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IRRADIATION OF EXTRUSION-CLAD URANIUM-2 w/o
ZIRCONIUM ALLOY FOR EBR-I, MARK III

by

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IRRADIATION OF EXTRUSION-CLAD URANIUM-2 w/o ZIRCONIUM ALLOY FOR EBR-I, MARK III

by

J. H. Kittel

ABSTRACT

The fuel material specified for the Mark III core of EBR-I was uranium-2 w/o zirconium alloy coextruded with Zircaloy-2 cladding. From previous work on swaged or rolled uranium-2 w/o zirconium alloy, it was anticipated that the extruded alloy would be dimensionally unstable under irradiation unless stabilized by suitable heat treatment. In order to determine an effective heat treatment, irradiation studies were made on both clad and unclad extruded uranium-2 w/o zirconium alloy specimens at irradiation temperature estimated to range from 200 to 750°C. The irradiation specimens included material with three different heat treatments, selected on the basis of previous studies, and material transiently melted in its cladding. For unclad specimens, it was found that the irradiation temperature strongly influenced the various irradiation growth rates resulting from different heat treatments. Growth rates of the clad specimens were relatively insensitive to either irradiation temperature or prior heat treatment. An exception was the transiently-melted material, which shortened under irradiation. The cladding had only limited ability to restrain the swelling rates of specimens irradiated at the more elevated temperatures. Clad transiently-melted material was found to be most resistant to high-temperature swelling under irradiation. The results of the present study combined with observations in earlier investigations resulted in a recommendation that the reference heat treatment for the core consist of gamma solution at 800°C followed by isothermal transformation at 690°C.

INTRODUCTION

The first two cores of the EBR-I reactor utilized a fuel element design in which the fuel and blanket slugs were not directly clad, but were bonded with NaK to loosely fitting jackets. With this arrangement, the slugs were able to develop appreciable amounts of surface roughening and dimensional changes resulting from irradiation without imposing undue stresses

or distortions in the jacket itself. In addition, in the second core the spacing ribs were removed from the jackets, so that the jackets as well as the fuel slugs were free to bow under thermal stresses.

Although the fuel elements in both cores performed satisfactorily from an engineering point of view, it was noted that, when the reactor was operated under certain conditions, control instabilities appeared, which were manifested in a prompt positive power coefficient.⁽¹⁾ One of the explanations advanced for this behavior was that the fuel rods were free to bow significantly under asymmetric radial temperature distributions. Analyses of all possible sources of instability in the reactor indicated that fuel rod bowing was the most likely.

Because of the importance of the phenomenon with regard to the future development of power from fast reactors, it was decided to continue operation of the reactor with a third core, constructed in such a manner that fuel rod bowing could be eliminated.⁽¹⁾ This core was designated "Mark III." In this design a metallurgical bond was required between the fuel and the cladding. Also, the rod in its fabricated form had to be stabilized against dimensional changes which might result from irradiation.

The maximum temperature expected in the fuel was approximately 390°C.⁽¹⁾ In this temperature range, cast or swaged uranium-2 w/o zirconium alloy had been found to possess adequate high-temperature swelling resistance, and, if properly heat treated, excellent dimensional stability.^(2,3) It was therefore decided to use this fuel alloy and to accomplish the required metallurgical bond between fuel and jacket by coextruding the fuel with Zircaloy-2 cladding. The finished dimensions of the fuel elements called for a fuel diameter of 0.364 in. with 0.020 in. of cladding, giving an overall diameter of 0.404 in.

This report describes the irradiation studies which were made to establish the optimum stabilizing heat treatment for the extruded and clad fuel alloy. Other reports have described the preparation of the fuel and blanket rod stock,⁽⁴⁾ coextrusion of the rods,⁽⁵⁾ assembly into finished elements,⁽⁶⁾ nondestructive testing of the finished elements,⁽⁷⁾ and thermal cycling tests.⁽⁸⁾

EXPERIMENTAL MATERIALS AND PROCEDURE

The irradiation specimens consisted of 1.000-in. lengths cut from a coextruded rod supplied by Nuclear Metals, Inc. and designated by them as No. 14326. The extrusion billet was a composite, consisting of a cast, natural uranium-2.07 w/o zirconium core made at Argonne and a cladding of reactor-grade Zircaloy-2. The composite billet was copper jacketed

and extruded by Nuclear Metals, Inc., at a temperature of 650°C and at a reduction ratio of 22 to 1. After removal of the copper coating, the rod was cold swaged to a final diameter of 0.400 in. The cladding thickness averaged 0.022 in., giving an average fuel diameter of 0.356 in.

Only a relatively short time was available between the selection of the reference fuel material and the required delivery date of the finished core. Because of this circumstance, only one group of specimens could be irradiated in the MTR and evaluated in the Laboratory's hot lab facilities. It therefore was necessary to narrow the heat treatments investigated to a minimum number, and to establish that those which were used on the irradiation specimens were of highest promise. Other considerations affecting the choice of heat treatment were that it should be readily adaptable to normal shop conditions and that conditions should be avoided which would promote an excessive amount of alloying between the cladding and the fuel alloy.

Information found in the literature on irradiations of uranium-2 w/o zirconium alloy coextruded with Zircaloy-2 pertained to a joint study by the Atomic Power Development Associates and Knolls Atomic Power Laboratory,⁽⁹⁾ and to experiments by Westinghouse.⁽¹⁰⁾ In the APDA-KAPL irradiations the coextruded material was found to develop undesirably large elongations. The specimens in the WAPD experiments were found to develop splitting of the Zircaloy-2 cladding after about 1 a/o burnup. In both the APDA-KAPL and WAPD irradiations a limited number of heat treatments were studied, and the heat treatments chosen appeared to have little influence on the behavior of the specimens.

Although a more extensive irradiation study had been made at Argonne on wrought uranium-2 w/o zirconium alloy,⁽²⁾ extruded material had not been included. Nevertheless, it seemed likely that the results obtained on swaged and rolled material might apply to extruded material as well. Results from this study were therefore used to guide the selection of heat treatments for the present extruded specimens.

The most promising heat treatment from the Argonne study consisted of holding the alloy at 800°C until completely transformed to gamma, followed by quenching and subsequent tempering for one week at 690°C. However, excessive grain growth occurred during the long anneal at 690°C. Later experiments indicated that when the alloy was irradiated at elevated temperatures, good stability could be obtained with transformation times at 690°C as short as one hour. It was therefore decided to use this heat treatment on some of the extruded specimens. The previous study also showed that gamma solution at 800°C followed by an isothermal anneal at 500°C was fairly effective in stabilizing swaged uranium-2 w/o zirconium alloy, without grain coarsening. This heat treatment was also selected for some of the extruded specimens.

In order to combine the best features of each of the above two heat treatments, i.e., the favorable texture which resulted in the 690°C quenched and tempered structure and the absence of distortion in the 500°C isothermal anneal, a third heat treatment, which consisted of a gamma solution anneal at 800°C followed by isothermal transformation at 690°C for one hour, was selected.

Finally, it was decided to irradiate material with an as-cast microstructure which had been produced by briefly melting and resolidifying the uranium-zirconium alloy in its Zircaloy-2 cladding. The procedure consisted of passing a clad rod through an induction coil under conditions such that a transient molten zone was formed in the fuel alloy. The technique had been developed by Aronin at Nuclear Metals⁽¹¹⁾ and independently by McCuaig at Argonne National Laboratory.⁽¹²⁾ The as-cast structure was known from earlier studies^(2,13) to be highly stable dimensionally under irradiation.

The details of the four heat treatments used are summarized in Table I. Figures 1 through 4 show the microstructures of the four types of material before irradiation.

Table I

HEAT TREATMENTS USED FOR IRRADIATION SPECIMENS

Treatment No.	Procedure
1	800°C for 15 min, water quenched; 690°C for 1 hr, water quenched.
2	800°C for 15 min, 500°C for 1 hr, air cooled.
3	800°C for 15 min, 690°C for 1 hr, air cooled.
4.	Transient melted in cladding.

Note: In heat treatments 2 and 3, the specimens were transferred from 800°C directly to the isothermal transformation temperature without intermediate cooling.

A total of 16 specimens were prepared for irradiation. Each of the first three heat treatments described in Table I was represented by five specimens. In each of the three groups of five specimens, three specimens were irradiated with their cladding intact, whereas the cladding was machined off the remaining two. It was believed that irradiation of unclad material would permit determination of the basic irradiation behavior of

21431

100X

Figure 1. Microstructure of extruded uranium-2 w/o zirconium alloy heat treated as follows: 800°C for 15 min, water quenched; 690°C for 1 hr, water quenched.

21429

100X

Figure 2. Microstructure of extruded uranium-2 w/o zirconium alloy heat treated as follows: 800°C for 15 min, 500°C for 1 hr, air cooled.

21430

100X

Figure 3. Microstructure of extruded uranium-2 w/o zirconium alloy heat treated as follows: 800°C for 15 min, 690°C for 1 hr, air cooled.

21432

100X

Figure 4. Microstructure of extruded uranium-2 w/o zirconium alloy subjected to transient melting in Zircaloy-2 cladding.

the fuel alloy, unhampered by physical restraint. On the other hand, the clad specimens were expected to provide information that would be helpful in predicting the irradiation behavior of a coextruded fuel element. The final specimen was prepared by transient melting and was left in its cladding. All clad specimens, however, were open at the ends, so that restraint was in a radial direction only.

Five NaK capsules were used to contain the specimens for irradiation. Each capsule contained three specimens representing the first three heat treatments shown in Table I. In addition, one of the capsules contained the transient melted specimen.

Each capsule also contained cobalt flux monitors for determination of the integrated thermal flux. Determination of the integrated exposure enabled the atom burnup in the specimens to be calculated, after suitable corrections were used to compensate for flux depression in the specimens. Also, it was intended to use the integrated flux as a basis for calculating specimen temperatures during irradiation, assuming that the specimens were in an essentially unvarying neutron flux. Unfortunately, approximately midway during their exposure, some of the capsules were moved from one flux level to a widely differing one. This circumstance seriously compromised the accuracy of the temperature calculations, which were made with the aid of an electrical analogue. The reported irradiation temperatures are therefore to be regarded largely as the best estimated values, and refer to maximum temperatures at the centerline of the specimen.

As mentioned earlier, five capsules were prepared for irradiation. Space limitations at the MTR, however, resulted in only four capsules being irradiated, or a total of 13 specimens. Measurements were made on the specimens to determine irradiation-induced changes in length, diameter, weight, and density (by liquid displacement). In addition, hardness changes were determined on the unclad specimens.

RESULTS AND DISCUSSION

The irradiation conditions and the changes observed are summarized in Table II. Figures 5 through 8 show photographs of the irradiated specimens. It can be noted that the specimens were subjected to burnups ranging from 0.051 to 0.16 percent of all atoms. The estimated maximum irradiation temperatures ranged from 200 to 750°C.

Unclad Specimens. The values of irradiation growth rates indicate that the irradiation temperatures greatly affected the differences in growth rates resulting from variations in heat treatment. For example, at an estimated maximum irradiation temperature of 200°C, the unclad specimen which

Table II
SUMMARY OF IRRADIATION EFFECTS ON SPECIMENS OF EXTRUDED URANIUM-2 w/o ZIRCONIUM ALLOY

Specimen No.	Heat Treatment	Cladding Thickness, in.	Estimated Max. Irradiation Temp., °C	Burnup, a/o	Length Change, %	% Length Change a/o Burnup	Max. Diameter Change, %	Density Decrease, %	% Volume Increase a/o Burnup	Weight Change, mg	Hardness Change, RA
CG-22-3	800°C for 15 min, water quenched, 690°C for 1 hr, water quenched	None	200	0.051	2.07	46	0.06	0.24	4.7	-1.4	9.9
CG-18-3	800°C for 15 min, water quenched, 690°C for 1 hr, water quenched	None	700	0.13	0.41	3.2	3.62	5.80	47.3	-6.9	-5.9
CG-24-3	800°C for 15 min, water quenched, 690°C for 1 hr, water quenched	0.022	500	0.13	1.41	11	-0.12	1.89	15.1	-4.3	--
CG-19-3	800°C for 15 min, water quenched, 690°C for 1 hr, water quenched	0.022	700	0.16	1.13	7.1	0.74	7.04	47.2	-3.6	--
CG-22-1	800°C for 15 min, 500°C for 1 hr, air cooled	None	200	0.051	0.26	5.2	0.61	0.74	14.5	-1.5	5.7
CG-18-1	800°C for 15 min, 500°C for 1 hr, air cooled	None	700	0.13	1.94	15	2.70	5.61	45.8	-1.8	0.4
CG-24-1	800°C for 15 min, 500°C for 1 hr, air cooled	0.022	500	0.13	0.66	5.1	0.57	1.27	10.0	-4.2	--
CG-19-1	800°C for 15 min, 500°C for 1 hr, air cooled	0.022	700	0.16	1.72	11	0.70	5.19	34.4	-4.1	--
CG-22-2	800°C for 15 min, 690°C for 1 hr, air cooled	None	200	0.051	1.84	36	-0.17	0.26	5.1	-6.3	0.7
CG-18-2	800°C for 15 min, 690°C for 1 hr, air cooled	None	700	0.13	0.25	1.9	2.68	5.67	46.2	-25.3	-7.9
CG-24-2	800°C for 15 min, 690°C for 1 hr, air cooled	0.022	500	0.13	0.90	6.9	0.37	1.46	11.5	-4.2	--
CG-19-2	800°C for 15 min, 690°C for 1 hr, air cooled	0.022	700	0.16	1.63	10	0.80	5.90	39.1	-4.2	--
CG-18-4	Transient melted in cladding	0.022	750	0.13	-0.31	-2.4	0.80	2.14	17.1	14.6	--

Micro No.

21937

21945

21934

21940

Specimen No.

CG-22-3

CG-18-3

CG-24-3

CG-19-3

Cladding Thickness, in.

none

none

0.022

0.022

Estimated Max. Irradiation Temp., °C

200

700

500

700

Burnup, a/o

0.051

0.13

0.13

0.16

Figure 5. Effect of irradiation on extruded uranium-2 w/o zirconium alloy heat treated as follows: 800°C for 15 min, water quenched; 690°C for 1 hr, water quenched. Magnification 2X. Melting damage is evident where specimens were in contact.

Micro No.

21939

21947

21936

21942

Specimen No.

CG-22-1

CG-18-1

CG-24-3

CG-19-3

Cladding Thickness, In.

none

none

0.022

0.022

Estimated Max. Irradiation Temp., °C

200

700

500

700

Burnup, a/o

0.051

0.13

0.13

0.16

Figure 6. Effect of irradiation on extruded uranium-2 w/o zirconium alloy heat treated as follows: 800°C for 15 min, 500°C for 1 hr, air cooled. Magnification 2X. Melting damage is evident where specimens were in contact.

Micro No.	21938	21946	21935	21941

Specimen No.	CG-22-2	CG-18-2	CG-24-2	CG-19-2
Cladding Thickness, in.	none	none	0.022	0.022
Estimated Max. Irradiation Temp., °C	200	700	500	700
Burnup a/o	0.051	0.13	0.13	0.16

Figure 7. Effect of irradiation on extruded uranium-2 w/o zirconium alloy heat treated as follows: 800°C for 15 min., 690°C for 1 hr, air cooled. Magnification 2X. Melting damage is evident where specimens were in contact.

21944

2X

Figure 8. Effect of 0.13 a/o burnup at an estimated maximum irradiation temperature of 750°C on specimen No. CG-18-4. The specimen is clad with 0.022 in. of Zircaloy-2, and before irradiation was transient melted in its cladding.

had been gamma quenched and tempered at 690°C showed a growth rate of 46% per a/o burnup, whereas the companion specimens isothermally transformed at 500 and 690°C showed growth rates of 5.2 and 36%, respectively. However, at an estimated maximum irradiation temperature of 700°C, differences are evident in the relative stabilities of the unclad specimens. The growth rate of the specimen which had been gamma quenched and tempered at 690°C decreased to 3.2% per a/o burnup, the growth rate of the specimen which had been isothermally transformed at 500°C increased to 15% per a/o burnup, and the growth rate of the specimen which had been isothermally transformed at 690°C decreased to 1.9% per a/o burnup.

The mean irradiation growth rate at 200°C was 29% per a/o burnup. At 700°C, the mean rate was 6.7% per a/o burnup. It is believed that the marked effect of irradiation temperature on the irradiation growth coefficients is related to gamma phases which appear in the alloy above 607°C. Irradiation above this temperature, where the alloy is at least partially gamma, would be expected to decrease anisotropic growth properties.

At the lower irradiation temperature of 200°C the specimen isothermally transformed at 500°C was least stable with respect to the rate of volume change. At the higher irradiation temperature of 700°C all unclad specimens showed a uniformly high rate of volume increase, which was approximately 46% per a/o burnup. Prior heat treatment thus had no influence on volume changes at the higher irradiation temperature. The volume increase resulted both from a general increase in diameter and from surface damage which, as can be seen in Figures 5 through 7, appeared to have resulted from partial melting. The diameter increase is believed to have resulted from high-temperature swelling. Comparable swelling rates were obtained in earlier studies on cast uranium-1.62 w/o zirconium alloys irradiated above 600°C.⁽¹²⁾

The origin of the surface damage appearing on the unclad specimens irradiated at the higher temperature evidently resulted from locally high temperatures. Analogue calculations indicate that it was not possible for the specimens to have reached melting temperatures provided that the NaK bond was maintained. On the other hand, if the three specimens were in contact with one another and the NaK vaporized in the triangular channel thus obtained, it is not unlikely that local hot spots could occur that would result in some melting damage. The general condition of the specimens indicates that the bulk of the material was far below the melting temperature.

Hardness changes resulting from irradiation were largely random. At the lower irradiation temperature, both the specimen gamma quenched and tempered at 690°C and the specimen isothermally transformed at 500°C showed an increase, while no significant change occurred in the specimen transformed at 690°C. At the higher irradiation temperature a negligible hardness change occurred in the specimen isothermally transformed at

500°C. Both the specimen gamma quenched and tempered at 690°C and the specimen isothermally transformed at 690°C decreased in hardness. At least part of the hardness decrease may be due to the influence of minute bubbles that have been observed in swelled specimens.⁽¹²⁾

Clad Specimens. Considering first the heat-treated specimens, length changes under irradiation were generally less affected by differences in heat treatment and irradiation temperature than were those encountered in the unclad group. The physical restraint provided by the cladding was no doubt responsible for the more uniform behavior of the specimens. On the basis of length changes in the clad specimens, none of the three heat treatments investigated appeared to be significantly better than the other two.

Irradiation temperature markedly affected the rate of volume change in the clad specimens almost to the same extent that it did in the unclad material. At the higher irradiation temperature, estimated to be near 700°C, the cladding showed only limited ability to restrain swelling of the fuel alloy, so that the volume changes which developed in the composite specimens were appreciable. The rate of volume increase ranged from 34 to 47% per a/o burnup. As can be noted in Figures 5 through 7, the cladding bulged outward where temperatures were highest, i.e., where adjoining specimens were in near contact.

The transient-melted specimen showed a significantly different behavior under irradiation than did the heat-treated specimens. Instead of elongating, it shortened, at a rate of 2.4% per a/o burnup. In the previous studies of uranium-2 w/o zirconium alloy⁽²⁾ no cast specimens were observed to shorten under irradiation, although shortening was observed in a group of specimens prepared from highly textured material rolled at 565°C. It therefore appears that the transient melting process used for the specimen imparted a nonrandom texture to the alloy on cooling, possibly from directional solidification of the melt.

It was also noted that the transient melted specimen was more resistant to swelling. It increased in volume at a rate of 17.1% per a/o burnup, or at less than half the rates shown by the clad heat-treated specimens irradiated at comparable temperatures. On the basis of the one specimen used in this investigation, transient melting appears to have considerable promise as a means of stabilizing uranium-2 w/o zirconium alloy coextruded with Zircaloy-2 cladding.

As stated earlier, the purpose of the study was to determine the most suitable heat treatment for the extruded and clad fuel alloy. Since the growth rates of the clad specimens were found to be largely insensitive to the heat treatments investigated, the selection of the reference heat treatment for the core was based primarily on considerations arising from other factors, such as the irradiation behavior of the unclad specimens.

prior studies on rolled or swaged materials, and on distortion and dimensional changes occurring during heat treatment of finished fuel elements. The latter factor ruled out the heat treatment which required a quench from 800°C and a subsequent quench from 690°C. Of the remaining two isothermal heat treatments, the one including the 690°C isothermal transformation was selected as the reference heat treatment. The choice was made on the basis of the known low growth rate imparted by tempering at 690°C.

CONCLUSIONS

1. At irradiation temperatures estimated to be near 200°C, extruded uranium-2 w/o zirconium alloy was best stabilized against irradiation growth by heating to 800°C, followed by isothermally transforming at 500°C.
2. At irradiation temperatures estimated to be near 700°C the alloy was best stabilized by heating to 800°C, followed by isothermally transforming at 690°C, or by quenching from 800°C and tempering at 690°C.
3. The average swelling rate of the alloy increased from approximately 8% volume increase per a/o burnup at 200°C to 46% per a/o burnup at 700°C.
4. The presence of coextruded Zircaloy-2 cladding on the fuel alloy provided sufficient restraint to make irradiation growth essentially unaffected by prior heat treatment.
5. The presence of cladding reduces only slightly the swelling rate of the fuel alloy at irradiation temperatures estimated to be near 700°C.
6. Clad material in which the fuel alloy had been subjected to transient melting within the cladding shortened slightly under irradiation and was more resistant to swelling than extruded and heat treated material.

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