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SWELLING OF ALUMINUM-CLAD ALUMINUM-PLUTONIUM ALLOYS ON POSTIRRADIATION ANNEALING

by

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Metallurgy Division

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TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

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ABSTRACT

The changes in density and hardness as a result of annealing at temperatures up to 600°C are reported for an aluminum-clad Al-5 w/o Pu and Al-10 w/o Pu alloy in which 89.3% and 75.6%, respectively, of the plutonium atoms were fissioned. The density of the irradiated specimens at room temperature decreased after pulse anneals of 15-min duration at temperatures above 525°C, whereas for lower annealing temperatures the density of the specimens increased. The hardness of the specimens at room temperature decreased substantially after annealing at temperatures between 275°C and 360°C.

A specimen of 1100 aluminum which was heavily deformed by rolling was also annealed in a manner similar to the irradiated materials. The density of the aluminum at room temperature increased after pulse anneals at temperatures up to 600°C, and the hardness decreased after annealing at temperatures above 300°C.

INTRODUCTION

During the course of a general investigation on the swelling of fissionable materials, aluminum-clad Al-5 w/o Pu and Al-10 w/o Pu alloys in which a substantial fraction of the plutonium atoms were fissioned became available for study. Since the specimens contained an appreciable quantity of gaseous fission products, it is of interest to determine their swelling behavior at elevated temperatures. This report describes the density changes and hardness of these specimens and of a rolled 1100 aluminum specimen on pulse annealing at temperatures between 125°C and 600°C.

EXPERIMENTAL MATERIALS AND PROCEDURE

The casting and fabrication of the Al-5 w/o Pu and the Al-10 w/o Pu alloys have been described previously.(1) The specimens were in the form of split cylinders, $1\frac{1}{4}$ in. in diameter and 2 in. long. The core alloy was

0.020 in. thick, clad on both sides by 0.020 in. of bonded-on 1100 aluminum and edge-sealed by an 1100 aluminum frame. The alloy in the Al-5 w/o Pu specimen weighed 3.342 gm and had a volume of 1.229 cc, and the alloy in the Al-10 w/o Pu specimen weighed 3.472 gm and had a volume of 1.163 cc.

The specimens were irradiated in the Materials Testing Reactor, and it is estimated that their central metal temperature was less than 100°C during irradiation. The Al-5 w/o Pu alloy received a thermal neutron exposure of 11.97×10^{21} nvt, and the Al-10 w/o Pu alloy received an exposure of 2.14×10^{21} nvt.

Since the specimens were in direct contact with the reactor cooling water, they were coated with a thin film of oxide after irradiation (see Figure 1). The oxide coating was removed by dissolution in boiling water containing 2% chromic acid and 5% orthophosphoric acid by volume. Visual examination of the specimens indicated that the only effect of irradiation was to deform the specimens to a slightly oval shape.

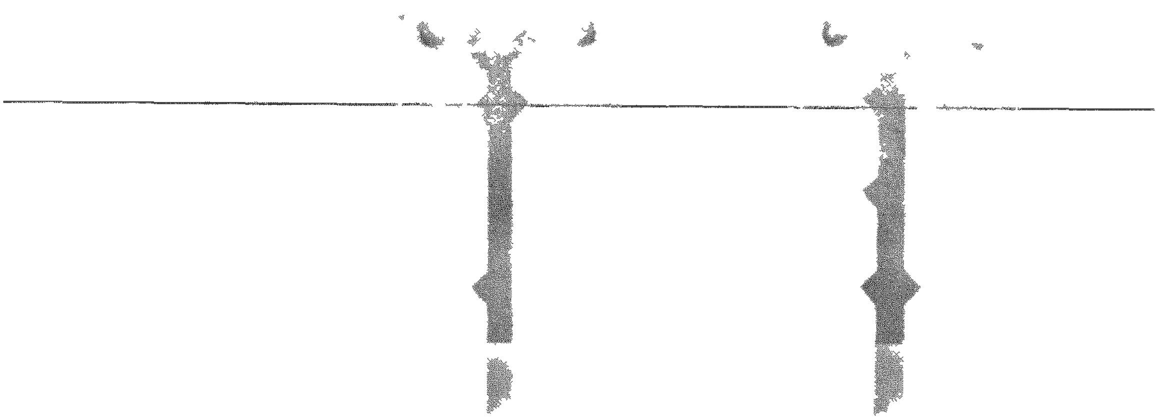

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Figure 1. Appearance of Aluminum-clad Aluminum-Plutonium Alloy Specimens after Irradiation

The specimens were immersed in a commercial heat-treating liquid for annealing temperatures between 125°C and 250°C. For annealing temperatures between 250°C and 500°C, the specimens were immersed in molten sodium nitrate-potassium nitrate, and for annealing temperatures between 500°C and 600°C the specimens were heated in a closed container containing an argon atmosphere.

The annealing temperature was determined with a chromel-alumel thermocouple placed adjacent to the specimen, and the temperature was controlled to ±1°C during annealing. The specimens were annealed 15 min at temperature.

The specimens were cleaned upon removal from the heat-treating medium, and the densities of the alloy core and cladding at room temperature were determined by weighing the specimens in air and carbon tetrachloride.

The hardnesses of the specimens at room temperature were also determined after several of the annealing treatments by use of a Rockwell Hardness Tester with a ball indenter of $\frac{1}{8}$-in. diameter and 60-kg load. The measurements gave the hardness of the composite specimen, i.e., core and cladding.

The irradiated aluminum-plutonium alloys were dissolved after the annealing studies. From a mass spectrographic analysis of the plutonium isotopes in the solutions, it is concluded that 89.3% of the plutonium atoms in the Al-5 w/o Pu alloy and 75.6% of the plutonium atoms in the Al-10 w/o Pu alloy were fissioned. These percentages of fissioned plutonium atoms show necessarily that a sizeable fraction of the Pu^{241} atoms, which are formed by neutron capture in Pu^{239} and Pu^{240} atoms, were fissioned.

A specimen of 1100 aluminum which was heavily deformed by rolling was also annealed in a manner similar to that for the irradiated aluminum-plutonium alloys, and density and hardness measurements were likewise obtained after the annealing treatments.

EXPERIMENTAL RESULTS

The changes in density of the irradiated aluminum-clad aluminum-plutonium specimens and the deformed aluminum specimen on pulse annealing 15 min at successively higher temperatures are shown in Figure 2, and the data are listed in Tables I-III. The room-temperature hardness values of the specimens after pulse anneals at elevated temperatures are shown in Figure 3, and the data are listed in the same tables.

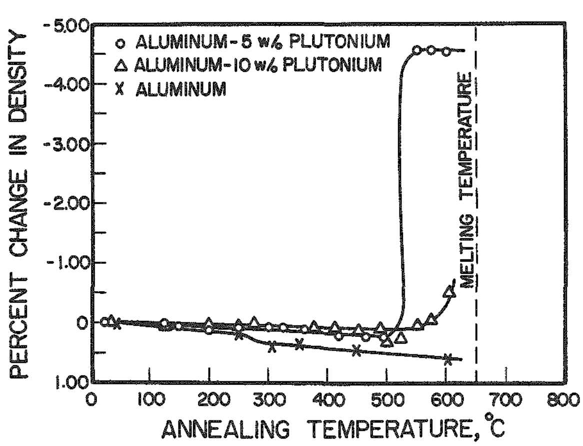

Figure 2

Change in Density of Irradiated Aluminum-clad Al-5 w/o Pu and Al-10 w/o Pu Alloys and of Rolled Aluminum on Annealing 15 Min at Successively Higher Temperatures.

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Table I

DATA FOR THE ALUMINUM-CLAD Al-5 w/o Pu ALLOY IRRADIATED TO 11.97×10^{21} nvt (89.3% Pu ATOMS FISSIONED) AND THEN ANNEALED 15 MIN AT VARIOUS TEMPERATURES

Annealing Temperature (°C)	Weight (gm)	Volume (cc)	Density (gm/cc)	Change in Density[(1)] (%)	Hardness (R_H)
Preirradiation	20.354	7.480	2.721		
Postirradiation	19.708[(2)]	7.281	2.707		61[(3)]
125	19.710	7.281	2.707	0	62
150	19.708	7.278	2.708	+0.04	59
200	19.708	7.272	2.710	+0.11	
250	19.707	7.275	2.709	+0.07	68
300	19.706	7.274	2.709	+0.07	57
325	19.705	7.274	2.709	+0.07	
360	19.706	7.271	2.710	+0.11	10
420	19.706	7.265	2.712	+0.19	
465	19.708	7.265	2.713	+0.22	15
496	19.704	7.260	2.714	+0.26	1
550	19.710	7.630	2.583	-4.58	15
575	19.702	7.628	2.583	-4.58	15
598	19.706	7.625	2.584	-4.54	16

(1)The per cent change in density is expressed as the ratio of the change in density on annealing to the density of the specimen after irradiation.

(2)This weight change is partially due to removal of an oxide film.

(3)This value is the average of three or more determinations.

Table II

DATA FOR THE ALUMINUM-CLAD Al-10 w/o Pu ALLOY IRRADIATED TO 2.14×10^{21} nvt (75.6% Pu ATOMS FISSIONED) AND THEN ANNEALED 15 MIN AT VARIOUS TEMPERATURES

Annealing Temperature (°C)	Weight (gm)	Volume (cc)	Density (gm/cc)	Change in Density[(1)] (%)	Hardness (R_H)
Preirradiation	20.670	7.569	2.731		
Postirradiation	20.599[(2)]	7.572	2.720		52[(3)]
125	20.599	7.571	2.721	+0.04	
150	20.592	7.570	2.720	0	59
200	20.592	7.567	2.721	+0.04	
250	20.590	7.568	2.721	+0.04	53
275	20.588	7.567	2.721	+0.04	15
325	20.588	7.568	2.721	+0.04	
375	20.586	7.562	2.722	+0.07	15
410	20.586	7.561	2.723	+0.11	15
453	20.586	7.560	2.723	+0.11	
487	20.586	7.561	2.723	+0.11	
499	20.588	7.544	2.729	+0.33	15
521	20.590	7.554	2.726	+0.22	
550	20.587	7.566	2.721	+0.04	
574	20.588	7.572	2.719	-0.04	
605	20.589	7.607	2.707	-0.48	15

(1)The per cent change in density is expressed as the ratio of the change in density on annealing to the density of the specimen after irradiation.

(2)This weight change is partially due to removal of an oxide film.

(3)This value is the average of three or more determinations.

Table III

DATA FOR 1100 ALUMINUM ROLLED AND THEN ANNEALED 15 MIN AT VARIOUS TEMPERATURES

Annealing Temperature (°C)	Weight (gm)	Volume (cc)	Density (gm/cc)	Change in Density (%)	Hardness (R_H)
As-received	25.986	9.619	2.702		106[1]
250	25.985	9.603	2.706	+0.15	106
302	25.986	9.582	2.712	+0.37	92
354	25.986	9.586	2.711	+0.33	71
450	25.986	9.580	2.713	+0.41	52
600	25.987	9.562	2.718	+0.59	22

[1] Value is the average of three or more determinations.

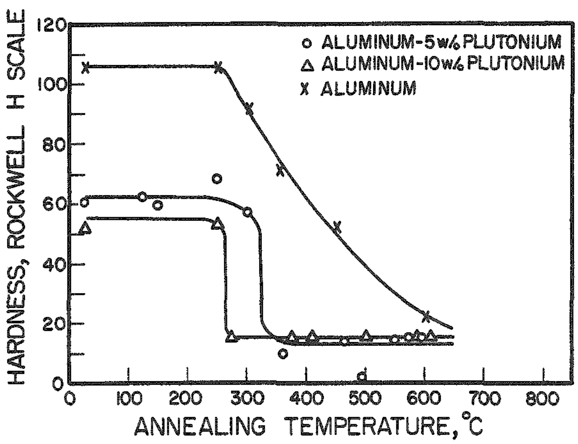

Figure 3

Change in the Room-temperature Hardness of Irradiated Aluminum-clad Al-5 w/o Pu and Al-10 w/o Pu Alloys and of Rolled Aluminum after Annealing 15 Min at Successively Higher Temperatures

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A visual examination of the irradiated specimens following the annealing studies revealed no perceptible change in the appearance of the specimens.

It is apparent from the curves in Figure 2 that a pronounced decrease in density of the irradiated aluminum-clad aluminum-plutonium alloys occurred on annealing at temperatures above 525°C. However, the density of the irradiated specimens increased on annealing at temperatures up to 525°C. The density of the rolled aluminum increased on annealing at temperatures up to 600°C.

The hardness of the irradiated aluminum-clad aluminum-plutonium specimens decreased substantially after annealing at temperatures between 275 and 360°C, and the hardness of the rolled aluminum decreased after annealing at temperatures above 300°C (see Figure 3).

DISCUSSION OF RESULTS

A significant result obtained by this investigation was the determination of a lack of swelling of the irradiated aluminum-plutonium alloys until they were annealed at temperatures within about 125°C of the solidus temperature of 1100 aluminum,* even though the specimens probably contained substantial quantities of krypton and xenon. It is estimated that at the conclusion of the irradiation, the Al-5 w/o Pu specimen contained 9.3×10^{19} krypton and xenon atoms in 1.23 cc of alloy, and the Al-10 w/o Pu specimen contained 1.6×10^{20} krypton and xenon atoms in 1.16 cc of alloy. The numbers of gas atoms in the specimens were calculated from experimental data which show that 24.75 krypton and xenon atoms are produced on the fission of 100 Pu^{239} atoms.(3) If the gas atoms in the Al-5 w/o Pu and the Al-10 w/o Pu specimens were contained in a single enclosed volume, they would occupy 3.5 cc and 5.9 cc (STP), respectively. Therefore, since the solubility of the inert gases in metals is negligible and the density of the irradiated specimens increased on annealing up to 525°C, the gas atoms were probably contained at a high pressure in small bubbles (of less than 0.05-μ diameter). The absence of a pronounced temperature dependence of swelling at temperatures below 525°C could then be accounted for by a large surface tension restraining pressure P determined by the relationship

$$P = 2\gamma/r \quad ,$$

where γ represents the surface tension of the bubble/matrix interface, and r the radius of the bubble. The surface tension is not expected to have a substantial temperature dependence.

The pronounced increase in the amount of swelling at annealing temperatures above 525°C may be due to the gas pressure in bubbles exceeding the yield strength of the matrix or to microstructural changes such as recrystallization or grain growth. It has been reported that a pronounced increase in the swelling of uranium occurs on recrystallization of the matrix during postirradiation annealing.(4)

It is not clear why the Al-5 w/o Pu specimen swelled more than the Al-10 w/o Pu specimen, since the latter specimen contained about twice as much fission product gas. It is perhaps significant that 10 w/o Pu in aluminum is near the eutectic composition in this alloy system.(5) Therefore, the plutonium in the Al-10 w/o Pu alloy would be more uniformly distributed in the aluminum matrix than in the Al-5 w/o Pu alloy. A more uniform distribution of fissionable material would result in gas-filled bubbles of a smaller diameter.

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*The solidus temperature of 1100 aluminum is 643°C.(2)

The krypton and xenon atoms produced in the specimens were probably retained during irradiation and annealing since the diffusion rate of inert gas atoms is low[6] and no cracks were visible in the aluminum cladding.

The decrease in hardness and the increase in density of the irradiated aluminum-clad aluminum-plutonium specimens and the rolled aluminum specimen suggest that defects of a similar nature in the irradiated and deformed materials commence to anneal at an appreciable rate at about 300°C.

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REFERENCES

1. Shuck, A. B., The Manufacture of Aluminum Clad Aluminum-Plutonium Alloy Irradiation Specimens, ANL-5043 (1952).

2. Metals Handbook Committee, Metals Handbook, 810 (1948).

3. Lewis, W. B., The Return of Escaped Fission Product Gases to UO_2, DM-58 (1960).

4. Loomis, B. A., and Pracht, D. W., Swelling of Uranium and Uranium Alloys on Postirradiation Annealing, ANL-6532 (1962).

5. Schonfield, F. W., "Plutonium Constitutional Diagrams," Progress in Nuclear Energy, Series V2, 579, Pergamon Press (1959).

6. Reynolds, M. B., Fission Gas Behavior in the Uranium Aluminum System, Nuclear Sci. and Eng., 3, 428 (1958).