	U ²³⁸ <u>Loaded</u>	<u>Unloaded</u>	QR-42 <u>Irradiated</u>	QR-45 <u>Irradiated</u>	Handbook ⁽¹²⁾ <u>Value</u>	Irrad. Element <u>Holder</u>
With-the-Grain	1.13	0.922	0.755	0.886	0.238	0.238	1.27	--
Against-the-Grain	0.685	0.621	0.558	0.638	0.145	0.180	0.814	0.290

Fig. 8. Electrical Conductivity vs. Sample Position after Irradiation of UHTREX Elements

3.4 Impact Strength

The impact strength of the irradiated elements was crudely compared to that of unloaded and unirradiated type AUC graphite. Cross sections 1/2 in. thick were cut from the irradiated QR-42 and QR-45 elements and placed in a vise in a reproducible manner, and a 446 g weight was dropped on them in a direction parallel to the axis of the elements so that shear failure across a diameter occurred. Of the 15 similar segments of unloaded AUC tested, all failed on the first impact in which the weight was dropped from heights of 4.5 to 10 in. The strongest test piece of irradiated material took up to four blows from a 10 in. drop height without failure, while the weakest failed under one blow from a 10 in. drop height. Qualitatively the impact strength of the irradiated material appeared to be greater for those samples in the higher temperature zones.

The conclusion is that the irradiated test material is somewhat more resistant to failure from this type of impact than the unirradiated and unloaded type AUC graphite.

3.5 Uranium and Fission Product Migration

Loss of fission products from the elements to the capsule walls can be shown by the analyses for Sr^{90} , Y^{91} , $\text{Cd}^{115\text{m}}$ and Cs^{137} done on samples from the ends and center of both elements and on the HCl solution with which the capsule walls were washed. Results of these analyses are shown in Table 5. More than 90% of some of these nuclides were removed from some of the samples and deposited on the relatively cold capsule

wall. It is interesting to note that the fraction of generated Cs^{137} retained in the elements is greater at the hotter centers (14-23%) than at the relatively cooler ends (4-6%). No explanation for this is offered. Analyses of the capsule contents also showed that Ce^{144} was almost completely retained in the graphite as was expected. Uranium removal from the elements was limited to the sloughing off of dust.

Table 5

RETENTION OF Sr^{90} , Y^{91} , $\text{Cd}^{115\text{m}}$ AND Cs^{137}
WITHIN THE ELEMENTS

Sample No.	Distance from Coolest End of Element, in.	Fraction Retained			
		Sr^{90}	Y^{91}	$\text{Cd}^{115\text{m}}$	Cs^{137}
QR-42-3	0.32	0.054	0.87	0.0106	0.064
QR-42-10	3.24	0.066	0.65	0.0109	0.140
QR-42-17	5.44	0.151	0.90	0.0023	0.057
QR-45-2	0.18	0.45	0.99	0.026	0.040
QR-45-10	2.94	0.032	0.55	0.016	0.23
QR-45-17	5.41	0.041	0.87	0.0046	0.058

Migration of materials within the elements is difficult to evaluate. Radiographs taken after irradiation (Fig. 9) show that there is a higher concentration of material at the ends compared with the centers of each element. The radiographs in Fig. 2, taken before irradiation, show a fairly uniform density. To account for such a change in radiographic density, U movement must be assumed, since fission products are not present in sufficient quantity to cause such an effect by themselves.

Fig. 9. Radiographs of QR-42 and QR-45 Taken after Irradiation

Densification of the graphite to the extent of a few percent cannot be the cause of it either. Analyses of samples for U^{235} , corrected for burnup, of the ends and center of the elements, shown in Table 6, have a maximum variation of 10%. These analyses are difficult to interpret because of the nonuniformity of the graphite.

Table 6

U^{235} CONCENTRATION IN ELEMENTS AND PERCENT BURNUP

Sample No.	Distance from Coolest End of Element, in.	(a) $g\ U^{235} / g\ of\ Sample$	(b) % Burnup of U^{235} Atoms
QR-42-3	0.32	0.0573	8.2
QR-42-10	3.24	0.0573	9.8
QR-42-17	5.44	0.0595	10.2
QR-45-2	0.18	> 0.0514	6.8
QR-45-10	2.94	0.0546	9.5
QR-45-17	5.41	0.0601	9.9

(a) U^{235} values corrected for burnup.

(b) Burnup includes fission plus capture with a capture-to-fission ratio of 0.20 and was determined by Ce^{144} analyses.

Gamma scanning of the irradiated elements in the energy range of 0.18 to 1.8 MEV after a cooling time of about 4 months showed the total activity decreased 11% from the hotter to the cooler end of QR-42 and 58% for QR-45. The large change in QR-45 must be discounted because of local loss of material from the cold end. Burnup determined by Ce^{144} analyses of the elements, also shown in Table 6, decreased from 10.2% to

8.2% burnup from the hotter to cooler end of QR-42, and for QR-45 the decrease was 9.9% to 6.8%. This is a decrease of 19.5% for QR-42 and 31.3% for QR-45. If both the U^{235} and the fission products remained in position in QR-42, there should be no difference between these percentage changes since they would both be proportional to the neutron flux profile assumed to be centered on the assembly. Migration of U^{235} and/or fission products from or along the element would change the ratio. It is known that the more volatile fission products were removed from the hotter spots, and this would account in part for the smaller percentage change in the gamma scanning of QR-42. Movement of fission products and/or U^{235} from the hotter center to the cooler ends could also tend to minimize the difference. The only conclusion to be drawn from these considerations is that the neutron flux variation from peak to minimum was probably greater than the 11% change in the gamma scan and more closely resembles the burnup profile.

4. CONCLUSIONS

From the results of this irradiation experiment, it would appear that the solution-impregnated elements tested suffered from two disadvantages compared with similar tests on other elements loaded by admixing with large diameter (> 100 microns) UC and UC_2 particles.^{15,16,17} First, the small particle size of the UC and UC_2 resulted in almost all the fission fragments being attenuated by the graphite matrix with increased radiation damage to the graphite. Second, the method of loading

causes a higher concentration of U at the element surfaces, which results in the rate of the release of material off of the surface of the element by fission fragment recoil being larger. With this, of course, the rate of the accompanying loss of U^{235} increases still more because of its higher concentration.

REFERENCES

1. L. E. Baker, H. M. Busey, B. M. Carmichael, A. J. Clark, Jr., J. P. Cody, H. J. Gay, R. P. Hammond, A. A. Heckes, D. N. Munro, R. E. Peterson, W. H. Schmidt, L. F. Stravasnik, W. R. Wykoff, "A Preliminary Study of the Turret Experiment," LA-2303, April 17, 1959.
2. J. A. Kircher, private communication, Los Alamos Scientific Laboratory (1961).
3. S. J. Basham, J. H. Stang, W. H. Goldthwaite and B. W. Dunnington, "Description of A Capsule for Irradiation of Fuel Specimens at High Temperatures," BML-1336, Apr. 22, 1959.
4. R. B. VanSice, "Hydraulic Flow Calculation for MTR-ETR Experiments-Standard Practices Manual," IDO-16368, Nov. 1, 1957.
5. R. J. Nertney, "Calculated Surface Temperatures for Nuclear Systems and Analysis of Their Uncertainties," IDO-16343, June 1, 1957.
6. A. I. Brown and S. M. Marco, Introduction to Heat Transfer, 3rd Edition, McGraw-Hill Book Co., Inc., New York (1958).
7. J. H. Perry, Editor, Chemical Engineering Handbook, 3rd Edition, McGraw-Hill Book Co., Inc., New York (1950).
8. E. A. Bryant and J. E. Sattizahn, "Results of Analysis of UHTREX Hi-burn Fuel Samples," Memo to G. A. Cowan, Group J-11, LASL, Oct. 12, 1962.
9. J. M. Davidson, E. M. Woodruff and H. H. Yoshikawa, "High Temperature Radiation Induced Contraction in Graphite," Proceedings of the Fourth Carbon Conference, Buffalo; 599-609, 1960.
10. J. M. Davidson and J. W. Helm, "The H-1 High Temperature Graphite Irradiation Experiment," HW-64286, Apr., 1961.

REFERENCES (continued)

11. J. M. Davidson and J. W. Helm, "The H-3 Irradiation Experiment: Irradiation of EGCR Graphite Interim Report No. 1," HW-71500A, Oct. 16, 1961.
12. "The Industrial Graphite Engineering Handbook," National Carbon Co., 1962.
13. W. N. Reynolds and J. N. W. Simmons, "The Effect of High Flux Neutron Irradiation on the Physical Properties of Graphite," AERE-R 3871, Nov. 1961.
14. R. E. Durand, D. J. Klein and F. R. Nykiel, "Effect of Reactor Irradiation on the Thermal Conductivity of Uranium Impregnated Graphite at Elevated Temperatures," NAA-SR-836, Aug. 15, 1954.
15. A. K. Smalley, R. J. Burian, A. F. Gerds, G. E. Raines and W. S. Diethorn, "The Effect of Radiation on Selected Properties of Spherical Graphite Fuel Elements," BMI-1483, Dec. 2, 1960.
16. R. J. Burian, R. L. Ritzman, J. E. Gates and R. F. Dickerson, "Evaluation of Irradiated Alumina-Coated UO_2 Particles Dispersed in Graphite Spheres," BMI-1572, Mar. 22, 1962.
17. G. B. Engle, "High-Energy Neutron-Induced Changes in Carbon and Graphite - A Review," GA-2802, Sept. 20, 1962.