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PERFORMANCE OF ALUMINUM-URANIUM ALLOY FUEL PLATES UNDER HIGH TEMPERATURE AND HIGH BURNUP CONDITIONS

by

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ABSTRACT

Irradiation experiments have been made to determine the irradiation temperature and burnup limits for X8001 aluminum alloy-clad Al-17.5 w/o U-2 w/o Ni-0.5 w/o Fe alloy silicon-bonded plates of the type developed for the SL-1 reactor. The irradiations were conducted on prototype SL-1 plates both in MTR capsules and in the ANL-2 High Pressure Water Loop in the MTR. The loop experiments were conducted under local boiling conditions in water at 215°C (420°F) flowing at 12 ft/sec. One of the plates being irradiated in the loop developed a clad defect after a period of operation in high pH water. At the time of clad penetration the plate had achieved a burnup of 58 percent of the uranium (1.3 percent of total fuel alloy atoms). No catastrophic corrosion of the fuel alloy or extensive fission product release occurred when the plate was irradiated with the defect. Because of heavy scale deposition on the loop specimens fuel alloy temperatures were calculated to be as high as 560°C (1040°F). Fuel swelling occurred above temperatures near 450°C (840°F). The silicon bonding technique appeared to be highly effective in maintaining sound metallurgical bonds between fuel and cladding.

INTRODUCTION

The fuel elements for the SL-1 reactor consisted of plates of Al-17.5 w/o U-2 w/o Ni-0.5 w/o Fe alloy, clad with X8001 alloy. Extensive corrosion testing of the cladding alloy(1) had indicated that it could be expected to withstand the 215°C (420°F) water conditions in SL-1 for the anticipated core life of three years. The nickel and iron additions to the fuel alloy had also been shown to improve the corrosion resistance of aluminum-uranium alloys.(2) Considerable data were also available concerning the irradiation behavior of aluminum-uranium alloy fuel plates at

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the relatively low temperatures existing in research and test reactors. On the basis of the above information it was considered unlikely that the operating temperature of SL-1, which was on the order of 170°C (300°F) higher than that of the MTR, would lead to excessive fuel element corrosion, fuel swelling, or other undesirable developments. Subsequent operation of the reactor more than verified these assumptions. Corrosion and scale buildup on the fuel elements was much less than anticipated and no dimensional instability was observed in the fuel plates.

There was, nevertheless, considerable interest in determining the burnup and operating temperature limits of aluminum-clad aluminum-uranium alloy plates of the type developed for SL-1. Prototype SL-1 plates were therefore made for experimental irradiations under conditions considerably more rigorous than those which would exist in the reactor. The experimental irradiations have recently been completed. Although the hot cell examinations are not yet finished for all test specimens, sufficient information has been obtained to define the high temperature behavior of the fuel at burnups ranging up to more than 50 percent of the uranium (1.1 total atom percent burnup). Information has also been obtained on the corrosion and scaling behavior of the fuel plates at high heat fluxes under local boiling conditions.

EXPERIMENTAL PROCEDURE

Two sizes of prototype elements were made for the irradiation studies. The smaller size was made for capsule irradiations. These plates were 15/16 in. wide and 5-1/4 in. long. The larger plates were made for loop tests, and were 2 in. wide and 16 in. long. Both plates were the same thickness (0.120 in.) as the full size SL-1 plates. The fuel alloy was 0.050 in. thick and the cladding was 0.035 in. thick on each side.

Both specimen types were fabricated in identical fashion to the SL-1 core loading. The fabrication procedure used for the SL-1 core loading was described in detail[3] at the Fuel Elements Conference held in Gatlinburg in May, 1958. The fabrication procedure utilized the customary picture frame assembly. Before assembly the surfaces to be joined were coated with elemental silicon powder. The assembled components were then pressed between dies at 600°C for a few minutes until the molten silicon-aluminum eutectic was squeezed out of the joints. The compacts were then given a 4-to-1 reduction at room temperature to the finished size. The silicon bonding technique consistently yields plates which have a higher quality fuel-to-clad bond than can be achieved by roll-bonding alone. Silicon-bonded plates are therefore more resistant to blistering, and hence more suited to the higher operating temperatures found in power reactors.

The capsules used for the small plates were 1-1/8 in. in diameter and approximately 6 in. long. Figure 1 shows one end of a partially disassembled capsule containing a test specimen. Each capsule was made with a rectangular opening so that a 1/16 in. thick coolant channel existed on either side of the specimen. The capsule was made in two halves, each of which contained a 1/8 in. diameter hole extending the length of the capsule. Al-0.5 w/o Co alloy flux monitor wires were placed in each hole. A pin at each end held the capsule halves together and also retained the flux monitors and the loosely held specimen in the capsule. As shown in Figure 1, the ends of the capsule were built so that a column of capsules would nest together in such a way that all plates were aligned in a parallel plane. Unimpeded coolant flow through the column of capsules was thus assured. The capsules were stacked five to a column in standard MTR X-baskets and irradiated in the MTR reflector in water at approximately 50°C flowing at a calculated velocity near 20 feet per second.

The capsule specimens were irradiated for periods ranging from four months to two years. Maximum estimated unperturbed thermal fluxes were near 1.3×10^{14}. The maximum calculated burnup achieved is approximately 80 percent of the uranium, or a total atom burnup of 1.8 percent. The maximum heat fluxes obtained are calculated to be near 210 watts/cm^2 (680,000 BTU/hr-ft^2). These small plate specimens were therefore irradiated under conditions very similar to those existing for the MTR fuel elements themselves.

Figure 2 shows schematically the arrangement of the in-pile tube of the High Pressure Water Loop[4] used for the irradiation of two larger prototype SL-1 plates. A simplified drawing of the flow diagram is shown in Figure 3. The loop is located in a radial beam hole through the graphite surrounding the MTR. The tip of the in-pile tube is located near the outer edge of the reactor core in a cylindrical well in the reactor tank. This arrangement provided a maximum unperturbed thermal flux near 1.7×10^{14} at the in-pile end of the specimen. A flux plot along the length of the test section is shown in Figure 4.

The plates were irradiated in water at 380 psi and 215°C (420°F). The desired coolant water conditions were neutral pH (7.0) and a resistivity greater than one megohm-cm. The water velocity in the test section was 12 ft/sec. The maximum heat flux was 218 watts/cm^2 (690,000 BTU/hr-ft^2). Under these conditions it was calculated that local boiling would exist along a distance of 4.3 inches at the inner end of the fuel plate. Examination of the plates after irradiation indicated that boiling occurred along a length of approximately 6 in. As shown in Figure 3, the out-of-pile section of the loop contained a section for a control prototype plate so that the effects due alone to exposure to loop water could be distinguished from those due to irradiation. After the first plate was irradiated the loop was chemically decontaminated[5] in order to remove corrosion products which had accumulated during the previous six years of operation.

RESULTS

To date, four of the small plates irradiated in the capsules have been subjected to hot cell examination. These specimens achieved uranium burnups ranging from 19 to 32 percent (0.41 to 0.71 percent of total atoms). The maximum heat flux was 181 watts/cm^2 (573,000 BTU/hr-ft^2). The plates did not show measurable changes in length. Average width and length increases respectively of 0.45 and 0.48 per cent were observed. These changes resulted in volume increases, measured by immersion, that occurred at a rate of approximately 1.2 percent per one percent burnup of all atoms. The average weight increase was 51.5 mg per specimen.

Figure 5 shows a typical specimen after irradiation. The only visible surface change is the lighter color of the cladding which is directly over the fuel. Similar results have been noted on aluminum alloy-clad oxide fuel irradiated in the MTR process water.[6] Small amounts of scale were deposited in scratches on the cladding. Metallographic examination showed no significant changes in the microstructure of the fuel and cladding. The bond between all components was sound and free of defects.

The first larger prototype plate (ANL-2-11) was in the loop for 227 days and subjected to the equivalent of 156 days of full power operation of MTR. Burnup analyses made on samples cut from the plate showed that a maximum uranium burnup of 58 percent (1.3 percent total atom burnup) was attained. A detailed description of this experiment and the results obtained have been described previously.[7] Irradiation of this plate was terminated when low level fission product activity was detected in the loop water. The clad rupture followed a period of seven days during which a faulty ion-exchange column caused loss of pH control. During most of this 7-day period the pH was above its normal value of 7 and at levels on the order of 9 and 10. The loop water activity charts indicated that the element was irradiated for at least two days after the initial cladding penetration occurred. Loop water activity at no time reached a level high enough to set off the high-level alarm, and no appreciable fission product contamination of the loop was noted.

The postirradiation appearance of the in-pile plate may be seen in Figure 6. The defect in the clad was in the form of an open blister that was located near the edge of the boiling zone 4-3/4 in. from the hot end of the plate. An apparently unopened blister was also noted in a similar area 4-1/2 in. from the hot end of the plate. Inside the blisters themselves, the cladding had corroded at a much faster rate than the exposed fuel alloy. The plate was unevenly but heavily scaled. The average thickness of the oxide layer was 0.0063 in. The corrosion loss averaged 0.0062 in. per side. It was also noted that swelling of the fuel plate occurred over a distance of about two inches at the hot end of the plate. In this area the plate had increased in thickness by approximately 20 percent.

A metallographic section through the clad defect is shown in Figure 7. The blister was found to be filled with white corrosion products. The apparently unopened blister was also sectioned and was found to have in reality a small opening. This blister was also filled with white corrosion product. In both defect areas clad-core separation had occurred around the blisters. The unbonded surfaces extended in one case as far as 0.4 in. from a blister.

Figure 8 shows a section through the swelled area. The laminar cracking which accompanied the swelling is believed to be related to laminar cracks which were observed in some of the core blanks[8] fabricated for the SL-1 fuel elements. Both laminar and spherical pores were observed, as shown in Figure 9.

The out-of-pile plate was covered unevenly with scale that averaged 0.0041 in. in thickness. The corrosion loss was 0.0052 in. per side.

Scale was removed from both plates and subjected to chemical and X-ray diffraction analyses. At the hot end of the in-pile plate the scale was approximately 50 percent corundum, 40 percent boehmite, and 10 percent spinel. Scale removed from near the defect area and from the out-of-pile plate was greater than 90 percent boehmite. The thermal conductivity of a piece of scale taken from the in-pile plate was measured to be 0.0023 cal/sec-cm^2-°C/cm (0.56 BTU/hr-ft^2-°F/ft).

From the known heat flux and thermal conductivity of the scale it was possible to calculate the operating temperature of the plate. The calculations indicate that the fuel alloy centerline temperatures ranged from 560°C (1040°F) at the hot end of the plate to 230°C (430°F) at the cool end. Fuel swelling occurred in the temperature range of 450° to 560°C (840° to 1040°F). The clad defect developed at a point where the fuel temperature was near 350°C (660°F).

As mentioned earlier, the loop was chemically cleaned before the second plate (ANL-2-12) was inserted in order to remove loose corrosion products that were being circulated. It was evident that the heavy scale deposit which formed on the ANL-2-11 plate raised its operating temperature well above desired conditions. It was hoped that by keeping the scale thickness down the ANL-2-12 plate could be operated at the same heat flux as the ANL-2-11 plate but at a temperature low enough to avoid swelling of the fuel alloy.

The ANL-2-12 plate was irradiated in the loop without indication of a fission break or other operating difficulty. The plate was in the loop for 223 days and subjected to an equivalent of 125 days of full power operation of MTR. The plate was given an interim inspection in the MTR hot cell midway during the irradiation period. Burnup analyses made on samples cut from the plate after irradiation showed that a maximum uranium burnup of 45 percent (1.0 percent total atom burnup) was achieved. The highest pH value recorded during this experiment was 8.0. A detailed report describing the experiment and the postirradiation examination is in preparation.(9)

The postirradiation appearance of the plate is shown in Figure 10. As in the case of the ANL-2-11 experiment, heavy scaling developed on the in-pile plate. The average thickness of the scale was 0.0069 in. The coating was generally nonadherent except at the hot end where boiling had been most vigorous. The plate showed no changes in thickness that resulted from fuel swelling. The plate increased in length and width 0.020 and 0.012 in. respectively. The corrosion loss averaged 0.004 in. per side, based on thickness measurements. Weight loss measurements gave a corrosion rate of 7.4 mg/cm^2-mo.

Metallographic examination of the ANL-2-12 plate confirmed that no fuel swelling had occurred and showed that no significant changes in microstructure resulted from irradiation. Figure 11 shows a typical area at the hot end of the plate. No variations in grain size or appearance of the UAl_4 particles could be detected from one end of the plate to the other. The silicon-bonded joints between core and clad were sound and no undissolved silicon was observed.

The out-of-pile plate from the ANL-2-12 experiment is shown in Figure 12. The scale on this plate had an average thickness of 0.0027 in. The scale was colored a grayish blue and was generally quite adherent. Weight loss measurements on the plate showed that it had corroded at a rate of 4.4 mg/cm^2-mo.

DISCUSSION OF RESULTS

The fuel plates made for the SL-1 reactor differed from aluminum-uranium alloy fuel plates made for other reactors in two important respects: (1) the addition of 2 w/o nickel to the fuel alloy for improved corrosion resistance, and (2) the use of the silicon-bonding technique for greater resistance to high temperature blistering. The irradiation experiments conducted in the high pressure water loop on the prototype plates have indicated that both modifications were effective in raising the performance of the fuel plates.

The excellent corrosion resistance of the fuel alloy was demonstrated in the ANL-2-11 experiment, in which a defect developed in the in-pile plate after substantial fuel burnup had been achieved. Metallographic examination of the defect area showed that corrosive attack of the cladding was more severe than corrosion of the fuel alloy (see Figure 7). This observation was confirmed by the fact that only a small increase in loop water radioactivity was noted when the defect occurred.

The extensive overheating of the ANL-2-11 plate because of scale accumulation did not result in unbonds or blisters between the fuel alloy and the cladding. It is believed that the two blisters which formed on the plate followed penetration of the cladding during the operation in high pH water. The silicon-bonding technique is evidently a highly effective method of joining aluminum alloy components which must remain bonded under severe thermal conditions. The fuel swelling which developed in the fuel alloy was not associated with the clad failure. It is quite possible that if unfavorable water conditions had not developed the plate could have been successfully irradiated to substantially higher burnup without difficulty.

The defects which developed during the high pH conditions apparently formed in the zone of highest temperature where liquid water was in contact with the cladding alloy. As shown in Figure 6, the defects were near the edge of the boiling zone, rather than in the boiling area where highest heat fluxes and clad temperatures were present. It is believed that during operation the scale in the boiling area was filled with steam with near-neutral pH, whereas in the somewhat cooler zone down the plate where the transition from boiling occurred the scale was saturated with the highly alkaline loop water. This lead to pitting attack and ultimate penetration of the cladding.

Although the second loop plate, ANL-2-12, developed a scale film under irradiation that was as thick as that which formed on the ANL-2-11 plate no fuel swelling occurred. The fact that the ANL-2-12 plate was not irradiated to quite as high a burnup as that attained in the ANL-2-11 plate is not considered to be significant in considering the difference in swelling behavior. It appears that the scale which formed on the ANL-2-12 plate had a higher thermal conductivity than the scale on the ANL-2-11 plate. The changed scale characteristics may have been the result of the fact that the ANL-2-12 experiment was the first use of the loop after the chemical decontamination.

CONCLUSIONS

1. Aluminum alloy-clad aluminum-uranium alloy fuel plates of the SL-1 type are capable of operation to burnups on the order of 50 percent of the uranium (1 percent total atom burnup) at fuel temperatures exceeding 400°C (750°F).

2. Clad failure of aluminum alloy-clad aluminum-uranium alloy fuel plates is more apt to result from penetration by pitting corrosion than from fuel swelling.

3. Aluminum-uranium alloy plates clad with X8001 aluminum alloy can be operated successfully in under local boiling conditions in neutral water at 215°C (420°F).

4. Exposure of defected highly-irradiated Al-17.5 w/o U-2 w/o Ni-0.5 w/o Fe fuel alloy to water at 215°C (420°F) does not result in catastrophic corrosion or cause the release of large amounts of fission product activity.

ACKNOWLEDGMENTS

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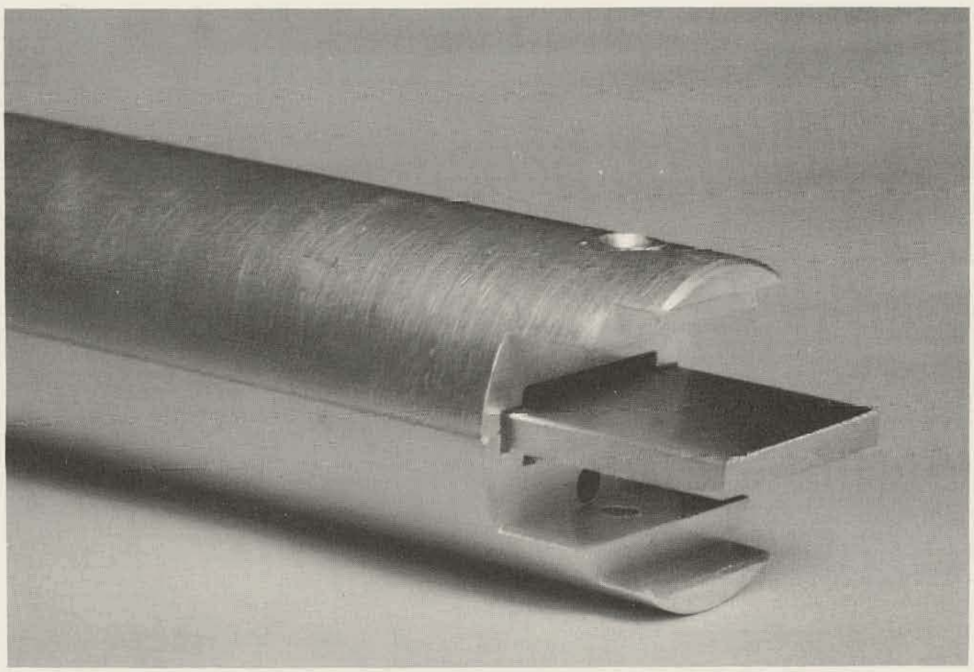

FIGURE 1. MTR CAPSULE WITH PARTIALLY WITHDRAWN
Al-17.5 w/o U ALLOY FUEL PLATE

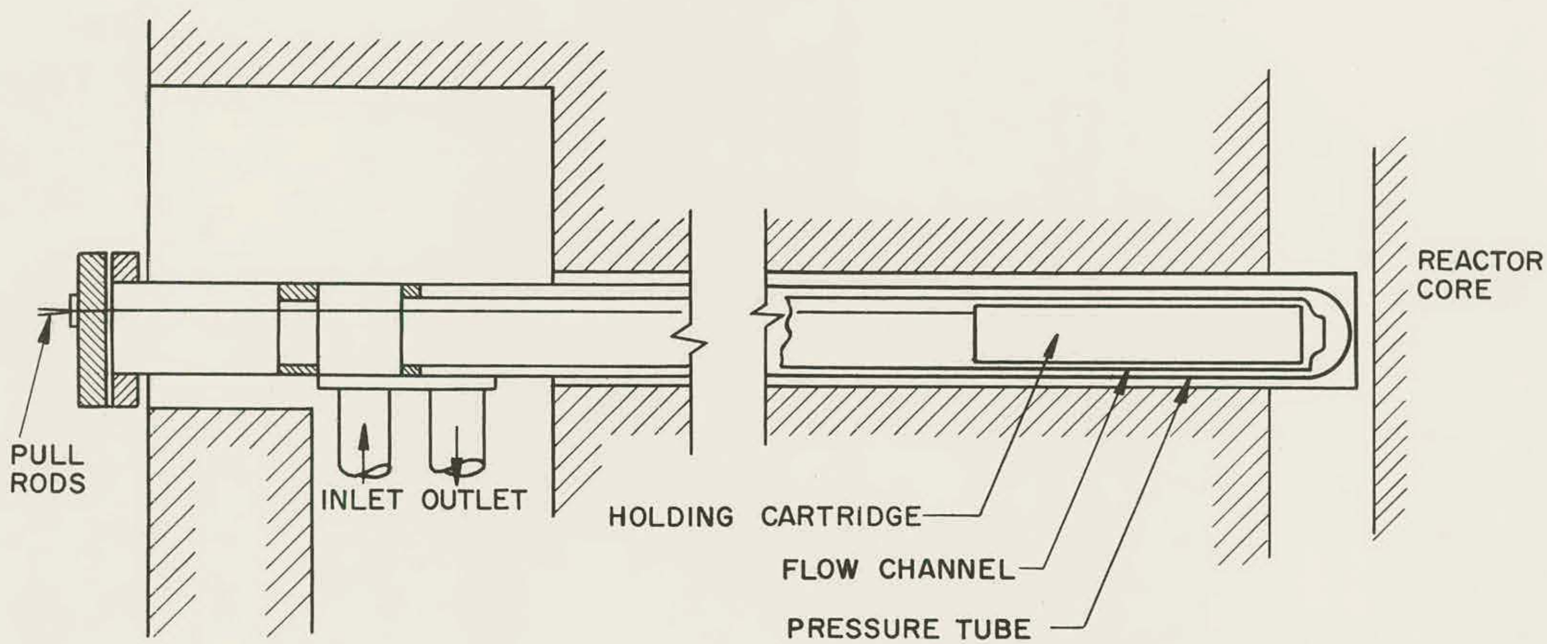

FIGURE 2. VERTICAL CROSS SECTION OF ANL-2 IN-PILE TUBE

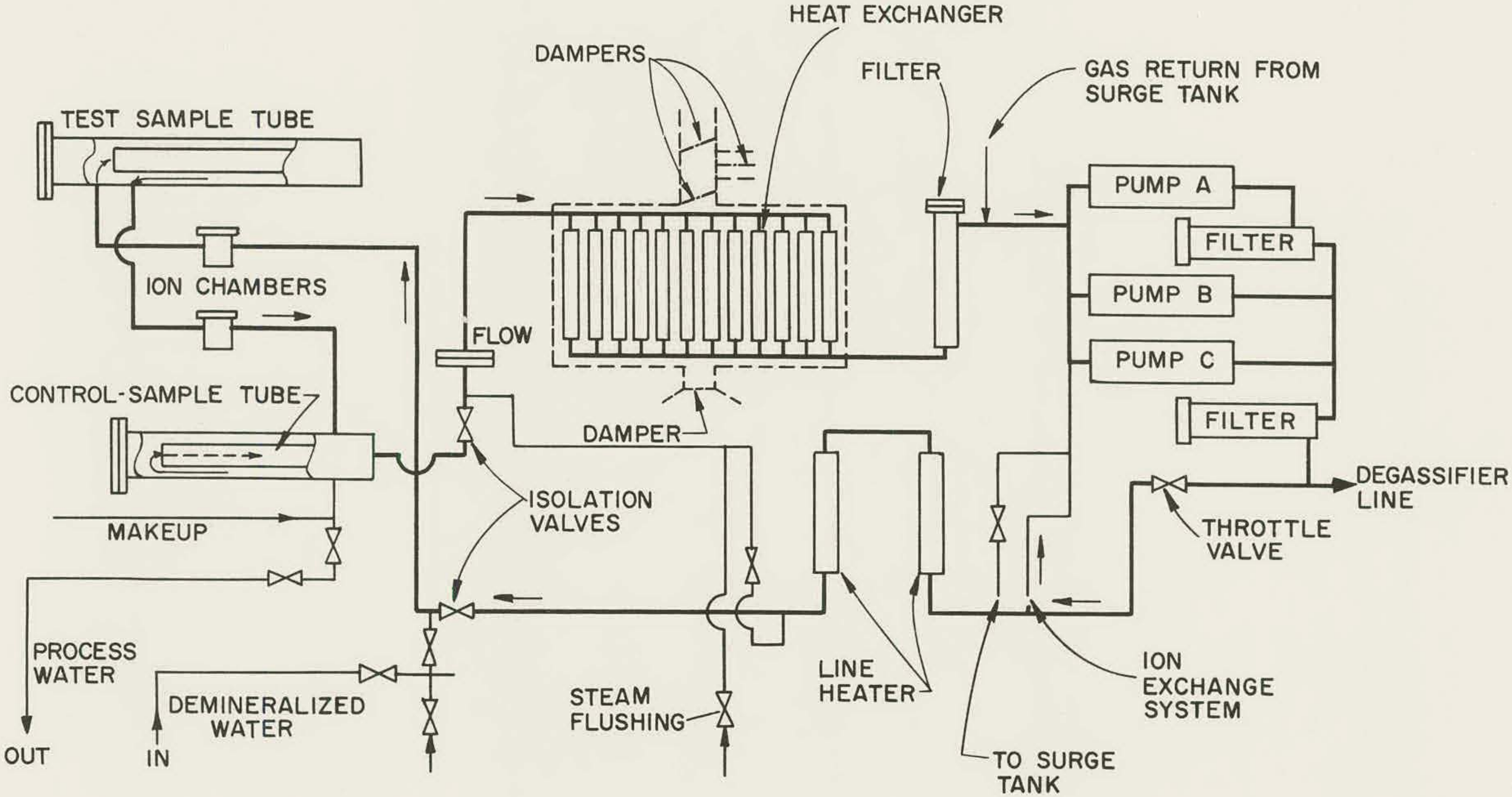

FIGURE 3. SCHEMATIC-FLOW DIAGRAM OF ANL-2 LOOP

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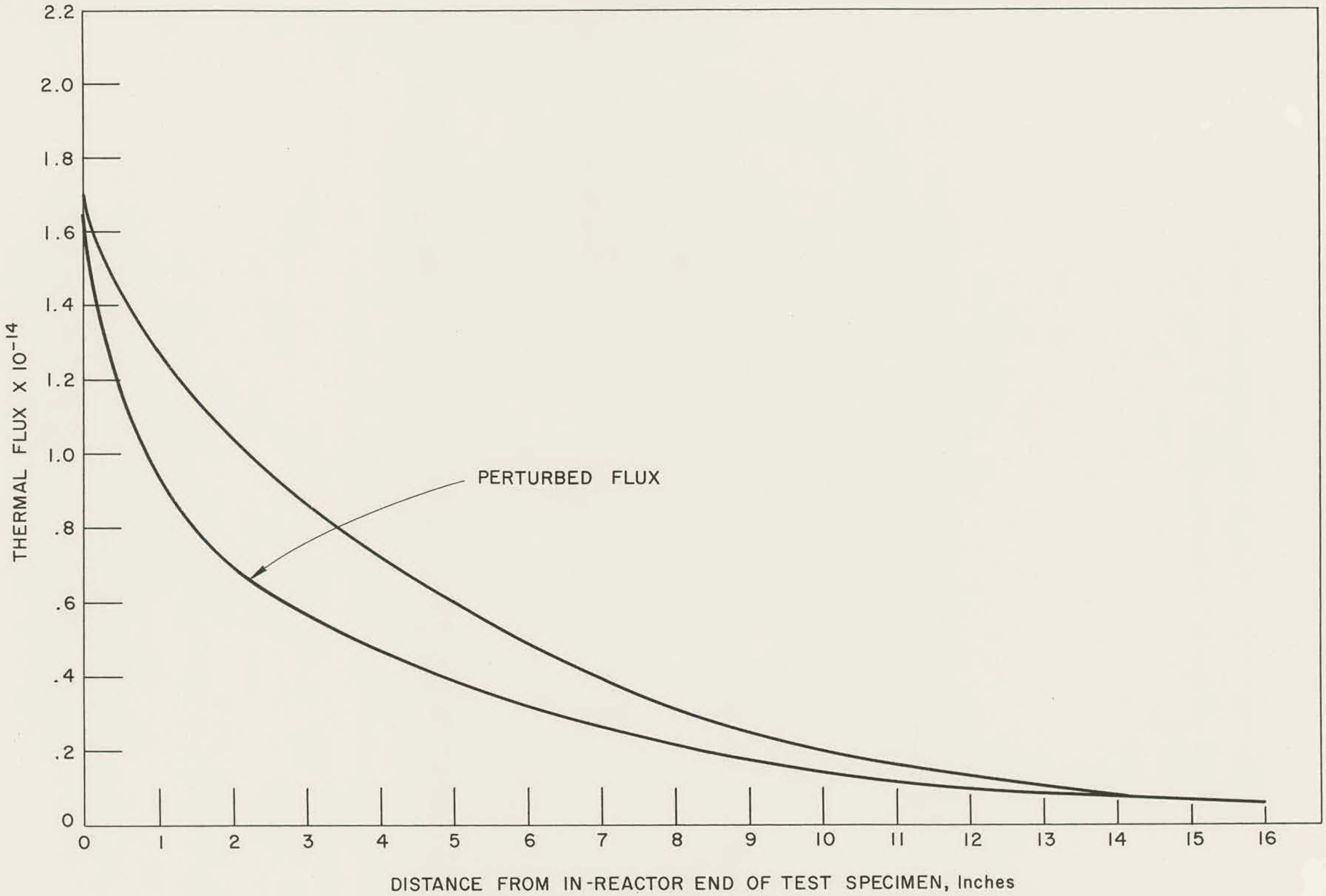

FIGURE 4. THERMAL NEUTRON FLUX PLOT FOR ANL-2 IN-PILE TUBE

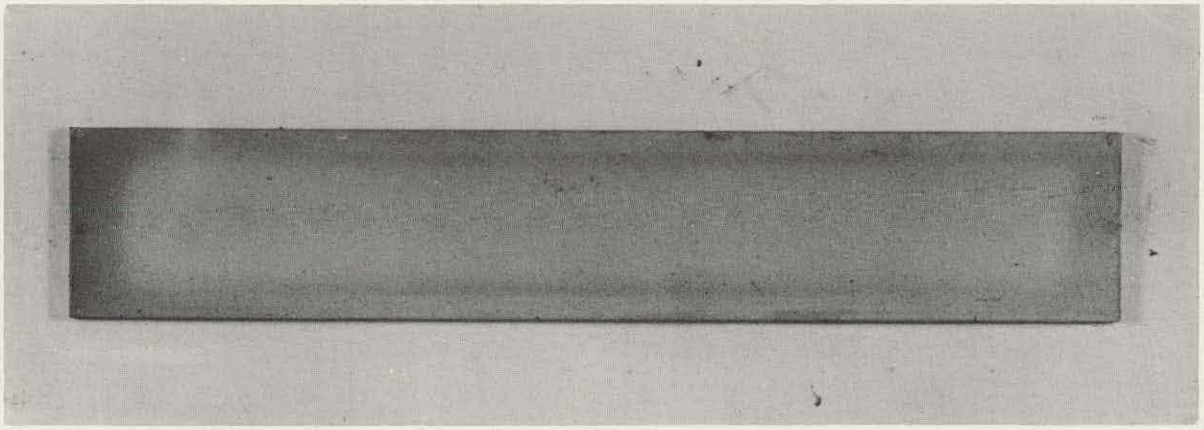

FIGURE 5. TYPICAL Al-17.5 w/o U ALLOY FUEL PLATE AFTER 32% URANIUM BURNUP IN WATER AT 55°C FLOWING AT 20 FT/SEC

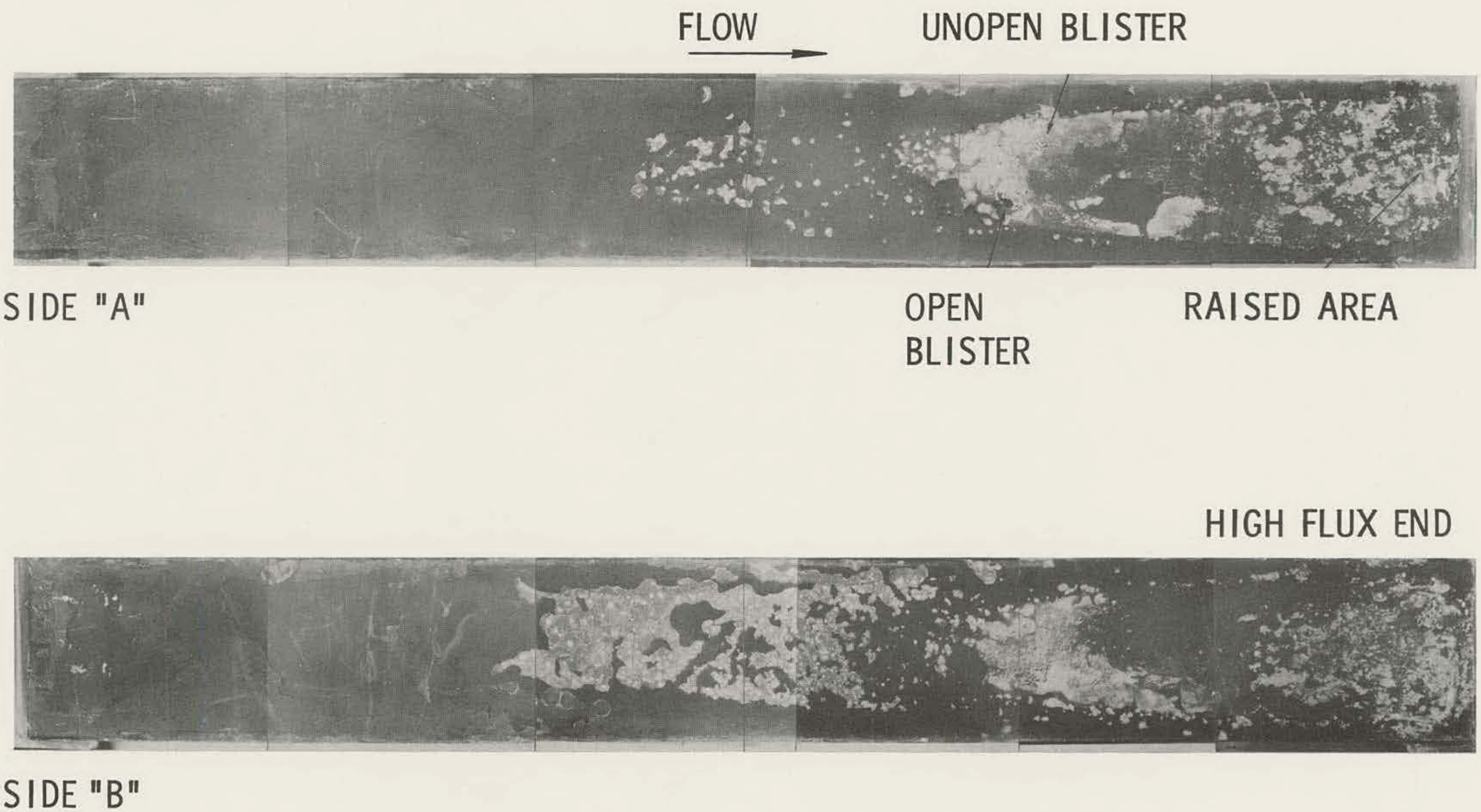

FIGURE 6. ANL-2-11 Al-17.5 w/o U ALLOY FUEL PLATE AFTER ~~55%~~ 58% URANIUM BURNUP IN WATER AT 215°C (420°F) FLOWING AT 12 FT/SEC

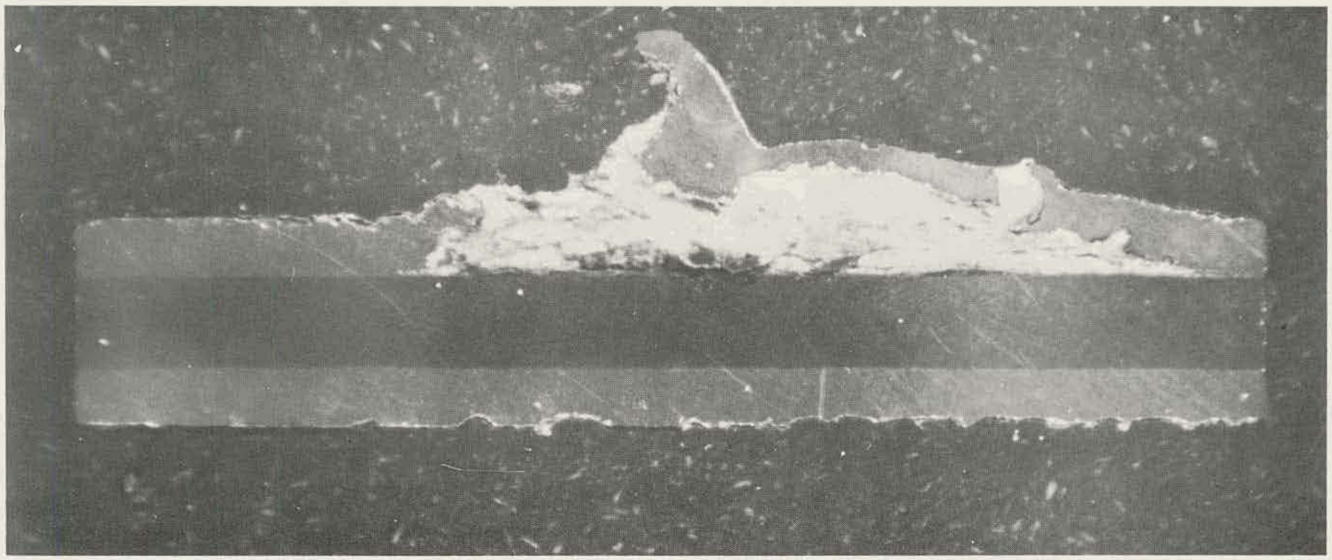

FIGURE 7. SECTION THROUGH FAILED CLADDING ON ANL-2-11
Al-17.5 w/o U ALLOY FUEL PLATE

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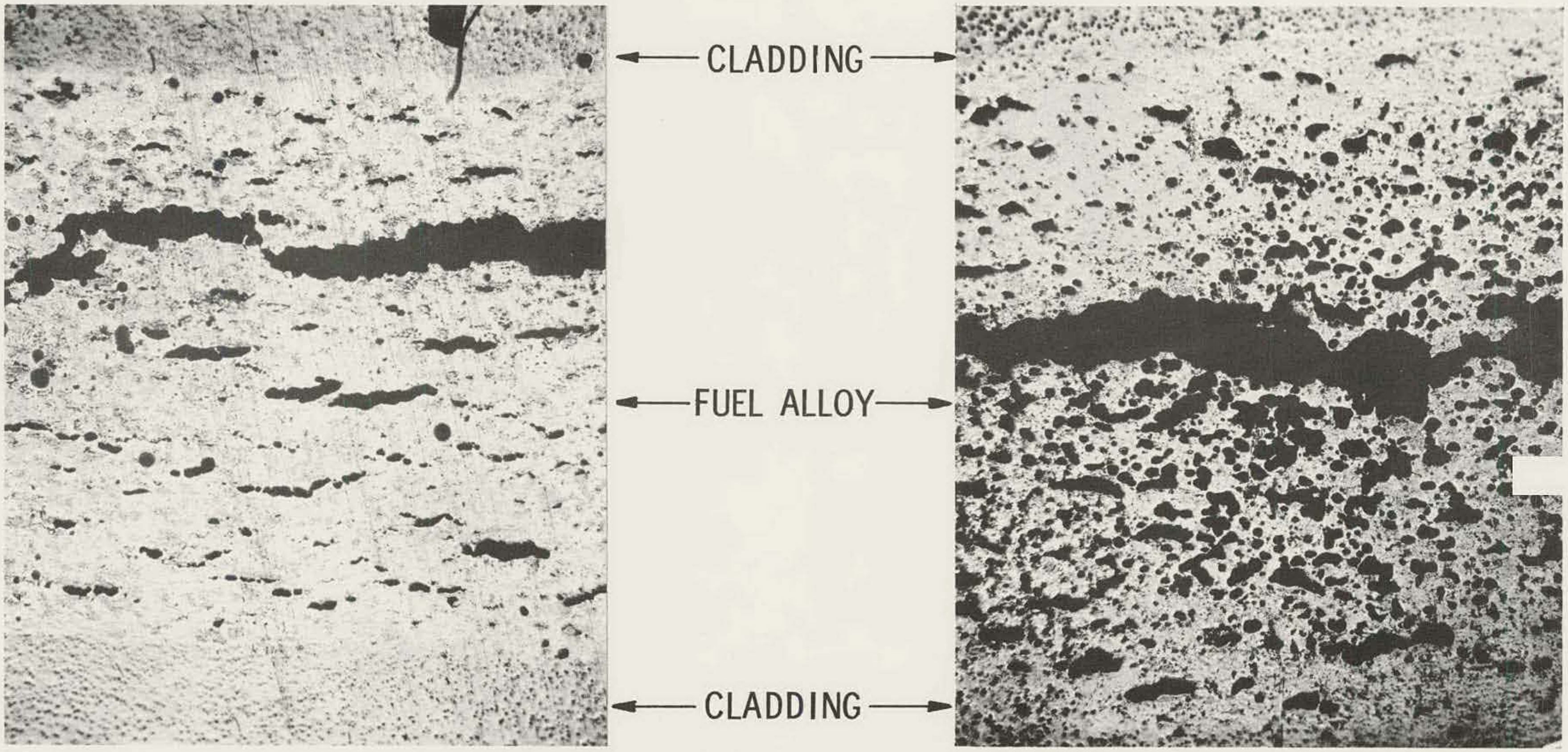
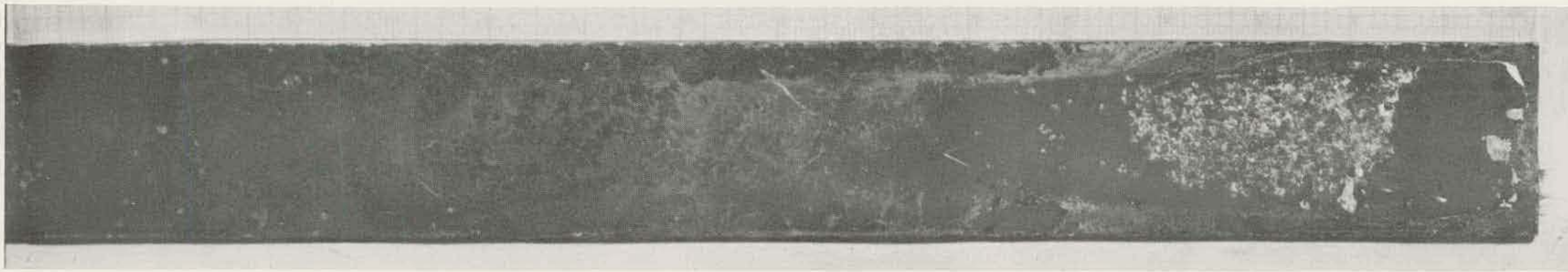
FIGURE 8. SECTION THROUGH SWELLED AREA OF ANL-2-11 Al-17.5 w/o U ALLOY FUEL PLATE



EDGE OF PLATE

CENTER OF PLATE

FIGURE 9. POROSITY IN SWELLED AREA OF ANL-2-11
Al-17.5 w/o U ALLOY FUEL PLATE

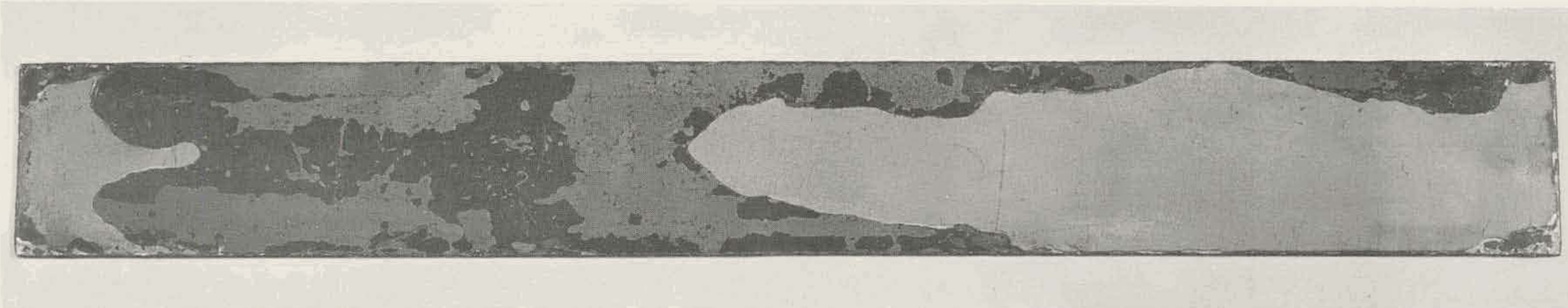


SIDE "A"

SIDE "B"

FIGURE 10. ANL-2-12 Al-17.5 w/o U ALLOY FUEL PLATE AFTER 42% URANIUM BURNUP IN WATER AT 215°C (420°F) FLOWING AT 12 FT/SEC

FIGURE 11. MICROSTRUCTURE OF Al-17.5 w/o U FUEL ALLOY IN PLATE ANL-2-12 AFTER ~~42~~ 45% URANIUM BURNUP



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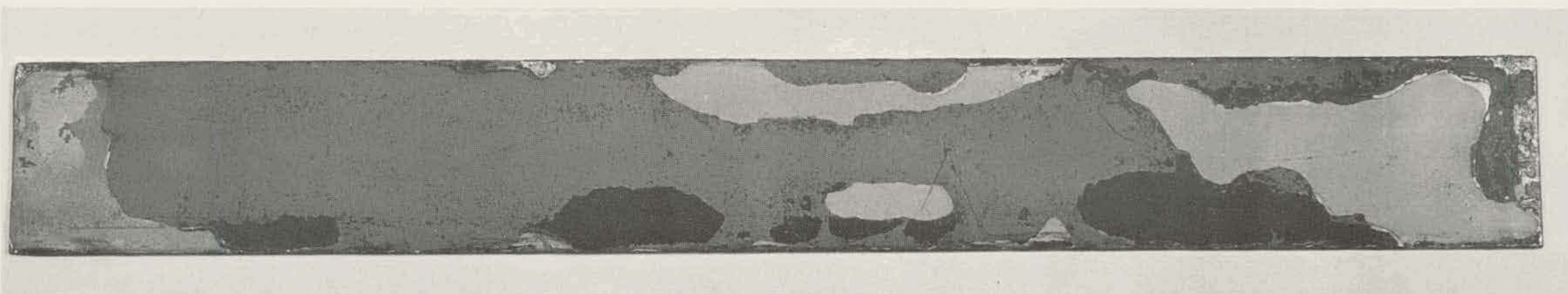

SIDE "B"

FIGURE 12. ANL-2-12 Al-17.5 w/o U ALLOY OUT-OF-PILE PLATE AFTER 125 DAYS IN WATER AT 215°C (420°F) FLOWING AT 12 FT/SEC