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UNITED STATES ATOMIC ENERGY COMMISSION

METALLURGY REPORT OF THE TECHNICAL
DEPARTMENT [FOR] MARCH, APRIL, MAY
1956.

By
C. A. Bruch
W. M. Cashin

Photostat Price \$ 7.80

Microfilm Price \$ 3.30

Available from the
Office of Technical Services
Department of Commerce
Washington 25, D. C.

Knolls Atomic Power Laboratory
Schenectady, New York

Technical Information Service Extension, Oak Ridge, Tenn.

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Metallurgy and Ceramics
(M-3679, 18th ed)

General Electric Company
KNOLLS ATOMIC POWER LABORATORY
Schenectady, New York

METALLURGY REPORT OF THE TECHNICAL DEPARTMENT
March, April, May 1956

Prepared by:

C. A. Bruch
W. M. Cashin

W M Cashin
Authorized Classifier

7/2/56
Date

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KAPL-1654REPORTS ISSUED ON METALLURGY AND CERAMICS CATEGORY LIST

The following topical reports containing metallurgical information were issued by the Knolls Atomic Power Laboratory during the period March, April, May 1956.

KAPL-1475, "Silica-Uranium Fuel Systems," W. M. Cashin and H. G. Sowman, March 6, 1956.

KAPL-1525, "Fabrication of Uranium-Rich Uranium-Magnesium Dispersion Fuels by Powdered Hot-Pressing," R. N. Honeyman, May 18, 1956.

KAPL-1518, "Uranium Metal Preparation by Electrolytic Reduction of Oxides," L. W. Niedrach and B. E. Dearing, April 2, 1956.

KAPL-1523, "Remote Operating "TUKON" Hardness Tester," E. R. Craig and R. F. Stearns, April 24, 1956.

KAPL-1493, "X-Ray Measurement of Annealed Samples of Damaged Graphite," D. R. Chipman and B. E. Warren, May 3, 1956.

The previous metallurgy Progress Report in this series is KAPL-1526, issued May 9, 1956 and covering the period December 1955 and January, February 1956.

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METALLURGY REPORT OF THE TECHNICAL DEPARTMENT
March, April, May 1956

SUMMARY

The delta phase region of the uranium-zirconium equilibrium phase diagram, based on new experimental results, is presented.

Melting points of boron and several oxides in the range above 2000°C have been determined. The apparatus used to obtain these data is briefly described.

The structure of the isomorphous calcium and strontium aluminates has been refined further preparatory to determining radiation changes; samples have been encapsulated for MTR irradiation.

High alumina - 5% UO_2 bodies have been made and tested in air and the preliminary results are quite promising.

A technique for the production of plutonium - uranium oxide breeder fuel compacts is being developed. The present extrusion method yields relatively low density. Future work will include a study of the effects of additives to the oxide.

Techniques for impregnating leached unfired VYCOR* are being standardized. Weight gain, as a function of concentration of impregnating solution, is being determined. Microstructural studies show a discrete dispersion of U_3O_8 in the fired silica matrix.

Hardness changes are given for metals which were exposed to 1.75×10^{22} thermal nvt. A review of all hardness measurements shows that radiation hardening has not saturated for titanium after this long exposure. The results for copper, molybdenum, iron, and zirconium show that the radiation hardening saturates at exposures below 1.75×10^{22} thermal nvt. An analysis is made of the possible effect of flux on the radiation hardening.

Irradiation results on 40 wt % uranium-zirconium alloys are summarized. The effect of melting on final behavior of these alloys is noted, and fuel distortion as a function of temperature is described. Preliminary results on 47 1/2% uranium-zirconium alloys have been obtained.

Irradiation results on VYCOR indicate that cracking occurs with no fuel present when a lead-bismuth alloy is used as a coolant.

Irradiation results on two samples of plutonium - uranium oxide have been obtained. Relatively low temperature irradiations still result in extensive sintering of the oxide; however, the addition of a liquid lead core-to-cladding thermal bond prevents sintering and central void formation in the 65% dense oxide solid solution.

*Trade Mark, Corning Glass Works

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I. ALLOY STUDIES

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I. ALLOY STUDIESURANIUM-ZIRCONIUM SYSTEM - J. F. Duffy, C. A. Bruch

5420-2

Work is continuing on the determination of the boundaries of the delta phase of the uranium-zirconium system.

The tentative phase diagram reported in the previous Progress Report (KAPL-1458) has been revised as the result of metallographic examination of annealed and quenched specimens. The suggested form of the delta phase region of the uranium-zirconium system is shown in Figure 1. As shown, there is a peritectoid reaction at 617°C and at approximately 38.5 wt % zirconium. The previously suggested eutectoid reaction was found to be inconsistent with new metallographic data. Alloys of the same composition, annealed and quenched from temperatures above and below the transformation temperature at 617°C, show a decrease in the amount of precipitate in alloys quenched from below 617°C. As a result, the reaction is considered to be a peritectoid.

The diagram also shows the solubility limits of the delta phase, and a eutectoid reaction at 606°C and at approximately 54.8 wt % zirconium.

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FIGURE I THE DELTA PHASE OF THE URANIUM-ZIRCONIUM EQUILIBRIUM SYSTEM

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II. CERAMIC STUDIES

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II. CERAMIC STUDIESMELTING POINT DETERMINATIONS - L. G. Wisnyi, S. Pijanowski

5430-1

The basis of any thermal investigation is the determination of melting points. To facilitate a rapid and fairly accurate determination of melting points, a small ribbon furnace has been set up (see Figure 2). This furnace may be used to obtain temperatures up to:

1. 1800°C in air or oxygen when a platinum - 20% rhodium ribbon is used, and
2. 3000°C when a tungsten ribbon is used in hydrogen, helium or a vacuum.

The ribbon, 1/8 inch wide and 0.005 inch thick, is bent in the form of a "V" wedge and is used as the container for the sample. As the temperature of the wedge is raised, by passing a current through the ribbon, the sample may be observed constantly by using an optical pyrometer. The melting point is determined at that temperature where the sample appears to vanish.

The explanation of this phenomenon lies in the relative emissivities of the wedge and the sample. The emissivity of the sample is much lower than that of the wedge; therefore, it always appears darker. When the sample melts, the resulting liquid permits the near-black body temperature of the wedge (emissivity of 0.90 to 0.95) to be measured.

The furnace and optical pyrometer are both standardized by melting very pure Al_2O_3 . The melting point of Al_2O_3 as given by A. S. Russell⁽¹⁾ in 1953 is 2040°C (on the International Temperature Scale of 1948). All melting points above 1900°C can then be given relative to Al_2O_3 . In other words, the melting point of pure Al_2O_3 is hereby accepted as the standard above the melting point of pure platinum (1769°C).⁽²⁾ The precision with which the measurements are made is that relating to the difference between the melting point of Al_2O_3 and that of the unknown.

Some of the observed melting points which can be reported at this time are:

<u>Material</u>	<u>Melting Point, °C</u>
1. UO_2	2750 $\pm$ 50
2. B	2040 $\pm$ 30
3. Sm_2O_3	2300 $\pm$ 50
4. Eu_2O_3	2050 $\pm$ 30
5. Gd_2O_3	2330 $\pm$ 20
6. Dy_2O_3	2340 $\pm$ 10

⁽¹⁾Russel, A. S., "Alumina Properties," Aluminum Company of America, 1953, p. 10.

⁽²⁾Sosman, R. B., "Temperature Scales and Silicate Research," American Journal of Sciences, 1952, p. 527.

STRUCTURE STUDIES ON ALKALINE EARTH ALUMINATES - E. R. Boyd, W. J. Pojasek
5420-1

A study of the high-temperature and radiation stability of the isomorphous compounds $\text{CaO} \cdot 2\text{Al}_2\text{O}_3$ and $\text{SrO} \cdot 2\text{Al}_2\text{O}_3$ is in progress.

The melting points of the compounds were determined to be:

<u>Compound</u>	<u>Melting Point</u>
$\text{CaO} \cdot 2\text{Al}_2\text{O}_3$	$1745 \pm 15^\circ\text{C}$
$\text{SrO} \cdot 2\text{Al}_2\text{O}_3$	$1780 \pm 20^\circ\text{C}$

The melting point was reported to be 1770°C .⁽¹⁾ Prolonged exposure at temperatures just below the melting points was found to have no effect.

X-ray photographs of the $\text{SrO} \cdot 2\text{Al}_2\text{O}_3$ were taken, and the lattice constants of the monoclinic cell were determined to be:

$$a = 13.04; b = 9.01; c = 5.55; \beta = 106.5^\circ$$

This is in agreement with the work of Lagerqvist, Wallmark, and Westgren:⁽²⁾

$$a = 12.95; b = 9.00; c = 5.54; \beta = 106.3^\circ$$

For the calcium salt, the reported density and ours, 2.90, corresponds to 4 molecules in a unit cell, however, no detailed structures have been reported on these compounds.

The compounds were found to belong to the space group $\text{C}2/c$ with the Sr in the special positions.

$$\text{Oy } 1/4; \text{O}\bar{\text{y}} 3/4; 1/2, 1/2 + y, 1/4; 1/2, 1/2 - y, 3/4; \text{where } y = 0.189$$

The x and y parameters of the two Al atoms in the asymmetric zones were determined to be:

$$\text{Al}_{\text{I}} \quad x = 0.170; y = 0.084$$

$$\text{Al}_{\text{II}} \quad x = 0.122; y = 0.436$$

From symmetry considerations, one oxygen atom must be in a special position. This was found to be: $0 \ 1/2 \ 0, 0 \ 1/2 \ 1/2, 1/2 \ 0 \ 0, 1/2 \ 0 \ 1/2$.

⁽¹⁾Goria and Burdese, Ric. Sci. Ital. 21, 1613 (1951).

⁽²⁾Lagerqvist, K., S. Wallmark and A. Westgren, Z. Anorg. Chem. 234, (1937).

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FIGURE 2

HIGH-TEMPERATURE MELTING POINT FURNACE

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The determination of the z parameters of the two Al atoms and the parameters of the other three oxygen atoms in the asymmetric zone is in progress.

Samples of the calcium and strontium aluminates are being encapsulated for irradiation in the MTR. Structural changes occurring will be observed by the use of X-rays.

HIGH ALUMINA FUEL BODIES - H. G. Sowman, C. W. Krystyniak, W. M. Cashin
4540-1

In the development of superior high-temperature fuel elements, high alumina 5 wt % UO_2 bodies have been fabricated and tested in air up to $1500^\circ C$. The results are encouraging in that without a concerted effort to obtain uranium-free, dense, surface structures, these bodies showed quite good dimensional stability, though some weight losses occurred - probably caused by oxidation of that UO_2 which was on the surface. All the bodies were prepared by cold pressing the powders containing Ceremul C binder and subsequently firing for one hour at $1750^\circ C$, in hydrogen.

The data on the materials are summarized in Table 1. The samples after the high-temperature test are shown in Figure 3. Future work will include the radiation testing of high alumina bodies containing enriched uranium. The potentialities of other refractory matrices are to be defined by future work.

TABLE I

HIGH ALUMINA - UO_2 REFRACTORIES

Body Designation	Composition, wt %	Test Conditions in Air		Initial Wt, g	Final Wt, g	Wt Change, g
		$^\circ C$	Hrs			
KB-138	93.1 Al_2O_3	1500	20	6.567	6.523	-0.034
	1.9 CaF_2	1200	24			
	5.0 UO_2					
KB-139	93.1 Al_2O_3	1500	20	6.478	6.399	-0.139
	1.9 TiO_2	1200	24			
	5.0 UO_2					
KB-140	95.0 GE 2548*1500		20	6.523	6.322	-0.29
	5.0 UO_2	>1200	24			

*GE 2548 is an electrical ceramic material containing 95% Al_2O_3 , 2% clay, 3% talc.

~~CONFIDENTIAL~~OXIDE FUEL EXTRUSION - J. N. Frank

4540-2

Attempts to obtain a satisfactory technique for the production of an extruded mixed uranium and plutonium oxide fuel element were continued. However, the handling procedures and the expense of the mixed oxide prohibited its use as an experimental material; therefore, normal UO_2 was used in all extrusion experiments to date. The UO_2 was produced from the reduction of ammonium diuranate in hydrogen, in agreement with the proposed manufacturing procedure for the mixed oxide.

Initial tentative requirements for the extruded fuel rod were: (1) close dimensional tolerances on rod diameter (1 to 2 mils), (2) absence of foreign material (additives), and (3) a minimum of 65% of theoretical density.

The basic variables governing the density of extruded UO_2 were found to be:

1. Particle size of fuel material
2. Type and per cent of binder material used
3. Per cent of water added to oxide
4. Precompaction of extrusion charge
5. Deaeration of material before extrusion
6. Sintering temperature
7. Sintering time

Possible control of the particle size of the fuel had been considered. It was decided that this control was both economically undesirable and hazardous in the manufacture of mixed oxide fuel; therefore, it was eliminated as a variable. The particle size distribution was assumed constant and fixed, because the oxide would be prepared in exactly the same manner and from the same starting materials, for each batch of material. The remaining variables in the extrusion process were investigated and the following results obtained:

1. Maximum densities of the fuel rod - when sintered at maximum obtainable temperature (1800°C) for 3 hours.
2. Good dimensional stability - when oxide was extruded with higher than 1 wt % binder and greater than 10 wt % water. However, this material fell appreciably short of the minimum acceptable density of 65% of theoretical.
3. Acceptable density (65% after sintering) - by reducing the binder and water percentages in the extrusion charge but resulting in very poor dimensional characteristics both in length, diameter and curvature of rod.

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KB 138

KB 139

KB 140

Tested in Air: 20 hrs 1500°C
24 hrs 1200°C

FIGURE 3

HIGH ALUMINA - 5 WT % UO_2 CERAMIC BODIES

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4. Precompaction and deaeration of material - had negligible effects on the final density of the UO_2 .

The conclusions gathered from experimental results are that an additive such as TiO_2 should be included in the extrusion mix to obtain both high density and dimensional stability in the sintered UO_2 .

U_3O_8 - SILICA DISPERSIONS - W. H. Howland

4540-1

The study of U_3O_8 dispersons⁽¹⁾ in VYCOR has been continued. Recent investigations have been made relating to the effect of varying the concentration of uranyl nitrate aqueous solution on the final concentration and distribution of uranium in porous VYCOR. Porous VYCOR, soaked in the nitrate solutions and subsequently heat treated, produces an element of dispersed U_3O_8 particles in a glass matrix. It has been reported⁽²⁾ that in the porous state, VYCOR has a surface area slightly above 200 square meters/gram and that the pores are in the form of cylinders with average radii of 20 Å. If heated to 1200°C for two hours or less, the pores are closed completely. The procedure for impregnating these pores with U_3O_8 is given below.

1. A short length of 1/8-inch rod is degassed under high vacuum until the pressure is less than 0.1 μ of mercury. The samples are then weighed in air at measured time intervals and the weight of the evacuated sample is determined by extrapolation to zero time. (The weight change is linear in time for the first ten minutes with this particular diameter rod. Then the weight rises as the square root of time, which is characteristic of a diffusion-limited process. Approximately 24 hours is required for saturation with water vapor.)
2. The rod is then re-evacuated and exposed to an aqueous solution of $\text{UO}_2(\text{NO}_3)_2$ which has been deaerated. Solutions containing 0.213, 0.319, 0.425 and 0.565 gram uranium per ml of solution were used in this study.
3. After the rod has been dried at 65°C for 100 hours or more, the temperature is raised above 300°C for at least 24 hours to decompose the salt. The temperature is raised to above 900°C for at least 4 hours, then to 1200°C for 2 hours to close the pores around the U_3O_8 particles. The uranium content is determined by weight difference.

(1) KAPL-1475, "Silica Uranium Fuel Systems," W. M. Cashin and H. G. Sowman, March 6, 1956.

(2) Nordberg, M. E., "Properties of Some VYCOR-Brand Glasses," J. Am. Cer. Soc. 27, No. 10 (1944).

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Figure 4 shows the relationship between per cent weight change of a saturated VYCOR rod and the concentration of the solution in which it was soaked (grams of uranium per ml of solution). The points representing the longer samples are at consistently lower concentrations than those for the shorter samples despite the fact that they were soaked for a longer period of time. The reason for this apparent systematic error is somewhat obscure at this time.

Preliminary attempts to determine the particle size of the fuel by metallographic methods have shown some promise. Photomicrographs of polished surfaces ranging from the center to the outside of one sample have shown that the particle sizes are between 0.01 and 0.08 mm, and that the radial concentration distribution is fairly uniform in a sample that soaked in solution for one week. Figure 5A shows a view of a surface perpendicular to the axis of symmetry of a 1/8-inch VYCOR rod containing U_3O_8 . Figure 5B shows a surface parallel to the axis of symmetry, at the center of the same rod, and Figure 5C a view of a surface parallel to the axis of symmetry and perpendicular to the surface of Figure 5B. The apparent difference in densities of sections of these photographs is a result of printing difficulties. It can be concluded from these observations that the fuel density is uniform and without preferred orientation.

Further time-temperature studies are in progress to obtain empirical relations between solution concentration, time and temperature, and the final density of uranium in VYCOR. With this information, samples of known initial properties will be prepared for irradiation studies.

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A. End View

B. Side View

C. Side View
(Rotated 90° from B)

FIGURE 5

VYCOR ROD IMPREGNATED WITH U_3O_8
100X

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III. IRRADIATION EFFECTS

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III. IRRADIATION EFFECTS

THE EFFECT OF IRRADIATION ON THE MECHANICAL PROPERTIES OF METALS -

C. A. Bruch, W. E. McHugh, J. Swota

5430-2

A study of the effect of three reactor variables, flux, time integrated flux, and exposure temperature on some mechanical and physical properties of a group of metals which represent the three common crystallographic structures is in progress. A detailed description of the metals being studied, copper, nickel, iron, molybdenum, titanium, zirconium, and Type 347 stainless steel, has been given in KAPL-1402, p. 59.

The results to be reported are for specimens which were irradiated in the MTR for one year.

Irradiation Conditions

The specimens were contained in tight-fitting aluminum tubes and irradiated in active lattice position L-41 of the MTR at a temperature of less than 95°C. The estimated exposure which varied from 6×10^{21} to 1.75×10^{22} thermal nvt, was as a result of the approximately cosine distribution of the neutron flux in this reactor position.

Hardness Results

Rockwell hardness measurements were made before and after irradiation. The results are given in Table II, together with Brinell numbers as converted from Rockwell values.

It was found for each metal that the change in hardness of each specimen was independent of position in the reactor. Therefore, average changes in property are reported in Table II.

All of the metals have shown large increases in hardness as a result of this irradiation. Type "A" nickel has shown an extremely large increase in hardness of 155 Brinell numbers.

25

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TABLE II

IRRADIATION INDUCED HARDNESS INCREASES FOR VARIOUS METALS*

Specimen Material	No. of Spec. Tested	Condition	Rockwell Hardness				500 kg Bhn
			Scale	Avg**	Min	Max	
Copper	6	Unirradiated	F	41.8	39.6	44.1	42.8
		Irradiated	F	93.8	93.0	94.5	100.9
		Change	F	+52.0	49.5	+54.0	+58.1
"A" Nickel	11	Unirradiated	B	50.7	48.9	51.9	83.7
		Irradiated	B	105.7	102.4	107.0	239.0
		Change	B	+55.0	+53.5	+57.0	+155.3
Zirconium	12	Unirradiated	B	33.5	30.8	37.0	69.6
		Irradiated	B	63.5	61.4	64.9	100.3
		Change	B	+30.0	+27.7	+32.8	+30.7
Titanium	12 (4)	Unirradiated	B	94.7	92.1	98.6	188.5
		Unirradiated	C	20.8	19.5	21.2	
	(8)	Irradiated	B	103.3	101.4	107.0	240.7
		Irradiated	C	32.8	31.5	33.5	
	(4)	Change	B	+8.6	+5.5	+11.3	+52.2
		Change	C	+12.0	+9.4	+12.8	
Molybdenum	5	Unirradiated	C	22.6	22.1	23.5	202.0
		Irradiated	C	29.5	28.6	30.2	230.3
		Change	C	+6.9	+6.3	+7.7	+28.3
Iron	12	Unirradiated	F	60.6	58.1	61.7	55.6
		Irradiated	F	92.4	90.9	94.8	98.4
		Change	F	+31.8	+29.3	+33.9	+42.8

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Ann. 347 SS	12	Unirradiated	B	82.7	81.5	83.6	136.4
		Irradiated	B	102.2	101.1	102.8	214.8
		Change	B	+19.5	+17.6	+20.6	+78.4
7% CW 347 SS	3	Unirradiated	C	22.1	21.8	22.3	200.0
		Irradiated	C	31.1	30.7	31.4	239.1
		Change	C	+9.0	+8.9	+9.1	+39.1
17% CW 347 SS	3	Unirradiated	C	28.7	27.6	30.0	227.0
		Irradiated	C	33.4	33.2	33.7	255.4
		Change	C	+4.7	+3.4	+5.6	+28.4
37% CW 347 SS	3	Unirradiated	C	31.1	30.9	31.4	239.7
		Irradiated	C	36.4	36.1	36.6	274.5
		Change	C	+5.3	+4.7	+5.5	+34.8
58% CW 347 SS	3	Unirradiated	C	34.8	34.2	35.3	262.9
		Irradiated	C	38.5	38.2	38.8	290.1
		Change	C	+3.7	+3.4	+4.2	+27.2

*Irradiated at the MTR to an estimated exposure of 6.0×10^{21} to 1.75×10^{22} thermal nvt and at a temperature of less than 95°C.

**At least 3 Rockwell impressions were made per specimen.

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The Effect of Exposure on Radiation Hardening

Data which may be used for an evaluation of the effect of nvt on the hardness increase resulting from irradiation are presented in Table III, and are shown in Figure 6. These data were obtained from previous experiments as well as from those results presented here. On each graph in Figure 6 is shown the error in Brinell Hardness numbers due to an error of ± 1 Rockwell number in the irradiated condition. As shown, the errors vary considerably and are a function of the Rockwell scale used and the hardness level of the metal.

On the basis of these data the following observations can be made concerning the effect of increasing neutron exposure on the hardening of the metals studied:

1. Copper - hardness data indicate saturation after an exposure of approximately 6.5×10^{20} thermal nvt.
2. Titanium - hardness change does not saturate after an exposure of 1.75×10^{22} thermal nvt.
3. Zirconium - hardness change may saturate after an exposure of 6.0×10^{21} thermal nvt.
4. Molybdenum - increase in hardness saturates after an exposure of approximately 6×10^{20} thermal nvt.
5. Iron - hardness increase has saturated after an exposure of 5.9×10^{20} thermal nvt.

This analysis of the data can be compared with a similar analysis (KAPL-1349, p. 109), based on fewer data points. The trends given in the previous analysis for copper, molybdenum, and zirconium are in good agreement with these latter observations. However, the earlier analysis of titanium indicated saturation after an exposure 2.2×10^{20} thermal nvt. The tensile test data (KAPL-1458, p. 101) were in agreement, and showed saturation at 2.2×10^{20} thermal nvt. The more recent data show that the earlier observations were in error, and that titanium continued to harden with increasing exposure. If only hardness measurements had been involved, it would have been relatively easy to understand the error. As shown in Figure 6, the potential error in hardness measurements is relatively large. However, since yield strength measurements were also used in the earlier analysis, the reason for the indication of saturation at lower exposures is not understood at this time.

There is a possibility that increasing exposure actually softens molybdenum. As shown in Figure 6, the hardening at an exposure of 6.9×10^{20} is much greater than at exposures of 6×10^{21} to 1.75×10^{22} . It was also observed that the change in electrical resistance was much greater for the corresponding lower exposure than in the highest exposures. An analysis of this possibility will be made after tensile data have been obtained.

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TABLE III

EFFECT OF INTEGRATED NEUTRON EXPOSURE ON THE HARDNESS INCREASE
OF SEVERAL METALS

<u>Metal</u>	<u>Irradiation Site</u>	<u>Estimated Thermal nvt x 10⁻²⁰</u>	<u>Estimated Exposure Temp, °C</u>	<u>Δ 500 kg Bhn</u>
Cu	BNL	0.08	60 to 100	+18.5
	MTR	2.1	90	+53.5
	MTR	6.4	90	+58.0
	MTR	60.	90	+58.1
	MTR	175.	90	+58.1
Ti	BNL	0.08	60 to 100	+ 6.4
	MTR	2.2	90	+33.0
	MTR	6.6	90	+33.0
	MTR	60.	90	+52.2
	MTR	175.	90	+52.2
Zr	BNL	0.08	60 to 100	+ 7.9
	MTR	1.9	90	+14.5
	MTR	5.9	90	+24.0
	MTR	60.	90	+30.7
	MTR	175.	90	+30.7
Mo	BNL	0.08	60 to 100	- 5.0
	MTR	1.9	90	+12.0
	MTR	5.9	90	+32.0
	MTR	6.9	90	+43.7
	MTR	60.	90	+28.3
Fe	MTR	175.	90	+28.3
	BNL	0.08	60 to 100	+10.4
	MTR	2.3	90	+35.0
	MTR	5.9	90	+50.0
	MTR	60.	90	+42.8
	MTR	175.	90	+42.8

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The Effect of Flux on Radiation Hardening

The data in Table III may also be used to evaluate the possible effect of neutron flux hardness increase resulting from the irradiation. The thermal neutron fluxes at the various sites at which the specimens were irradiated are as follows:

<u>Site</u>	<u>Estimated Thermal Flux</u>
BNL	$3.5 \text{ to } 4.1 \times 10^{12}$
MTR	$1.0 \text{ to } 3.1 \times 10^{14}$

To evaluate the flux effect, specimens were irradiated in the MTR for times suitable to give approximately equal nvt values. A comparison may be made of the effect of these irradiations on the hardness changes. The indications for the various metals, assuming that the fast neutron fluxes differ by approximately the same amount as do the thermal neutron fluxes, are as follows:

1. Copper - there are not sufficient data available at this time to permit an evaluation.
2. Titanium - 2.2×10^{20} thermal nvt at MTR, has produced a change of 33.0 which indicates that there may be a small flux effect.
3. Zirconium - 1.9×10^{20} thermal nvt at MTR has produced approximately hardness increases of 14.5 and 14.1, respectively. There does not appear to be a flux effect.
4. Molybdenum - 1.9×10^{20} thermal nvt at MTR has produced hardness changes of 12.0 which indicates that there may be a small flux effect.
5. Iron - the 2.3×10^{20} thermal nvt at MTR for which the hardness increase was 35.0. Most of the difference in hardening can be assigned to difference in total exposure; however, there is possibly a small flux effect.

In summary, the indications from the data are that there may be small flux effects on radiation hardening, in the range studied, but that the pre-dominating factor is the total exposure.

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FIGURE 6 THE EFFECT OF INTEGRATED NEUTRON EXPOSURE ON THE HARDNESS INCREASE OF SEVERAL METALS

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URANIUM-ZIRCONIUM ALLOY IRRADIATIONS - DD Van Horn

4540-1

Specimens of zirconium - 40 wt % uranium alloys were irradiated for various times and temperatures to determine their dimensional stability. The alloys contained uranium enriched to 93% U-235, and were prepared by induction and arc melting methods. Before irradiation, the specimens were annealed by heating to 1050°C for one hour and furnace cooled.

The results are summarized in Table IV. A comparison of arc melted and induction melted specimens irradiated at 90°C to 1.21% burnup of all atoms is shown in Figure 7. The induction melted alloy is much more stable than the arc melted alloy, presumably because of the higher carbon content in the former which modifies the microstructure.

Figure 8(B and C) shows a comparison of arc and induction melted specimens which were irradiated at 90°C to a burnup of 2.90% of all atoms. Here again the induction melted material appears to be more stable. Figure 8(D) shows an induction melted specimen after 4.83% burnup at 90°C. This sample showed a decrease in length and an increase in diameter, whereas the opposite was true for the specimen irradiated to a lower burnup.

The effect of exposure temperature on the induction melted material is shown in Figure 9. The samples were irradiated at 90, 400 and 750°C; the 750°C temperature was in the gamma phase region. The sample irradiated at 400°C shows approximately 5 to 10 times less damage than those irradiated at 90°C. The sample irradiated at 750°C is very extensively damaged and shows great swelling in all directions. The spiral grooves are caused by the constraining action of the wire basket in which the specimen was irradiated.

Six samples of 47 1/2 wt % uranium-zirconium alloys, 10% enriched uranium, have been irradiated to 2 1/2 total at. % burnup. Three of the six samples were heat treated to give quenched γ alloy, the remaining three were treated to give the stable σ (previously termed ϵ) structure. One of each phase was irradiated at three temperatures, 150, 550°C (below the σ to γ transformation), and 650°C (above the σ to γ transformation temperature). In a preliminary examination of the irradiated material, it appears that both σ and γ alloy performed very well at 550 and 650°C, and some slight bowing occurred at the lowest temperature. Detailed measurements and microstructural examinations are to be made.

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TABLE IV

EFFECT OF IRRADIATION ON DIMENSIONAL STABILITY
OF ZIRCONIUM - 40 WT % URANIUM ALLOYS

<u>Figure No.</u>	<u>Material</u>	<u>Burnup (% all atoms)</u>	<u>Temp of Irradiation, °C</u>	<u>Growth Coefficient</u>	
				<u>Growth (%/ % Burnup)</u>	<u>Diameter</u>
1 A	Induction melted	1.21	90		
1 B					
1 C					
1 D	Arc melted				
1 E					
1 F					
2 B	Arc melted	2.90	90		
2 C	Induction melted	2.90	90	0.07	-1.0
2 D	Induction melted	4.83	90	-2.0	0.7
3 (20-2)	Induction melted	4.83	90	-2.0	0.7
3 (20-5B)	Induction melted	3.76	400	0.2	-0.1
3 (20-5A)	Induction melted	3.4	750		7.0

SILICA IRRADIATIONS - W. M. Cashin

4540-1

Past work on the performance of silica-uranium fuel dispersions has been summarized in KAPL-1475 (March 6, 1956). It has been found that a 3 wt % dispersion of U-235, as U_3O_8 , in VYCOR did not release any fission products when irradiated in the MTR to 3% depletion of the uranium. The coolant used in the irradiation was a lead-bismuth eutectic alloy. Some cracking of the VYCOR pieces occurred; this was attributed possibly to reaction of the molten coolant with the VYCOR (which contains sufficient B_2O_3 to make such an interaction quite probable).

VYCOR containing 3 to 5 wt % B_2O_3 but no fuel was irradiated in the MTR for one cycle at a flux near 0.5×10^{14} nvt - a level equivalent to that attained in the U-235 VYCOR specimen. The specimens were somewhat discolored, and cracked in the same manner as the samples containing U-235. Figure 10 shows the irradiated samples; the cracks appear to have been initiated at the surface. In the early work, silica pieces containing 3 wt % unenriched uranium were irradiated in helium and no cracking occurred. One may infer, therefore, that the coolant, lead-bismuth, may be the prime contributor to the break up observed. Further irradiation testing of silica matrix material with higher fuel loadings is planned.

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A, B, C - INDUCTION MELTED
D, E, F - ARC MELTED
G - STANDARD

FIGURE 7 EFFECT OF METHOD OF PREPARATION
ON STABILITY OF ZR - 40 WT. % U ALLOYS.

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A - STANDARD C - INDUCTION MELTED
B - ARC MELTED D - INDUCTION MELTED

FIGURE 8 EFFECT OF BURN-UP ON THE
STABILITY OF ZR - 40 WT. %
U ALLOYS.

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FIGURE 10

VYCOR SAMPLES IRRADIATED IN LEAD-BISMUTH
COOLANT. NVT $\sim 10^{20}$

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PLUTONIUM-URANIUM OXIDE FUEL PERFORMANCE - W. M. Cashin

4540-2

Additional samples of dry-pressed mixed plutonium-uranium oxide have been irradiated in the MTR. The results of the first irradiation have been reported in KAPL-1446 (December 7, 1955).

All elements tested to the present time are similar in that they contained one inch of dry-pressed powder, 0.1875 inch diameter dry-pressed to 65% of theoretical density. The fuel content is 0.41 to 0.42 gram of plutonium in a 1 to 5 solid solution of PuO_2 and UO_2 .

The second irradiation test (KAPL 26-2) was placed in the MTR in September for an 18-week irradiation - to a calculated 35% depletion of the original plutonium. Since the conversion of uranium to plutonium is high (~ 0.5) in this system, the total plutonium present after irradiation is near 0.3 gram. The capsule was a low-temperature capsule, as shown in Figure 11, without heater or thermocouple leads. The estimated coolant temperature was 300°C and the power generation near 2500 watts/gram, or 1000 watts for the capsule. The capsule was sectioned, examined and the oxide dissolved in the RML. Sectioning was performed near the top and bottom of the fuel and through the approximate midplane. The oxide was sintered, cracked and a central void had been formed, quite similar in appearance to the fuel which operated at 660°C (KAPL 26-1). Figure 12 shows a section at the top of the fuel; the center section of the sample because it was too close to the camera is out of focus. The oxide was unsintered and easily removed from the periphery, but sintered near the center. Figure 13 shows the section through the midplane. Here the central void formation is evident, along with major cracking. It has not been possible to determine on these samples whether any settling of the fuel has occurred.

In the third (KAPL 26-3) PuO_2 - UO_2 irradiation, the fuel was contained in a 0.25-inch-ID, 0.34-inch-OD tube. The void space between the 0.188 diameter fuel specimen and the tube walls was filled with lead. This fuel element was irradiated at the MTR for one cycle at a flux near 0.7×10^{14} nv. The capsule design is identical to that reported in KAPL-1446 for irradiation KAPL 26-1. The NaK coolant operated at a maximum temperature of 550°C , the estimated heat generation for the sample was 1 kw and the plutonium depletion is estimated to be 3%.

The capsule was opened in the RML. The steel element showed no dimensional changes. The element was sectioned at top and bottom of the fuel, and three other sections were made through the fuel. It was apparent that the fuel had floated on the lead for there were void spaces near the top. It is important to note that the addition of lead increased the core-to-cladding heat transfer sufficiently to prevent much sintering of the oxide and no central void could be found.

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Figure 14 shows a section at the top of the fuel. The cracking is not typical of the sintered irradiated fuel observed in KAPL 26-2, but is reminiscent of dried mud flats. The oxide was quite friable, as checked with a probe, which indicated that little, if any, sintering occurred.

A section through the fuel midplane is shown in Figure 15. At this point the maximum temperature was attained and there is still no evidence of sintering. It appears that it would be quite feasible to use a liquid metal bond such as lead or sodium in this system, and such a bond would prevent central coring of 65% dense oxide. It is not by any means certain that such coring must be avoided.

Another fuel capsule similar to KAPL 26-2 is being prepared for an MTR irradiation and to provide additional long burnup fuel for chemical processing studies. The reprocessing studies on the fast oxide breeder fuel is described in KAPL-1536 (to be issued).

DIFFUSION OF FISSION GASES IN URANIUM-ALUMINUM ALLOY M. B. Reynolds 2722-2

The study of fission krypton diffusion in low-burnup irradiated 20 wt % uranium-aluminum alloy has been continued. There is now strong evidence that, although no gas diffuses through the aluminum below its melting point, there is appreciable diffusion of the gas within the dispersed UAl_4 phase present in the uranium-aluminum alloy. Thermal cycling, which has a marked effect on fission gas diffusion in metallic uranium, has no effect on uranium-aluminum alloy of this composition if the upper temperature is below that of the eutectic.

If a specimen is heated immediately to a temperature above the eutectic, gas diffusion takes place at a rate dependent upon temperature and shows approximately the proper square root dependence upon time. As described in the previous Progress Report, KAPL-1526, when the temperature is increased to a value above the decomposition temperature of UAl_4 , the total free gas activity rises rapidly to a value essentially constant and independent of previous annealing history. Calculations based on burnup (which was determined by chemical analysis) show that essentially all of the radiokrypton has been released.

A summary of the diffusion studies is contained in Table V. It will be noted that diffusion was studied at temperatures above and below the eutectic as well as above the temperature of decomposition of UAl_4 . Results of these experiments are given in Figure 16 in which total krypton activity as counts per minute is plotted against time at temperature. Curves 4B and 5B indicate that the quantity of fission krypton evolved from a specimen annealed for a long period at a temperature just below the eutectic and then for a short time just above the eutectic is approximately equal to the quantity evolved from a like specimen annealed for the same total time at a temperature just above the eutectic temperature. Since no gas is evolved until the matrix is molten, gas diffused from within the UAl_4 particles at

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FIGURE 11

IRRADIATION CAPSULE, KAPL 26-2

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Central Section

1/32"

FIGURE 12

IRRADIATION CAPSULE KAPL 26-2

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Top View

1/32"

FIGURE 13

IRRADIATION CAPSULE KAPL 26-2

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Cracked
Oxide
Fuel

Lead

Steel

Annular
Gap

Top View

1/32"

FIGURE 14

IRRADIATION CAPSULE KAPL 26-3

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Central Section

1/32"

FIGURE 15

IRRADIATION CAPSULE KAPL 26-3

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FIGURE 16

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approximately the same rate in both cases, but was trapped at the interfaces in the specimen annealed with the matrix solid. Upon finally melting the matrix, escape of this gas would bring the total quantity of gas evolved up to the level achieved by the specimen annealed the same length of time with the matrix molten. The small "step" on curve 4B may be caused by the escape of this trapped gas since it corresponds very closely to the difference in level between the extrapolated plateau of 5B and the level which would have been reached by 4B in the same total time if diffusion from the particles had not started until the time matrix was finally melted.

The times for the radiokrypton activity to reach 1100 counts/minute were determined from curves 5B, 7B, and 8B, respectively. By using these times and their corresponding temperatures an activation energy of 200 ± 50 kcal/mol was computed for the diffusion process within the UAl₄ particles. The significance of this unusually high activation energy is not apparent at this time.

An additional fuel dispersion system being studied is the UO₂ - silver system. Two series of specimens, having different particle size ranges, have been prepared by powder metallurgy techniques. With these the effect of particle size on gas diffusion in a UO₂ dispersion may be observed.

TABLE V
ANNEALING SCHEDULE

<u>Experiment Number</u>	<u>Time, days</u>	<u>Temperature, °C</u>
3B	5.8	670
	1.2	795
4B	21	610
	7	655
	2	750
5B	19.7	660
	4.3	755
6B	19.1	605*
	6.9	655
	11	750
7B	7	700
	2	750
8B	-	680

*Cycled to room temperature 103 times

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FISSION PRODUCT DAMAGE VERSUS NEUTRON INDUCED DAMAGE - F. W. Kunz, C. A. Bruch
4540-1

This program is designed to study and separate fission product damage from neutron induced damage in a 1 wt % uranium-zirconium (0.387 at. % uranium) solid solution alloy. The enrichment of the uranium will be varied to produce changes in the number of fission events for three neutron exposure levels, 10^{18} , 10^{19} and 10^{20} nvt. The irradiation effects will be studied by measuring the changes in tensile properties, hardness and electrical resistivity and also the recovery rates of these properties on annealing. Details of the program are given in KAPL-1458 (February 10, 1956).

Five ingots of the 1 wt % uranium-zirconium alloy, composed of hafnium-free - crystal bar zirconium and 1 wt % uranium have been prepared, the depleted uranium alloy by consumable arc melting and the enriched uranium alloys by tungsten arc melting in vacuum. The uranium-zirconium alloys contain the following enrichments:

<u>Alloy</u>	<u>% Enrichment</u>
A	0.712
B	10.5
C	40.3
D	93.13
E	U-235 100 to 300 ppm

Each ingot was worked and triple melted to improve the homogenization.

The billets are in the process of being fabricated into rod suitable for specimen requirements.

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