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"PIN-CUSHION" IRRADIATION OF CAST URANIUM-PLUTONIUM
ALLOY SPECIMENS

by

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ABSTRACT

The irradiation behavior of small pin specimens of chill-cast U-10 w/o Pu and U-15 w/o Pu specimens projecting from heat transfer "cushions" is described. The specimens were coated with a thin layer of carbonyl-deposited nickel. The 15 w/o Pu pins showed excellent dimensional stability at 1/3 a/o total burnup. The 10 w/o Pu specimens were somewhat inferior, failure of one of them indicating that the improvement in performance over unalloyed uranium may be due to the restraint imposed by the cladding.

A. INTRODUCTION

Preliminary irradiations of uranium-235 containing 10 w/o and 15 w/o plutonium, which were made in 1952-1953, are the subject of this report. At that time attention had been directed to the combination of U^{238} and Pu^{239} as prospective fuel for large fast breeder reactors, and it was desired to know how stable such an alloy might be under neutron irradiation.

The ANL CP-3' reactor had been employed to screen certain other uranium alloys for radiation damage by a technique which became known as the ANL "pin-cushion" test.^(1,2) This method was modified somewhat because of the added hazard due to alphas and was applied to the study of small uranium-plutonium pins cast and coated with nickel at Los Alamos Scientific Laboratory.

Because of the low neutron flux in CP-3' the use of U^{235} was considered advisable to increase the damage rates. Some observations resulting from this initial work have appeared in abbreviated form in the classified literature.^(3,4,5) Collateral work on other plutonium alloys is the subject of a companion report.⁽⁶⁾ These two papers record the modifications of technique required for the early handling of irradiated plutonium and review all the test results. In subsequent investigations the pin-cushion method has been superseded by improved experimental procedures.^(7,8)

B. EXPERIMENTAL PROCEDURE

The fuel pins were produced in the plutonium facility of the Los Alamos Scientific Laboratory by chill casting. After machining to a final size of approximately $1/16$ " diameter, $3/16$ " long, they were given a protective nickel coating by the carbonyl process. The plating was left on the specimens when they were mounted in heat transfer blocks of aluminum for encapsulation. As a further precaution against escape of alpha activity in case the plating failed, the mounting blocks, or "cushions," were provided with tightly fitted aluminum caps, and the bolted assembly of cushions and caps was enclosed in a special hermetically sealed aluminum can containing helium as an atmosphere and heat transfer agent. A cutaway section of the irradiation capsule and its contents is shown in Figure 1. A more detailed description of the encapsulating technique, which had to be repeated after each intermediate examination of the irradiated pins, is given in Appendix A.

The capsule was irradiated in a vertical thimble of CP-3' in a thermal neutron flux of about 2×10^{12} nv. The specimens were examined three times over an irradiation period of 1-1/2 years. The total burnup achieved in this time was approximately 1/3%, averaged over the entire pin. Maximum burnup of surface atoms was probably 2-1/2 times this value. Irradiation data and burnup computations are given in Appendix B.

Visual examination and a photograph record were made of the pins following each irradiation period (see Appendix C). Measurements of the length of each pin from the base to the top were made with a dial gage. At the conclusion of the study, all pins were submitted to the ANL Chemical Engineering Division for fission product analysis of burnup (Table I of Appendix B).

C. DISCUSSION OF RESULTS

Changes in length after all the exposure periods are recorded in Table III Appendix C. In general, they were small and somewhat random in quality. One of the 15 w/o Pu specimens showed a slight tendency to shrink after initial elongation; the other showed a gradual increase. One of the 10 w/o Pu specimens showed essentially null behavior; the appreciable elongation of the other was apparently due entirely to a gross distortion of its tip, which resulted from a failure of the cladding.

A stereophotographic record of the appearance of the specimens after the various exposures is also appended in Figures 3 through 6. Of immediate interest is the 10 w/o Pu specimen which failed (Figures 5 and 6).

Figure 1. Sketch of Irradiation Capsule, Cut Away
to Show Assembly Details.
(Enlarged 1.5X)

After the first period in the reactor and before any surface bumping was apparent, it was noted that there was a well-defined fissure of the nickel coating along one side of the edge of the projecting end. This was opened and stretched by the subsequent exposures and the fuel appeared to extrude through it to a certain degree. On the cylindrical surface of the same specimen may also be seen lesser evidences of gross surface roughening beneath the nickel coating (See Figure 5-c). Careful examination of the other 10 w/o Pu specimen revealed a hint of the same effect. A gap between the aluminum cushion and one side of the pin is to be seen after the second exposure (Figure 3-c), indicating that the pin has departed from its cylindrical shape in this region.

Close examination of the pins containing 15 w/o Pu did not disclose any distinct surface distortions on their sides. However, rounding of the ends appeared more pronounced after irradiation than before.

The behavior of the U-10 w/o Pu alloy specimens is probably similar to what might be expected of coarse-grained, randomly oriented unalloyed uranium clad with a light nickel can. Comparison of the two pins indicated that the nickel coating, while intact, had a surprising degree of strength relative to the contained fuel under the conditions of the irradiation, and was capable of exerting considerable mechanical restraint against surface roughening. In the case of the failed specimen, rupture of the coating and extrusion of the somewhat plastic fuel reduced the hoop stress in the can enough to permit surface bumping to develop more freely than in the specimen which did not fail.

The excellent behavior of the U-15 w/o Pu specimens probably indicates that this composition has a greater degree of stability than U-10 w/o Pu, but the deduction is obviously risky because no information is available concerning the relative degree of restraint produced by the cladding. Rounding of the end surfaces probably indicates that the underlying fuel metal has undergone some uniform plastic deformation.

D. CONCLUSIONS

It is concluded that under the conditions of the test:

1. Chill-cast U-15 w/o Pu alloy shows excellent dimensional stability when coated with a jacket of carbonyl-deposited nickel. It is uncertain, however, whether the improvement over bare, cast uranium is due primarily to the intrinsic characteristics of the fuel material or to the appreciable restraint imposed upon it by its coating.

2. Chill-cast U-10 w/o Pu alloy is probably inferior to the 15 w/o Pu alloy and that it behaves essentially as coarse-grained uranium, enclosed in a strong can, would be expected to.

3. Surface roughening of U-Pu specimens under irradiation may be successfully inhibited by a relatively thin layer of nickel cladding.

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The assistance of the following ANL employees is acknowledged. Initial mounting and encapsulation of the specimens was performed by A. B. Shuck, and post-irradiation operations were done with the help of R. J. Fousek, both of the Metallurgy Division. The cave welding apparatus was set up and operated by C. T. Szymko of the Central Shops Division. Radiochemical analyses of burnup were made by R. P. Larsen, Chemical Engineering Division.

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APPENDIX A

Encapsulation of Specimens

Initial Preparation of Capsule Outside of Cave

The chill-cast pins as received from Los Alamos were coated with a thin layer of nickel deposited by the carbonyl process, making their size somewhat greater than the machined dimensions ($1/16"$ x $3/16"$). They were prepared for irradiation in a glovebox under a slight negative pressure maintained by a small blower discharging through an SC-7 filter to a standard hood stack. Pins were mounted in the end surfaces of two 2S aluminum cylinders, 0.800" diameter by 1" long. One of these holders is shown in Figure 2. Mounting holes were drilled 0.002" - 0.003" larger than pin diameter and 0.045" less than the length of the specimen in depth. Metal around the mounted pin was upset by means of a hollow conical punch, thus insuring good contact for heat transfer.

Figure 2. Photograph of Aluminum Heat Transfer Block with Specimen Pins in Place.

M-12631

2X

As a precaution against oxidation and dispersion of the fuel material during irradiation, each of the pin cushions was covered by an aluminum cap, fitted closely by means of a tongue and 60° V groove made circumferentially on the top of the cushion. Caps and cushions were held firmly together by a central tie bolt passed through their axis after stacking them. One end of the tie bolt was anchored to a bottom disc of steel, and the top end was threaded to receive a nut. This assembly was placed

inside an outer can of aluminum and hermetically sealed by heliarc welding after evacuation and introduction of a helium atmosphere (see Figure 1).

Handling of Capsule in Cave - Decanning and Canning

Decanning and disassembly of the capsule by remote procedures in a cave after irradiation proved to be quite simple. The aluminum can was opened with a hack saw, after which the tie bolt holding the cushions together was removed by means of a nut driver. The capsule then was immersed in carbon tetrachloride as a precaution against spread of contamination in the event of specimen disintegration, and the protection cup was pried off with a screw driver.

This procedure was used without special difficulty for all three decanning operations. Re-assembly of the cushions and caps for further irradiation also proved to be a simple task. They were easily placed on the central tie bolt and the single nut was readily tightened with a nut driver. Sealing the bolted assembly within the aluminum outer can, however, was a somewhat more complicated and time-consuming task.

The outer can had been made from a section of aluminum tube which was closed at the ends by close fitting aluminum plugs. One of these, terminating in a handling or grappling knob, was welded to the tube and the weld was leak tested before commencement of the loading operation. The plug at the other end was drilled and tapped to serve also as an evacuating stem and screw-set valve, and it was necessary to weld it to the aluminum tube in the cave remotely after insertion of the bolted pin cushion assembly.

This was accomplished by placing the assembled capsule in a massive heat-absorption block mounted on the magnetic chuck of a motorized turntable set up within the cave. The rotation of this device (approximately 5 rpm), when properly aligned, placed the edge to be welded in proper reference to a mounted heliarc welding torch. The latter was equipped with automatic starting and stopping devices mechanically indexed and controlled from outside the cave. After welding of the evacuating plug into the end of the can the torch head was moved out of the way and a vacuum line was attached to the stem through a special brass adapter. This device slipped over the evacuating stem and held an Allen wrench and aluminum conical point set screw in axial alignment for temporary gas-tight closure of the capsule. Slip-fit surfaces of the adapter were made vacuum-tight by the use of stop-cock grease on wrench handle and capsule stem. The attached vacuum line was passed outside the cave, where the vacuum pump and helium supply were located. Pump discharge was made to the cave interior.

After careful leak testing of the assembly weld previously made, the capsule was finally evacuated, filled with helium, and closed tightly by insertion of the conical screw. The vacuum adapter was then removed and the end of the evacuating stem and the conical screw were finally fused together by the heliarc torch.

APPENDIX B

Irradiation and Burnup Analysis Data

All four pin specimens were analyzed radiochemically for burnup. The reported values are shown in Table I. Percentage error is estimated to be $\pm 5\%$. The over-all average of 0.33 a/o burnup was used in further computations.

TABLE I

Determination of Burnup by Cs¹³⁷ Radiochemical Analysis

Cushion No.	Pin No.	Composition, w/o Pu	Average Total Atom Burnup, a/o	Cushion No.	Pin No.	Composition, w/o Pu	Average Total Atom Burnup, a/o
Pu-4	B	10	0.31	Pu-4	A	15	0.34
Pu-5	B	10	0.35	Pu-5	A	15	0.31

Neutron exposure data are recorded in Table II, together with estimated cumulative surface depletions computed therefrom, and assigned average total atom burnup values for each exposure. The estimated surface depletions have been computed without any adjustment for flux depression. They are therefore appreciably higher than they should be; however, the tabulated values provide a reasonable relative system which has been used in assignment of total atom burnups to the intermediate exposures.

TABLE II

Irradiation Data, Surface Depletion of Fissionable Atoms,
and Average Total Atom Burnup

Exposure No.	Request No.	Irradiation Dates		Exposure Data			Estimated Per Cent Cumulative Surface Depletions ^b			Estimated Per Cent Cumulative Average Burnup
				Nominal Flux (10 ¹² nv)	Equivalent Time at 275 kw ^a (10 ⁶ sec)	Nominal Flux-Time (10 ¹⁸ nvt)	U ²³⁵ Atoms	Pu ²³⁹ Atoms	Total Atoms ^c	
		In	Out							
1	5229	8-18-52	10-27-52	1.30	3.52	4.58	0.23	0.29	0.24	0.07
2	5284	11-10-52	11-24-52	2.04	0.86	10.96	0.55	0.69	0.57	0.18
		11-24-52	2- 9-53	1.30	3.56					
3	5386	6-29-53	11- 9-53	1.39	6.99	20.67	1.03	1.30	1.07	0.33

^aComputed from CP-3' power log.

^bBased on nominal flux-time values and fission cross sections of 500 and 630 barns for U²³⁵ and Pu²³⁹, respectively.

^cApplicable to both 10 and 15 w/o Pu alloys within a relative deviation of approximately 1%.

APPENDIX C

Post-Irradiation Observations

Observed per cent length changes of the pins after each exposure are listed in Table III. They are based on the total length of the pins (3/16") and do not take into account any restraints involved in the mounting method.

TABLE III

Per Cent Changes in Length of U-Pu Pins
after Exposures in CP-3' Reactor

Cushion No.	Pin No.	Composition, w/o Pu	% Change in Length after Irradiation Exposures ^a		
			First Exposure	Second Exposure	Third Exposure
Pu-4	B	10	-0.3	0.4	-0.6
Pu-5	B	10	1.8	2.3	7.5
Pu-4	A	15	1.4	0.9	-0.5
Pu-5	A	15	1.6	1.7	2.1

^a Apparent changes in reference pin lengths ranged from -0.9% to 0.5%. Tabulated data are corrected for this variance.

Stereophotographs of the cushions and their pins are shown in Figures 3 through 6. The views in Figures 3 and 5 were obtained through the ANL-designed stereobinocular; those in Figures 4 and 6 were made with the Bausch and Lomb stereobinocular periscope.

Figure 3. Appearance of Cushion Pu-4: (a) Before Irradiation, (b) After 0.07 a/o Average Burnup, and (c) After 0.18 a/o Average Burnup.

a) 15 w/o
Pu

13107

Zr

5X

b) 10 w/o
Pu

13312

15 w/o
Pu

5X

c)

Zr

13839

10 w/o
Pu

15 w/o
Pu

5X

Figure 4. Appearance of Cushion Pu-4 and Free Ends of Its Fuel Pins After 0.33 a/o Average Burnup.

14

a)

10 w/o
Pu

18031

15 w/o
Pu

Zr

4X

b)

15 w/o
Pu

c)

10 w/o
Pu

18032, 18033

34X

Figure 5. Appearance of Cushion Pu-5: (a) Before Irradiation, and (b and c) After 0.18 a/o Average Burnup.

a) 15 w/o
Pu

10 w/o
Pu

13108

Zr

5X

b) 15 w/o
Pu

10 w/o
Pu

13840

5X

c)

10 w/o
Pu

13841

13X

Figure 6. Appearance of Cushion Pu-5 and Free Ends of
Its Fuel Pins After 0.33 a/o Average Burnup.

16

a)

10 w/o
Pu

18035

15 w/o
Pu

Zr

4X

b)

15 w/o
Pu

c)

10 w/o
Pu

18036, 18037

34X