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METALLURGICAL INVESTIGATION OF ALUMINUM-CLAD CADMIUM CONTROL RODS

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METALLURGICAL INVESTIGATION OF ALUMINUM-CLAD CADMIUM CONTROL RODS

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ABSTRACT

Cadmium (mp 321°C) clad with aluminum was used satisfactorily for control rods for a water-cooled nuclear reactor operating at a neutron flux of $\sim 10^{15}$ n/(cm^2)(sec). Laboratory tests, to assure the integrity of this combination, had shown that aluminum is attacked very slowly by molten cadmium at 400-600°C under conditions of static stress and also under continuous strain. The aluminum cladding of two control rods used in operation at high flux was not attacked even though the cadmium had partially melted in one rod during reactor operation.

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INTRODUCTION

In the high-flux demonstration lattice[1] developed for a Savannah River production reactor, aluminum-clad cadmium control rods (Figure 1) were used to provide the necessary neutron absorption and yet operate within the capacity of the rod-cooling system.

Cadmium was chosen over lithium-aluminum for the high-flux charges. In the lithium-aluminum rod, heat is generated by the $^{6}Li\ (n,\alpha)$ reaction; the energy of this reaction is dissipated entirely within the rod, and the heat removal capacity of the control rod cooling system would be exceeded. Only about 20% of the emission energy is retained in the cadmium rod because cadmium reacts by an (n,γ) reaction.

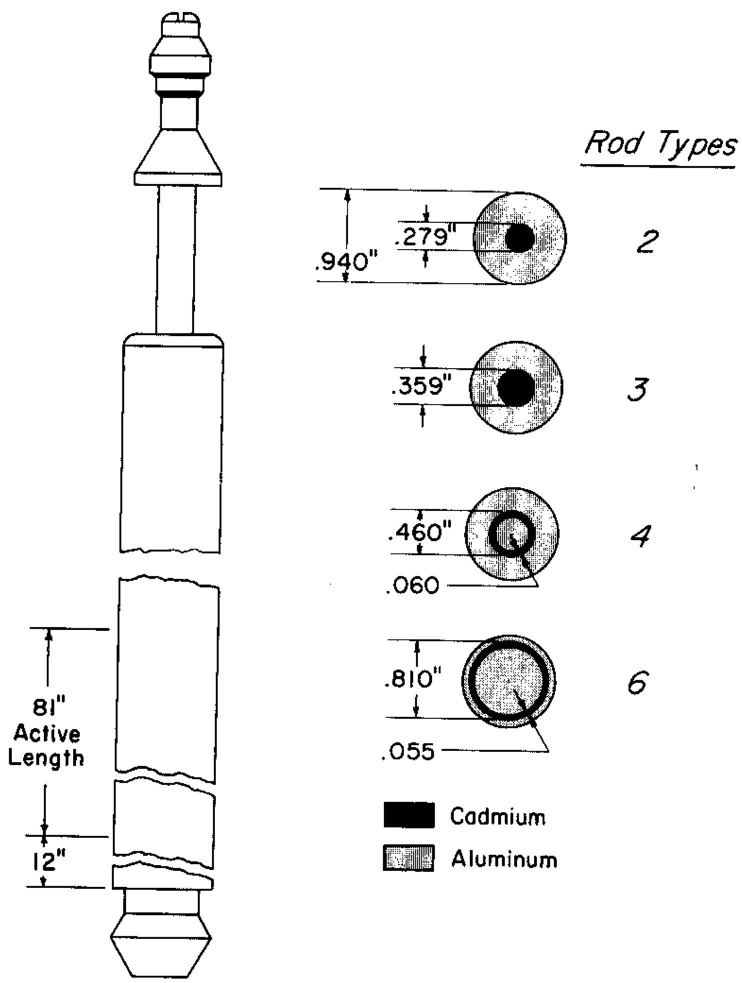


FIG. 1 CADMIUM CONTROL RODS

Because cadmium is almost black to thermal neutrons, neutron absorption is determined by the diameter of the cadmium. Calculations showed the diameter of solid cadmium rods was limited to <0.45 inch to prevent melting (321°C) in the highest flux regions. If the cadmium should melt, penetrate the aluminum sheath, and escape from the control rod, the loss of the cadmium core would cause a rapid increase in reactor reactivity. Solid cadmium control rods were clad by swaging extruded-and-drawn aluminum tubes over the cadmium rods. Rods >0.45 inch in diameter were prepared by plating cadmium on aluminum rods, and then swaging the extruded-and-drawn aluminum tube over the plated rods.

Calculations predicted that if the gap around the cadmium exceeded about 1.5 mils, the thermal resistance of the gap would cause the cadmium to melt. However, the molten cadmium should solidify immediately after touching the aluminum cladding since the maximum cladding temperature was calculated to be 160°C.

Aluminum is a very reactive metal, but is normally protected by a thin film of oxide. To react with aluminum metal, the reactants must first penetrate the oxide film by diffusion, dissolution, or mechanical rupture. Aluminum oxide films will probably not react with molten cadmium because of the high positive value of ΔF^{o} (+215 kcal/mole) for the reaction:

$$3\ Cd(l) + Al_2O_3(s) \longrightarrow 3\ CdO(s) + 2\ Al(s)$$

At 400°C, ΔF is +207 kcal/mole of Al_2O_3. Therefore, if the core melted, the aluminum sheath of a control rod would not be exposed to liquid cadmium unless the natural aluminum oxide film is ruptured by some mechanism other than reactions with cadmium, such as thermal stress, mechanical abrasion, or creep.

Little is known about the reaction between solid aluminum and liquid cadmium. The Liquid Metals Handbook[2] states that resistance of aluminum to cadmium is low at its melting point, and that solubilities are believed to be small. Rostoker, et al.[3] mentioned that cadmium did not embrittle aluminum alloys. This is an exception to the rule that low miscibility often results in embrittlement.

The phase diagram for the aluminum-cadmium system shows that the solubility of cadmium in aluminum is very low, <1% at 500°C (Figure 2[4]). Since the dissolution of cadmium in aluminum is small, attack, if it does occur, would be expected along grain boundaries. Because of the minimal amount of specific data, the reaction between aluminum and molten cadmium was investigated to determine experimentally the predicted reliability of aluminum-clad control rods.

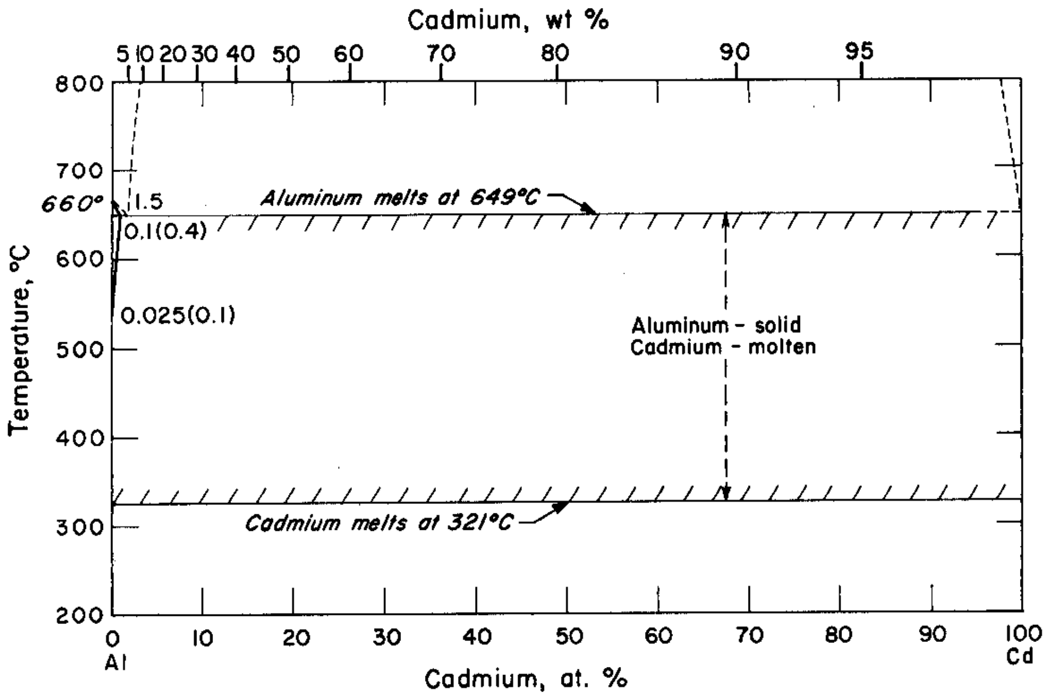


FIG. 2 ALUMINUM-CADMIUM PHASE DIAGRAM

SUMMARY

A variety of test exposures of aluminum to molten cadmium showed neither rapid nor severe attack. Reactor conditions would be very much less severe than those tested, so good reliability of aluminum-clad cadmium control rods was predicted with confidence even if the cadmium should melt.

The penetration of aluminum by molten cadmium was 3 to 7 mils/day in statically loaded U-bend specimens at 400°C, and 8 mils/day in continuously deformed specimens at 400°C. In continuously deformed specimens at 600°C, the penetration of cadmium was only 11 mils/day. The aluminum was not embrittled by molten cadmium. An 8-day period of alternately heating at 400°C and quenching changed the diameter of test control rods less than 1%.

Postirradiation examination showed that cadmium had melted and resolidified in one rod that was inadvertently inserted in the reactor for a full cycle; as predicted, the cadmium did not attack the aluminum cladding. Control rods that received the highest nominal exposure showed no discernible effects from the irradiation.

DISCUSSION

STATIC STRESS TESTS

To determine the extent of cadmium attack on aluminum, seven U-bend specimens of 1100 aluminum were immersed in cadmium at 400°C. While immersed, the samples were abraded with a hacksaw blade to remove some of the protective oxide film in the area of maximum stress. After immersion, samples were then stressed again. At one-hour intervals, one sample was removed; the last sample was exposed for 7 hours.

All seven U-bends were sectioned and examined metallographically to determine cadmium penetration. Figure 3 illustrates the microstructure in an area where the aluminum oxide film had been ruptured, and shows the deepest penetration in the 7-hour-test sample. Slight penetration along grain boundaries is shown. The average depth of penetration for this sample was 1 to 2 mils at irregular intervals.

Note that cadmium had penetrated only 4 mils after 7 hours exposure at 400°C. Penetration along grain boundaries occurs at approximately the same rate as the local dissolution of the grains.

FIG. 3 LIQUID CADMIUM ATTACK OF ALUMINUM U-BEND SPECIMEN (250X)

CONTINUOUS STRAIN TESTS

Three test control rods, 19.5 inches long, were fabricated, installed in a creep-test machine, and heated between 400-600°C to melt the cadmium core. The elongation produced by the creep deformation continuously ruptured any protective aluminum oxide film on the interior surfaces of the control rod, and assured maximum reaction of aluminum with liquid cadmium. The tests lasted about 6 days; results are shown below.

Control Rod Elongation During Creep Test

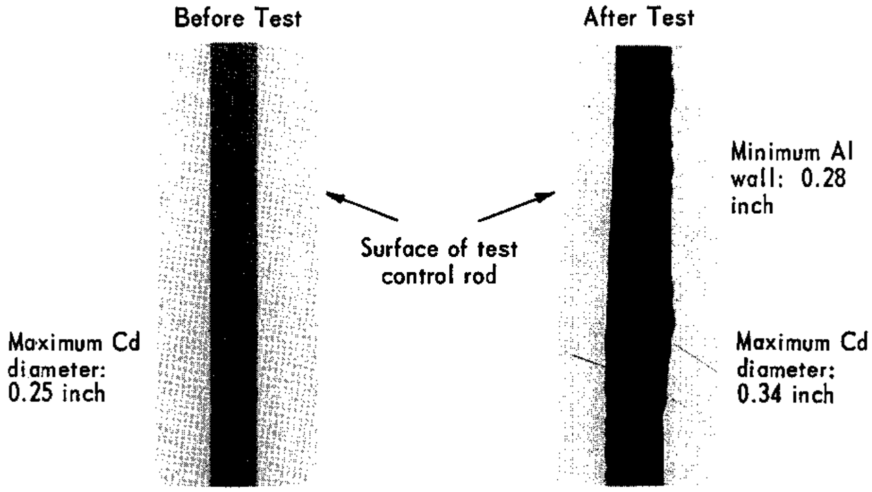
Rod No.	Cd Diameter, inch	Temp, °C	Stress, lb/in²	Elongation, % in 6 days
1	0.25	600	60	0.65
2	0.25	500	140	0.16
3	0.47	400	210	0.27

The most severe attack of the aluminum sheath occurred after 6 days at 600°C, during which the cadmium reached a maximum radial penetration of about 65 mils. This penetration corresponded to an increase in cadmium diameter from 0.25 to 0.34 inch as shown in Figure 4a. Microscopic examination of the area of maximum penetration indicated slow penetration by the cadmium as shown by the rounded ends at attack sites down grain boundaries, Figure 4b. The presence of aluminum dendrites within the cadmium showed that some of the aluminum had dissolved. Embrittlement of the aluminum was not observed.

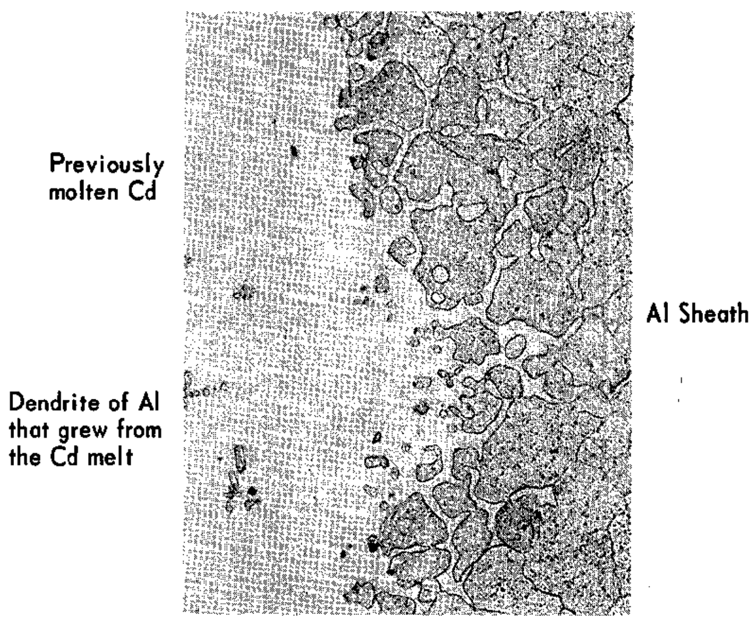
A radiograph of the rod (Figure 5) proved that the molten cadmium had filled the voids created by assembly clearances and had penetrated between the end plug and sheath wall during the continuous strain tests.

The continuous strain tests compared favorably with the static stress tests. The average rate of penetration into the U-bends was 3 to 7 mils/day at 400°C; the rate into test control rod cladding subjected to continuous strain was 8 mils/day at 400°C and 11 mils/day at 600°C. The continuous deformation of the aluminum increased the rate of attack, but even the highest rate was extremely slow for the proposed application in control rods.

If cadmium melted in a control rod, it would freeze almost immediately at any point of contact with the aluminum cladding, and the time for penetration of the cladding would be very short. The maximum temperature of the cladding in reactor service was calculated to be 160°C. The central portion of the cadmium core might remain molten for a longer time, but would be separated from the aluminum cladding by a frozen crust of cadmium.

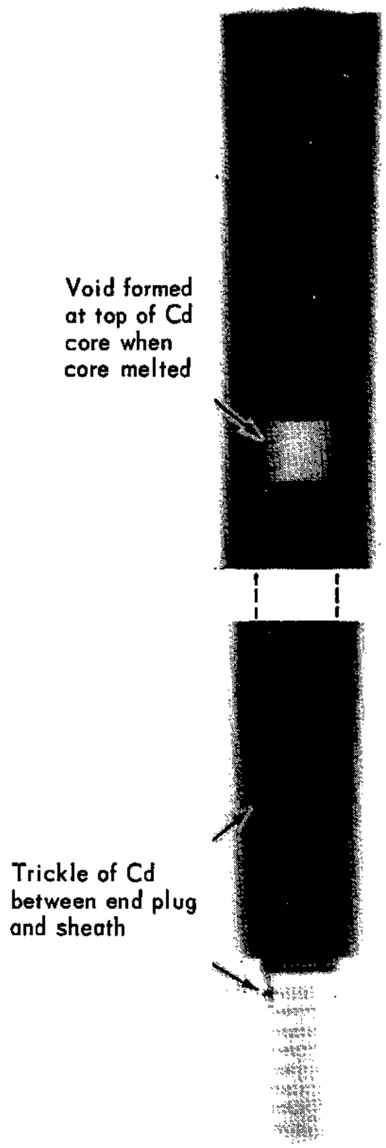


a. Radiographs showing maximum Cd penetration during creep test for 142 hours at 600°C (1X).



b. At 600°C, intergranular nature of the attack is evident in region of maximum penetration (100X).

FIG. 4 LIQUID CADMIUM ATTACK OF ALUMINUM CREEP TEST SPECIMEN



Note that cadmium filled the assembly clearance spaces and penetrated between end plug and cladding. (1X)

FIG. 5 RADIOGRAPH OF CADMIUM-FILLED CREEP TEST SPECIMEN

THERMAL CYCLING TESTS

Three test control rods, 0.940-inch OD by 19.5 inches long, were supported vertically in 12-inch-long furnaces at 400°C. Once a day the samples were water quenched and the OD measured in three positions, two in the heated area and one outside. Two of the rods were heated for about 8 days (198 hours). The test on the third rod was terminated after 47 hours because the furnace temperature reached 500°C between 30 and 47 hours because of a faulty controller.

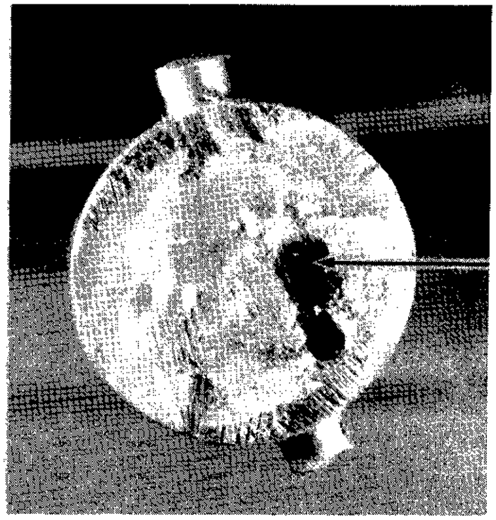
Dimensional changes of the rods did not follow a consistent pattern. The OD of 0.940 inch generally varied by ±3 mils. The OD increased by a maximum of 8 mils, or less than 1%, after being heated and quenched for 8 days.

After the first 5 hours, a brown solid was observed on the weld at one end of the rod that had been heated for 47 hours. The solid became enlarged, was removed, and was identified as cadmium covered with CdO. Metallographic examination showed that the cadmium had penetrated the gap between the cladding and end plug, and then leaked through a porous aluminum weld as shown in Figure 6. Radiographs of the other two rods showed that the cadmium had penetrated between the cladding and the end plug as in the continuous strain tests.

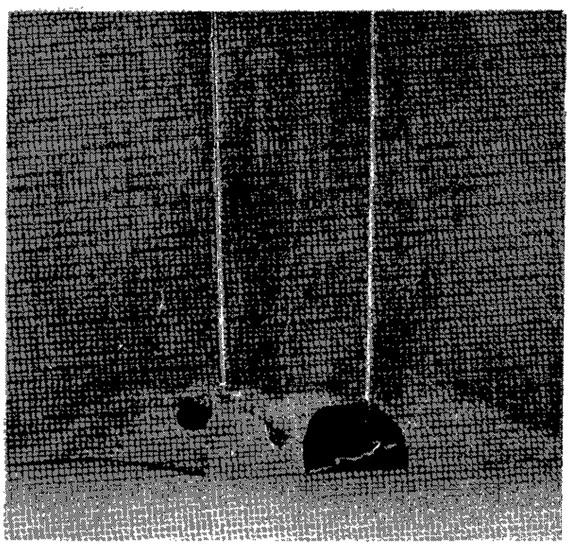
In actual operation, the temperature of the 1-inch-long end plug would be sufficiently below the 321°C freezing point of cadmium to ensure that no cadmium could escape, even through a defective weld.

CONTROL ROD CHARACTERISTICS

Cross sections of the rods used in the first high-flux charges are shown in Figure 1. The only gap found during pre-irradiation examination of the control rods was 0.09 mil wide. Because the thermal expansion of cadmium is greater than that of aluminum, even this gap would decrease in size, due to heating during irradiation. Both the type 2 (solid Cd rod = 0.279-inch diameter) and type 6 rods (Cd electroplate = 0.055 inch thick) were examined after irradiation.



a. CdO deposit on end of test control rod after 5 hours at 400°C (2X).



b. Longitudinal section of rod through end plug and cladding. CdO deposit in weld, high porosity throughout weld (5X).

c. Enlargement of top of CdO in weld showing Cd penetration between end plug and cladding (100X).

FIG. 6 PENETRATION OF POROUS WELD BY MOLTEN CADMIUM DURING THERMAL CYCLING TESTS

POSTIRRADIATION EXAMINATION OF CONTROL ROD WITH SOLID CADMIUM CORE

An examination was made of a type 2 control rod (Cd = 0.279-inch diameter) that had accumulated the highest exposure [flux level = 3.4×10^{15} n/(cm^2)(sec)] in the first high-flux cycle. A transverse section of the rod is shown in Figure 7. The folds

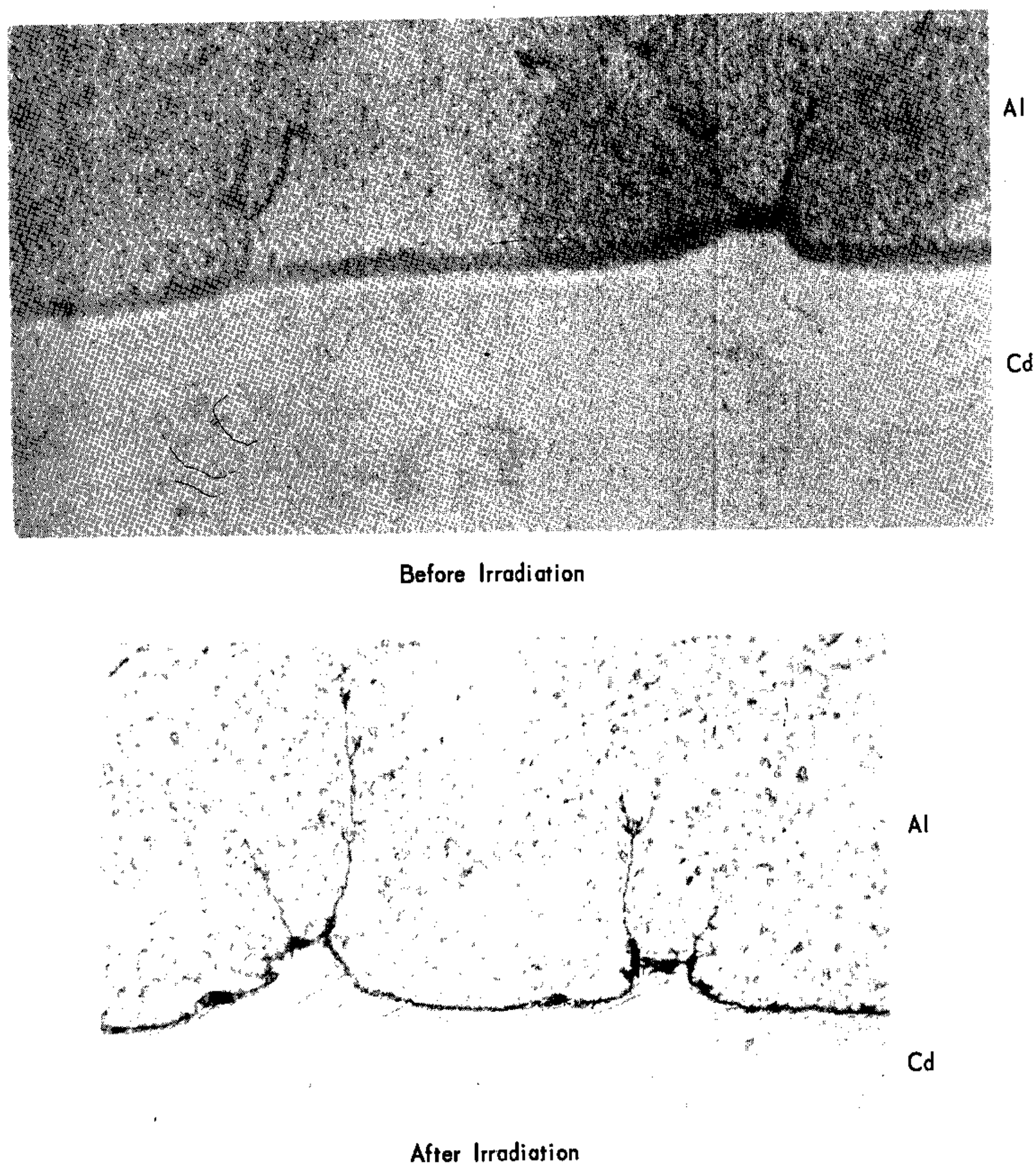


FIG. 7 TYPE 2 CADMIUM CONTROL ROD INTERFACES (500X)

in the aluminum cladding were due to a sinking operation performed during the manufacture of the tubing. The aluminum tubing was reduced in diameter by drawing it through a die without using a mandrel to control the inside diameter. After a mandrel was used, the folds were eliminated.

Diameter measurements and metallographic examination of the rod showed no evidence of either distortion, cadmium melting, or attack on aluminum. The diameter was found to be 0.940 ±0.004 inch. The gap between the cadmium and aluminum was a maximum of 0.08 mil in the irradiated rod, which was essentially equal to the gap of 0.09 mil before irradiation.

POSTIRRADIATION EXAMINATION OF CONTROL ROD WITH CADMIUM-PLATED ALUMINUM CORE

Postirradiation examination of a type 6 control rod (Cd electroplate = 0.055 inch thick) that was inadvertently fully inserted in the reactor for one cycle [flux level = 4.8×10^{15} n/(cm^2)(sec)] revealed a separation between the cadmium and aluminum at both the inner and outer surfaces of the cadmium. The inner cadmium surface was initially formed when the cadmium was plated on an aluminum rod, and the outer was formed by swaging an aluminum tube over the plated cadmium. The separation was unexpected because preirradiation examination of these rods showed the average gap between the cadmium and aluminum to be <0.01 mil. The separation may have occurred during swaging but more probably developed during thermal cycling in the reactor.

About 40% of the gap at the inner circumference of the cadmium was filled with cadmium that had melted and resolidified, as shown in Figure 8. The maximum thickness of the resolidified metal was 0.5 mil. The maximum gap was about 0.6 mil wide and formed on the outer circumference of the cadmium. No evidence of melting was found on the outer circumference of the cadmium in any of the specimens.

Measurements of the OD of the control rod showed no distortion greater than the nominal variation of about 4 mils. Examination of all the specimens from this rod showed that the integrity of the aluminum had been maintained in spite of the fact that the cadmium core had partially melted during reactor operation.

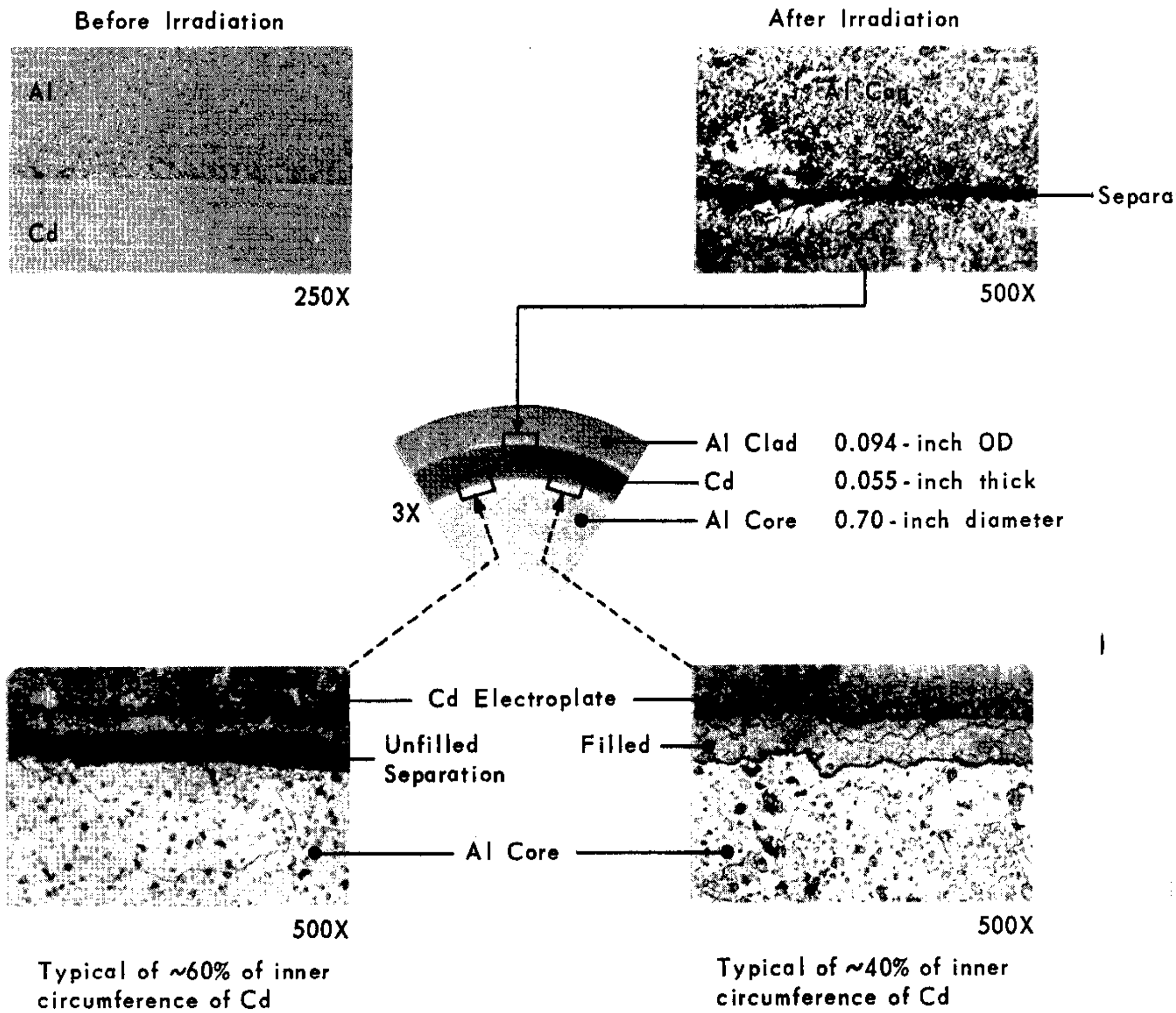


FIG. 8 TYPE 6 CADMIUM CONTROL ROD INTERFACE

CONCLUSIONS

On the basis of both laboratory experiments and reactor operating experience, unintentional melting of the cadmium core of an aluminum-clad reactor control rod will not cause penetration of the aluminum cladding and loss of the cadmium core. The rate of penetration of molten cadmium under conditions of continuous strain was found to be a maximum of 11 mils/day at 600°C. The conditions of the experiment produced the maximum possible attack on the aluminum and were greatly in excess of anticipated conditions in control rods. If the cadmium melts (mp 321°C) in a control rod it will be in contact with the aluminum cladding as a molten metal for only a short time. The cadmium will solidify at the cladding (calculated maximum temperature = 160°C). Because of the short reaction time and relatively low temperature, the aluminum will not be penetrated, and there will be little or no intergranular attack. Embrittlement of the aluminum cladding should not occur.

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