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ARGONNE NATIONAL LABORATORY
P. O. Box 299
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EXAMINATION OF IRRADIATED EXPERIMENTAL NRU FUEL RODS

by

S. H. Paine and W. F. Murphy

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ABSTRACT

Examination of flat, natural uranium plates mechanically clad with aluminum sheaths revealed a tendency for thermal contact between clad and core to diminish as a result of elongation and roughening of the uranium. The assemblies containing beta-quenched uranium showed less damage than those in which material rolled in the high alpha range was used.

A clad rod having square cross section was found to have failed by typical internal aqueous attack of the core material. Cause of initial perforation cannot be assigned with certainty.

I. INTRODUCTION

Under the Joint US-Canadian Sheath Program[1,2] a request was received from the Canadians early in 1954 that a composite rod containing a number of experimental flat plates of uranium sheathed in aluminum[3] be made the object of a cooperative laboratory study after irradiation in the Chalk River NRX reactor. Use of the hot laboratory facilities at Argonne National Laboratory was envisioned, and the scope of the examination was subsequently determined in a conference at ANL attended by representatives from AECL, AEC-SROO, AEC-COO, and the Laboratory.[4] Just prior to the scheduled operations the scope, at the request of the Canadians, was broadened to include study of sections from another clad experimental rod of square cross section.

Most of the hot laboratory work was performed during the period September 15 to 24, 1954. Three members of the AECL, D. G. Boxall, N. S. Spence and F. Abrams, were present during much of this time to observe and to work in collaboration with the ANL personnel. At the request of the Canadians a memorandum summarizing results of the examination and transmitting macrophotographs was issued by the Laboratory on October 4, 1954,[5] pending receipt from the Canadians of preirradiation data. This part of the agreement was evidently overlooked, and the data were obtained only when the Canadian final report became available early in 1955.[6] The plate elements were re-examined more carefully by ANL workers when new, improved photographic equipment became available some time later. The opinions resulting from this work are not entirely in accord with those obtained from the initial examination. All the information available to the study, therefore, has been critically re-examined, and the program is closed by issuance of the present report.

II. DESCRIPTION OF SPECIMENS

Flat Plate Elements

The metallurgical history of the specimens is detailed in Appendix A. The flat plate rod was essentially a composite of ten separately sheathed elements cut from the same flat bar of uranium and reassembled end to end. Each core element was 12 in. long and had a cross section 0.170 x 1.340 in. Aluminum cladding, 0.040 in. thick, was sized to the core by drawing through a die. Dovetail end plugs brought the total length of the sheath to approximately 13 in. A schematic representation of the construction is shown in Figure 1-a. Elements were numbered 1 to 10 from bottom to top in the composite rod; odd numbered pieces were in the as-rolled condition (high alpha rolling), whereas even numbered pieces had been water quenched from the beta phase after the rolling operation.

The total exposure of the rod in the reactor was computed to be 675 MWD/T. Upon radiographic examination, Plate No. 6 was found to be ruptured and No. 4 was badly warped. Plates Nos. 2, 3, 5, 7, and 8 were selected for examination at ANL after disassembly of the rod in the NRX canal.

Square Rod

The core of the square rod was a natural uranium bar, 10 ft. long, having a cross section 1.150 in. square. It was essentially in the beta-annealed condition and was clad with a 0.040-in. aluminum sheath. Details of its fabrication are given in Appendix A, and a schematic cross section is shown in Figure 1-b. Total exposure in the reactor was computed to be 362 MWD/T at the time the sheath was found to have failed. Examination showed a 3-in. crack along one corner of the assembly near the middle of the rod. Two pieces, each a foot long, were sheared from this portion of the rod for examination at ANL.

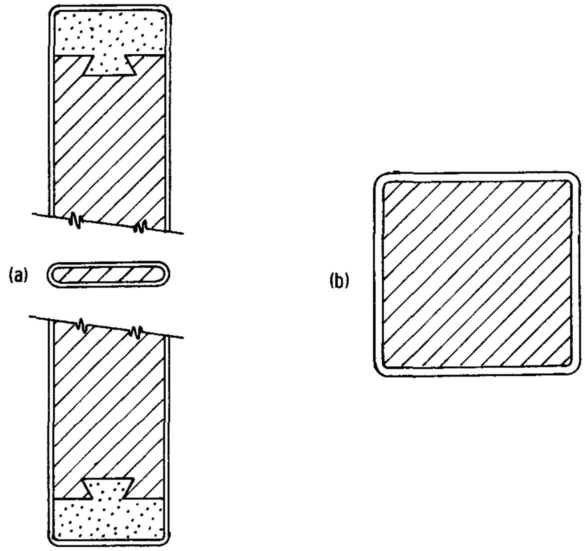

Figure 1

Sectional Diagrams of (a) Flat Plate and (b) Square Rod Aluminum-clad Uranium Assemblies (Not to relative scale)

III. EXAMINATION PROCEDURE AND OBSERVATIONS

A stereophotographic study of specimens is given in Appendix B, Figures 3 to 13.

Flat Plate Elements

The Canadians had radiographed the composite flat rod before irradiation and after exposure to 423 and 675 MWD/T. By means of small fiducial notches machined on the edges of the core at 3-in. intervals, they were able to determine from the negatives whether length changes had occurred.(6) Their data are presented in Table I.

At ANL the five flat elements were examined as received. The cladding was clean, except for a few rust-colored spots, and no pitting or surface roughness was observed. One end of all plates was bent somewhat, the damage having occurred, in the opinion of the AECL men, at the time of removal from the assembly. Plates Nos. 2 and 8 were both visibly necked down over a length of 3 in. at one end. Observations are summarized in Table II.

Width and thickness measurements were made at the middle of Plates Nos. 2 and 5 by micrometer, and lengths were measured with an inch scale to an estimated accuracy of $\frac{1}{64}$ in. Cladding was then stripped from these two plates and the measurements were repeated. Table III presents this information for comparison with preirradiation data supplied by the AECL report.(6)

TABLE I

Dimensional Changes Observed Radiographically in Composite Flat Plate Rod

Plate No.	Heat Treatment	Distance Above Calandria Floor (in.)	First Exposure		Second Exposure	
			% Change in Length	Relative Exposure [a]	% Change in Length	Relative Exposure
2	As alpha-rolled	17	0.25	0.28	0.33	0.45
4	As alpha-rolled	44	0.62	0.55	2.17	0.89
6	As alpha-rolled	70	1.01	0.62	4.13	0.99
8	As alpha-rolled	96	0.23	0.44	0.27	0.70
10	As alpha-rolled	122	0.07	0.10	0.00	0.16
1	Beta-quenched	4	0.05	0.10	0.00	0.16
3	Beta-quenched	30	-0.05	0.44	-0.33	0.70
5	Beta-quenched	57	0.08	0.62	-0.40	0.99
7	Beta-quenched	83	-0.07	0.55	0.03	0.89
9	Beta-quenched	109	-0.07	0.28	0.00	0.45

[a] Assuming cosine buckling and setting maximum exposure reached at middle of rod equal to 1.00.

TABLE II

Examination of Flat Plate Elements as Received at ANL

Plate No.	Metallurgical Condition	Condition of Cladding	Macro Features as Received [a]
2	Alpha-rolled	Clean except for a few rust spots. No pits or roughness.	Reduced cross section for 3 in. at one end. Pronounced bend.
8	Alpha-rolled	Clean except for a few rust spots. No pits or roughness.	Reduced cross section for 3 in. at one end. Pronounced bend.
3	Beta-quenched	Clean except for a few rust spots. No pits or roughness.	Bent at one end.
5	Beta-quenched	Clean except for a few rust spots. No pits or roughness.	Bent at one end.
7	Beta-quenched	Clean except for a few rust spots. No pits or roughness.	Bent at one end.

[a] Some of the bending observed may be due to mechanical damage in disassembly of rod.

TABLE III

Dimensional Measurements of Plates Nos. 2 and 5 before and after Irradiation

Specimen Measured[a]	Sheathed Measurements			Stripped Measurements		
	Thickness (in.)	Width (in.)	Length (in.)	Thickness (in.)	Width (in.)	Length (in.)
Before Irradiation Plate No 5	0.257	1.450	$13\frac{1}{8}$	0.170	1.340	12
After Irradiation						
Plate No. 2 (alpha-rolled) at Uniform End	0.255	1.450	$13\frac{3}{8}$	0.171	1.336	$12\frac{3}{16}$[b]
at Necked End	0.237	1.377		0.164	1.279	
Plate No. 5 (beta-quenched)	0.266	1.451	$13\frac{1}{8}$	0.177	1.349	12

[a] Thickness and width measurements have an estimated tolerance of ±0.002 in. or less; length is measured to $\frac{1}{64}$ in.

[b] Entire elongation occurred in a 3-in. length at one end of plate.

It will be seen from Table III that the specimen irradiated as-rolled (No. 2) remained nearly constant in thickness and width, but elongated appreciably. Elongation was practically confined to the end of the plate which had necked down. The plate which had been beta quenched (No. 5) remained unchanged in length, but the thickness and width dimensions increased slightly, of the order of ten mils.

Appearance of the stripped plates is recorded in Appendix B, Figures 3 to 10. These will be discussed more fully below.

Square Rod

It is reported(6) that the length of the square rod had remained constant during irradiation. Two sections, each approximately a foot long, sheared from the middle portion of the rod and still enclosed in the aluminum sheath, were received at ANL for examination. They are shown in Figure 11. Unfortunately, shearing was done through the very center of the portion which had the visible sheath rupture in it. Part of the distortion observed in this region, therefore, is due to mechanical disturbance after irradiation. All four sides of the aluminum sheath were stained by rust-colored material on the surface. Handling of the specimens dislodged copious amounts of uranium oxide from the sheared ends. Careful examination of the corner edges showed them to be bright, and the metal was dented and scraped in places.

Thickness of the sheathed rod varied, in general, from 1.254 to 1.257 in. over the flat surfaces. On one of the sections, however, a raised portion, about an inch in diameter, appeared and gave the rod a dimension approximately $\frac{1}{16}$ in. greater than the average. Cladding was removed by hammer and chisel from the piece on which this raised area was seen. Large quantities of loose, powdery oxide were present everywhere under the cladding. However, along the edges and in the vicinity of the raised spot a layer of tightly adherent oxide was present. Most of the oxide was removed by scraping, brushing, and washing with acetone. The appearance of the surfaces at this time is shown in Figures 12 and 13.

The dimensions of the rod as cleaned varied from 1.150 to 1.190 in. The larger spans were found along edges and in those portions where an oxide layer was quite evidently still present. Measurements over the bump mentioned above ranged up to 0.173 in., so it is evident that the distortion was caused by a heavier than average accumulation of oxide, most of which was removed in the cleaning operation. This spot may be seen in Figure 12-a.

A network of surface cracks was observed on one face of the bar, extending from its middle to the end remote from the sheath rupture. It may be seen plainly in Figure 13-d.

IV. DISCUSSION

Flat Plate Elements

On the basis of initial examination it was reported that there was no visible deterioration of the sheath, and that the sheath uranium contact appeared to be very good on those specimens which were stripped.(6) Additional examination indicates that this view may be overoptimistic. That contact was very good at the edges of the plates is evident from an examination of Figures 4 and 8, for here a fairly general metallurgical bond had formed between core and clad. However, contact away from the edges was evidently not so good (Figures 5 and 9), especially as surface roughness increased with irradiation time.

Judging from Figure 9 and applying a proper scaling factor, the peak-to-valley relief on the surface of the beta-quenched specimen (No. 5) is approximately 0.020 in. The thickness data in Table III indicate that nearly half of this relief was produced during irradiation. The Canadian report(6) mentions that surface roughness was slightly increased at the end of irradiation. Only the highest peaks were apparently in metallurgical contact with the cladding, judging from the appearances of the specimens as shown in Figures 7 and 8. Thus, even for the dimensionally stable beta-quenched material, progressive worsening of the flat surface contacts would tend to increase the heat flux and the temperature at the edges. There are quite definite indications that incipient eutectic melting between clad and core had occurred (Figure 10).

Incipient eutectic melting appears to have been worse on the as-rolled plate (No. 2) than on the beta-quenched specimen, although the former was subject to a much lower neutron flux. Additional evidence of overheating in this plate is indicated by an area of discoloration (Figure 3-b) in the center of which is a group of fusion pits (Figures 3-b and 5). These evidences are in the portion of the plate where localized elongation occurred. It is likely that clad-to-core contact here was practically nonexistent except at the edges and that the eutectic melting damage was greatest in this specimen because the greater share of all heat produced had to be dissipated through the edge contacts. Judging from the relative exposures of the two types of material, the beta-quenched assembly should be able to stand at least twice the exposure that would rupture an as-rolled element, and probably many times more.

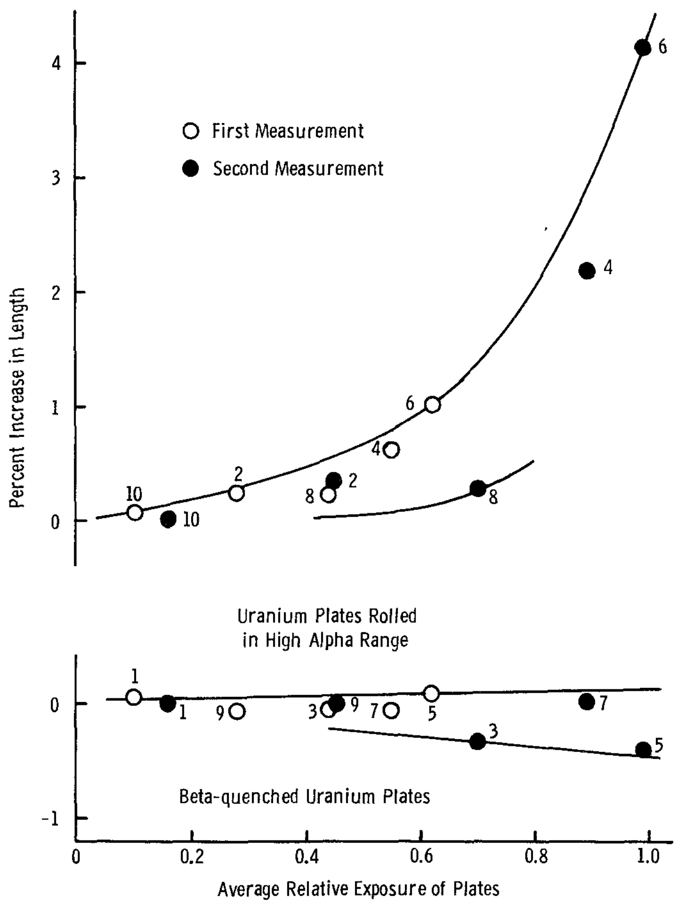

Figure 2 Elongation of Flat Plates as a Function of Relative Exposure

A typical difference between the dimensional stability of as-rolled and beta-quenched materials is shown by the group of flat plates. If it be assumed that buckling of the reactor flux is essentially a cosine function falling off to zero at the ends of the composite rod, the relative exposures of the various plates may be computed and dimensional changes may be plotted as a function of a burnup ratio. The data which result from these assumptions are given in Table I and have been plotted in Figure 2. It will be seen at once that the as-rolled material, after an initial low-growth exposure, tends to elongate rapidly, whereas the beta-quenched material either remains nearly unchanged or tends to a slight shrinkage. This relative behavior is to be expected from the metallurgical history of the materials,[7,8] but, in the test under consideration, the sheath has apparently provided considerable restraint to the as-rolled specimens so that the elongation is initially retarded. The reactor flux probably is not exactly defined by a cosine function. However, so interrelated are the data from the various plates (plate numbers are shown on the graph) that reducing the scatter of the points would accentuate the restraining effect just mentioned.

Square Rod

No evidence of metallurgical bonding between clad and core was seen on the stripped section of square rod, nor on the cladding removed from it. The oxide accumulations were typical of corrosive aqueous attack and were great enough at the edges of the bar to explain much of the rupture damage to the rod sheath. However, the initial perforation which permitted water to enter cannot logically be attributed to the increase in dimensions caused by the oxide.[6] From the thinned and beaten appearance of the sheath material at the rupture and on the corners adjacent (see Figure 11), it seems probable that the initial flaw might have been caused by an inadvertent bump suffered in handling the heavy rod.

The cracks appearing on one face of the bar (Figure 13-b) are clean and angular, do not appear to have suffered modification by aqueous corrosion, and are wide enough to have changed the dimensions of the bar appreciably. However, the measurements across this face are identical with the preirradiation data. It is most likely, therefore, that they are a craze crack system in an oxide layer ready to spall from the surface and not a series of cracks in the uranium itself.

V. CONCLUSIONS

1) The test confirms the fact that beta-quenched and beta-annealed uranium bar materials are essentially dimensionally stable under neutron irradiation, and that they are more subject to surface roughening than material rolled in the high alpha range.

2) There is evidence that the flat plate fuel element with unbonded aluminum cladding is susceptible to deteriorating heat transfer conditions when used either with as-rolled or beta-quenched uranium. The design may be adequate for short irradiation periods or low rates of heat production, but its limitations must be recognized. Beta-quenched material appears to be able to withstand at least twice as much exposure as the alpha-rolled uranium, and probably more.

3) The 0.040-in. aluminum cladding exerts considerable initial restraint upon elongation of alpha-rolled uranium. However, its yield strength is apparently smaller than the stress which causes elongation of the core.

4) It is suggested that failure of the square bar probably started as a mechanical flaw and proceeded by the corrosion mechanism of uranium oxide formation and swelling.

ACKNOWLEDGMENTS

The valuable technical assistance of A. C. Klank in performing the examinations is acknowledged. F. L. Brown and R. J. Fousek assisted in the photographic work.

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7. Paine, S. H., Persiani, P. J., and Murphy, W. F., Irradiation of Prototype CP-4 (EBR-I) Reactor Fuel Slugs in the CP-3' Goat Hole, ANL-5855 (1958).

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APPENDIX A

Metallurgical and Irradiation History of Specimens(6)

Flat Plate Composite Rod

Plates were all cut from a single bar of natural uranium, 11 ft long and 0.170 x 1.340 in. in cross section, which had been rolled in the high alpha range. Plates were 12.0 in. long and were numbered consecutively from 1 to 10, starting at the end which corresponded to the original ingot bottom.

Odd-numbered specimens were heated in a lead pot at 1350°F (732°C) for $2\frac{1}{2}$ min, then quenched in 68°F water (20°C). Even-numbered specimens were left in the as-rolled condition. All plates were pickled in hot nitric acid to remove lead and rolling scale. Dovetails and fiducial edge grooves were machined, the general surface, however, being left as it came from pickling.

Aluminum end plugs were fitted into the dovetails at ends of plates, and the plates were introduced into individual extruded aluminum tubes having 0.040-in. walls which thickened to 0.055 in. at the edges of the plates. Each assembly was then drawn through a die to insure close contact between sheath and core, and the ends were heliarc welded after preheating weld area. The sheathed plates were loaded end to end in a special tube which permitted light water to circulate past them when placed in the reactor, and kept the elements in line and under lateral restraint by means of four, $\frac{1}{2}$-in. round aluminum tubes nested against them in the water channel.

The assembly was given 675 MWD/T of exposure in the NRX reactor, and was dimensioned radiographically at the 423 and the 625 MWD/T levels. After the latter examination, irradiation was discontinued because of the discovery of a ruptured sheath on the No. 6 element in the composite.

Square Rod

The square bar was rolled in the high alpha range to $1\frac{1}{2}$-in. dimensions from a standard $4\frac{1}{2}$-in. diameter ingot. It was roller straightened and then beta treated by heating for 15 min in a lead bath at 1000°F (538°C), transferring to a second bath at 1350°F (732°C) for 5 min, quenching back into first lead bath for 5 min, and then removing to a water quench. Roller straightening was required after heat treatment. The bar was then machined to the final core dimension of 1.146-1.150 in. by 10 ft long, leaving a radius of $\frac{1}{32}$ in. at all corners. Sheathing was accomplished by placing it in a 0.040-in. wall aluminum tube and drawing the assembly through a die. Closure was completed by standard welding techniques.

The rod was irradiated to an exposure of 362 MWD/T and was removed because of radioactivity in the coolant water flowing through its channel. Examination disclosed that the sheath had split along one corner of the rod for an approximate length of 3 in. near its middle.

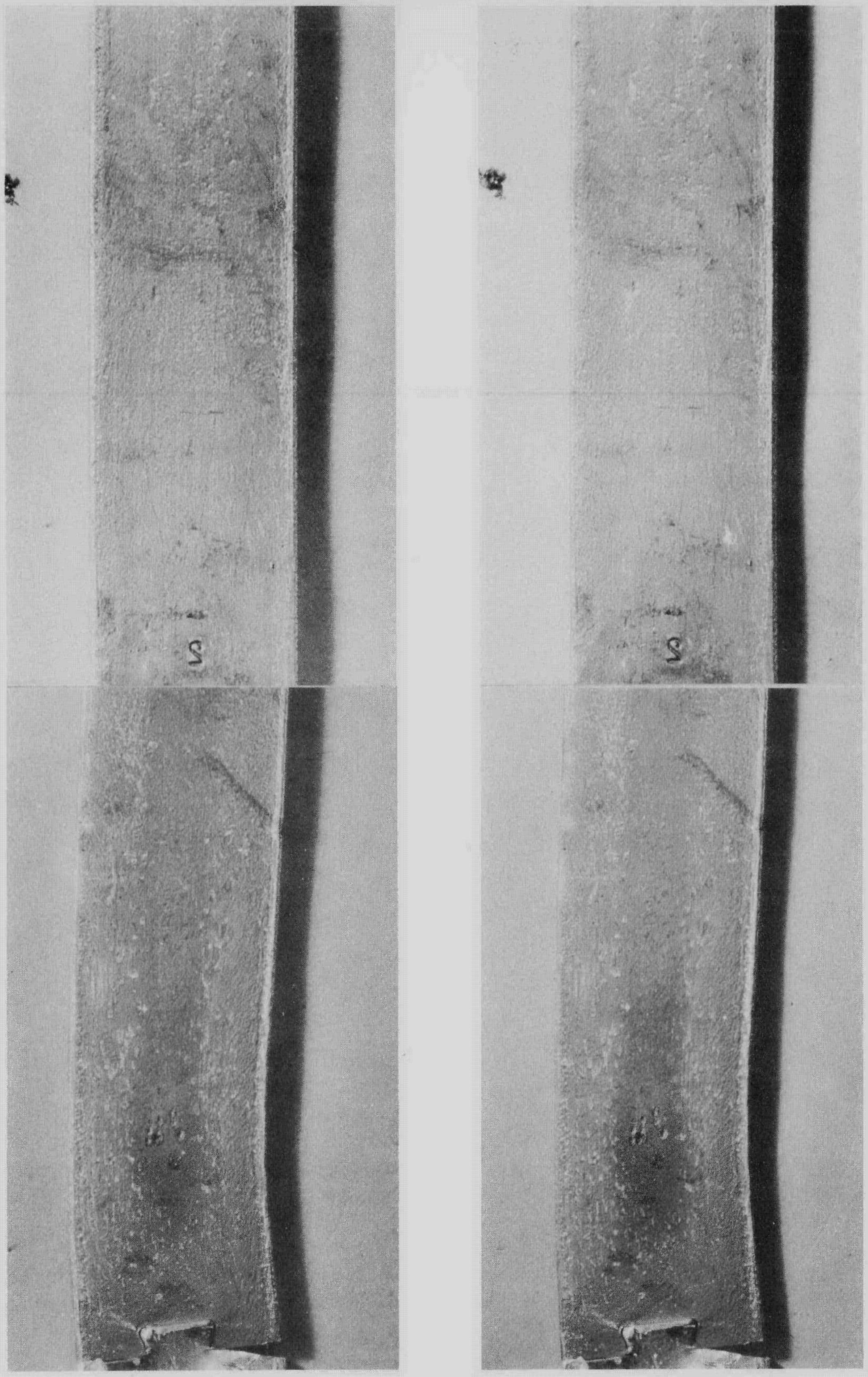
APPENDIX B

Stereophotographs of Specimens

The photographs shown in Figures 3, 7 and 11-13 were made with a special monocular camera constructed at ANL to take stereo views serially through the wall of the cave. Figures 4-6 and 8-10 were taken with a Bausch and Lomb stereobinocular constructed specially for cave use.

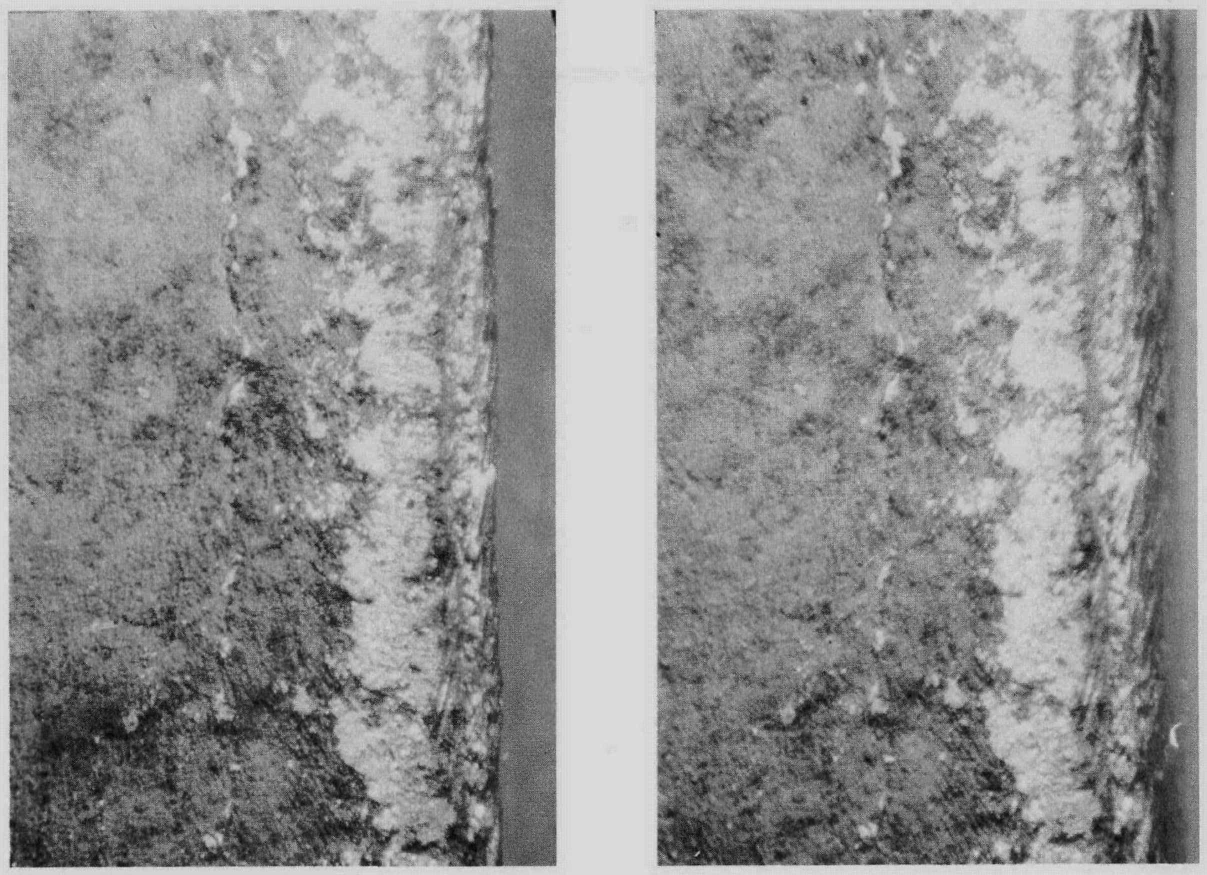
a)

b)



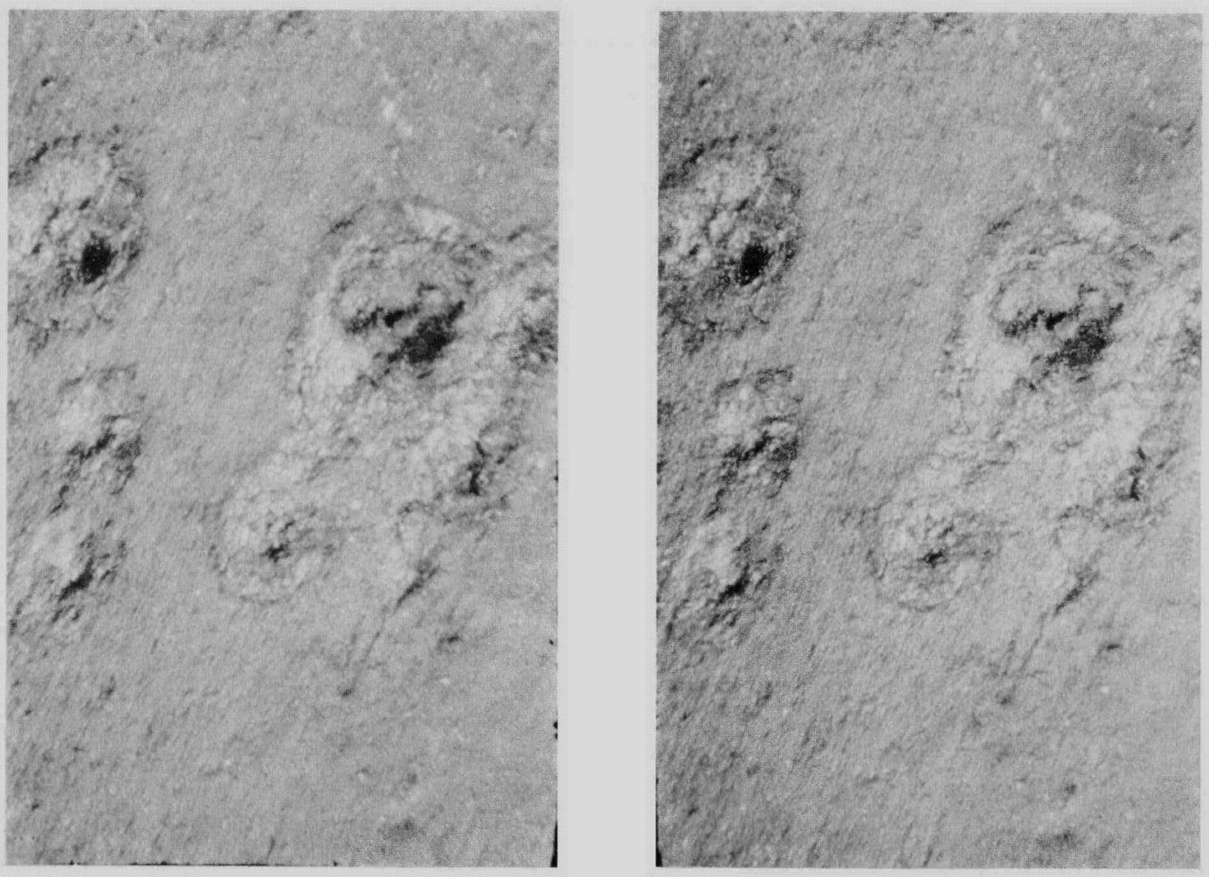
16325, 16319 1X

Figure 3. Views of Both Sides of Flat Plate No. 2 With Sheath Removed (As-rolled material)



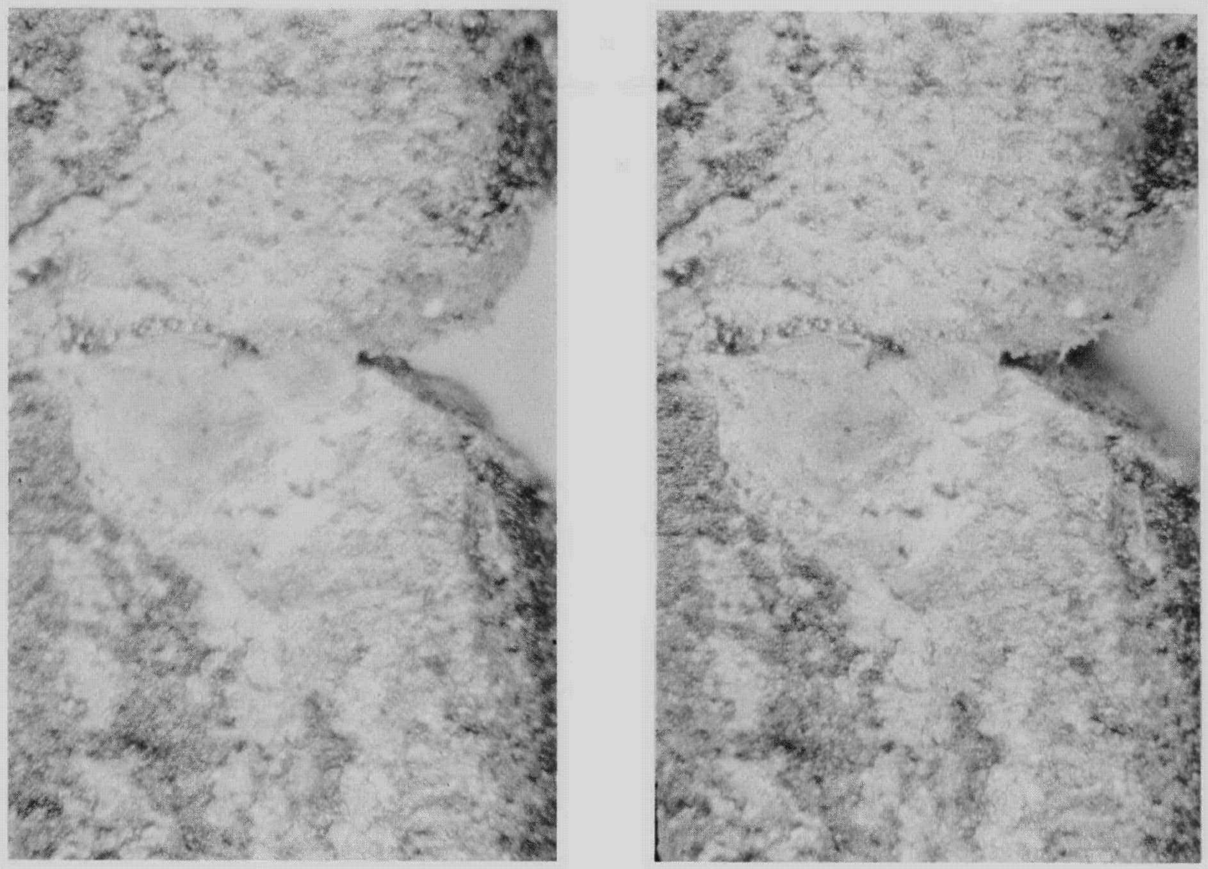
17254 12X

Figure 4. Magnified View of Plate No. 2 at Edge. Raised edge, parallel fissure, and adhering aluminum are seen clearly in this stereo.



17250 12X

Figure 5. View of Surface Pits on Damaged End of Plate No. 2

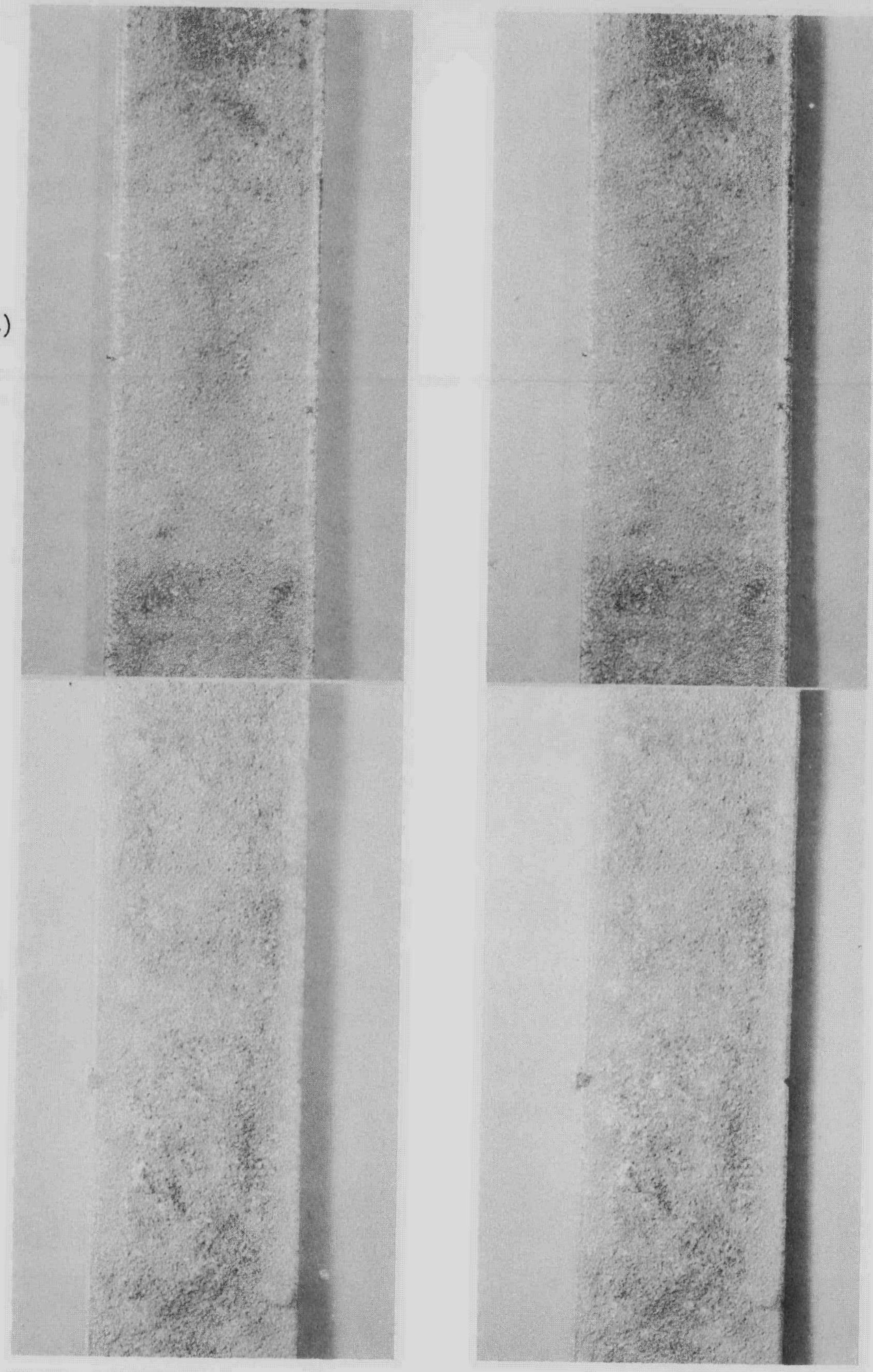


17252 34X

Figure 6. Greatly Enlarged View of Aluminum Fused to Edge of Plate No. 2. Notch is fiducial mark used for radiographic dimensioning of plate before removal of sheath.

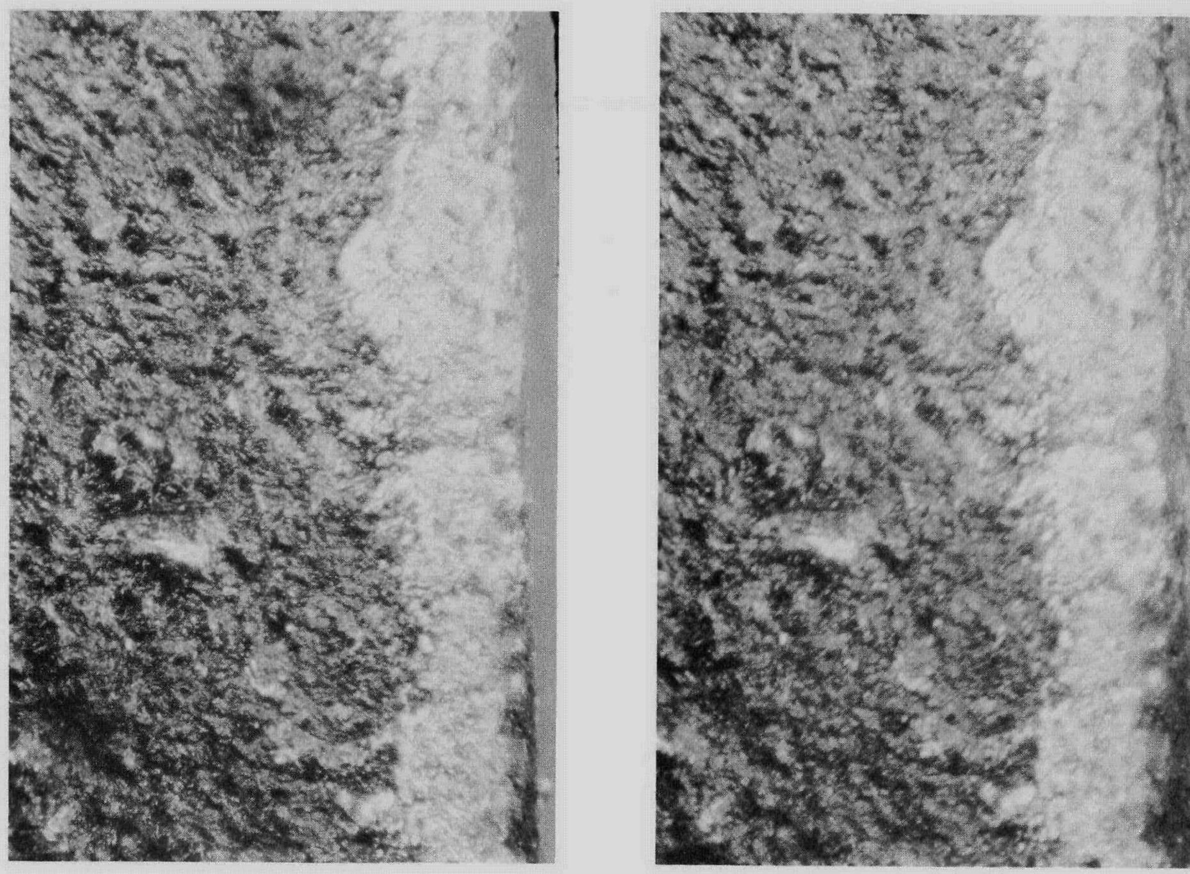
a)

b)



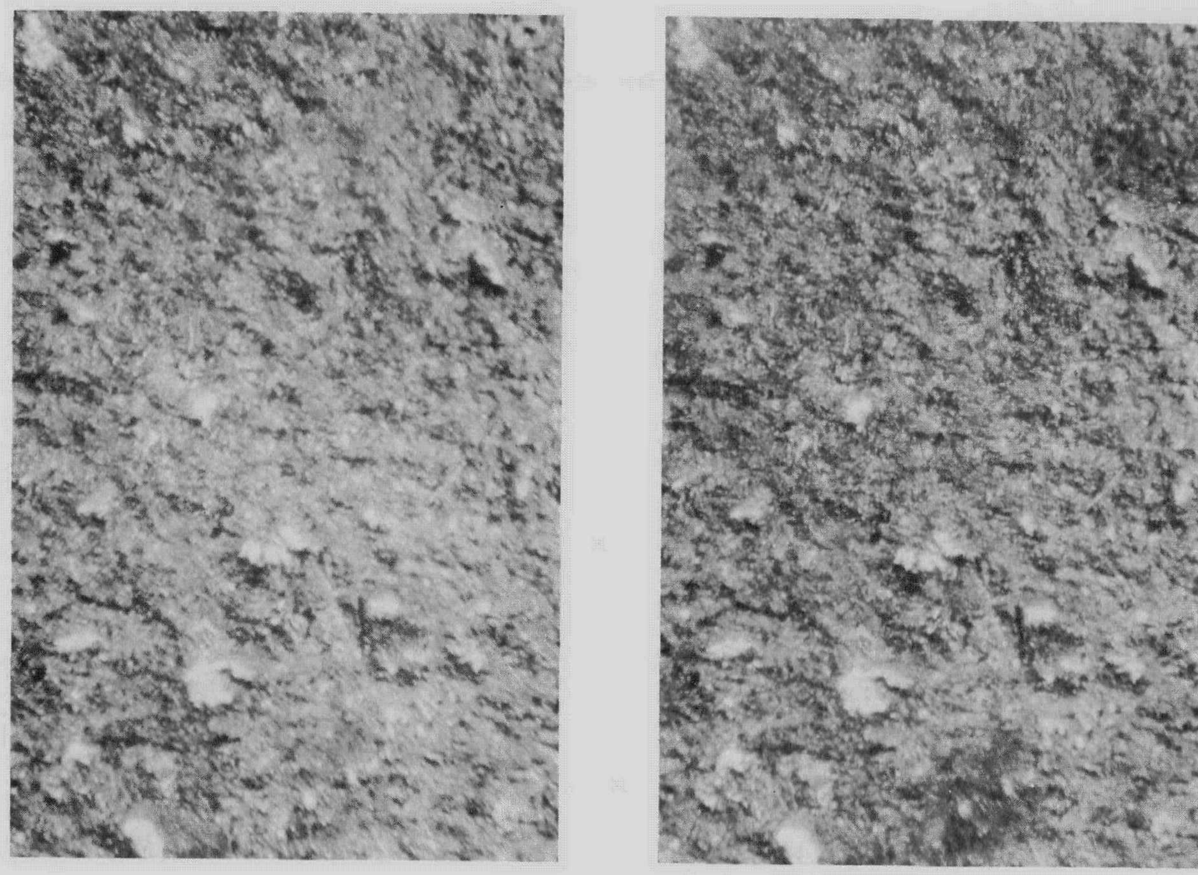
16324, 16327 1X

Figure 7. Views of Both Sides of Flat Plate No. 5 With Sheath Removed (Beta-annealed material)



17249 12X

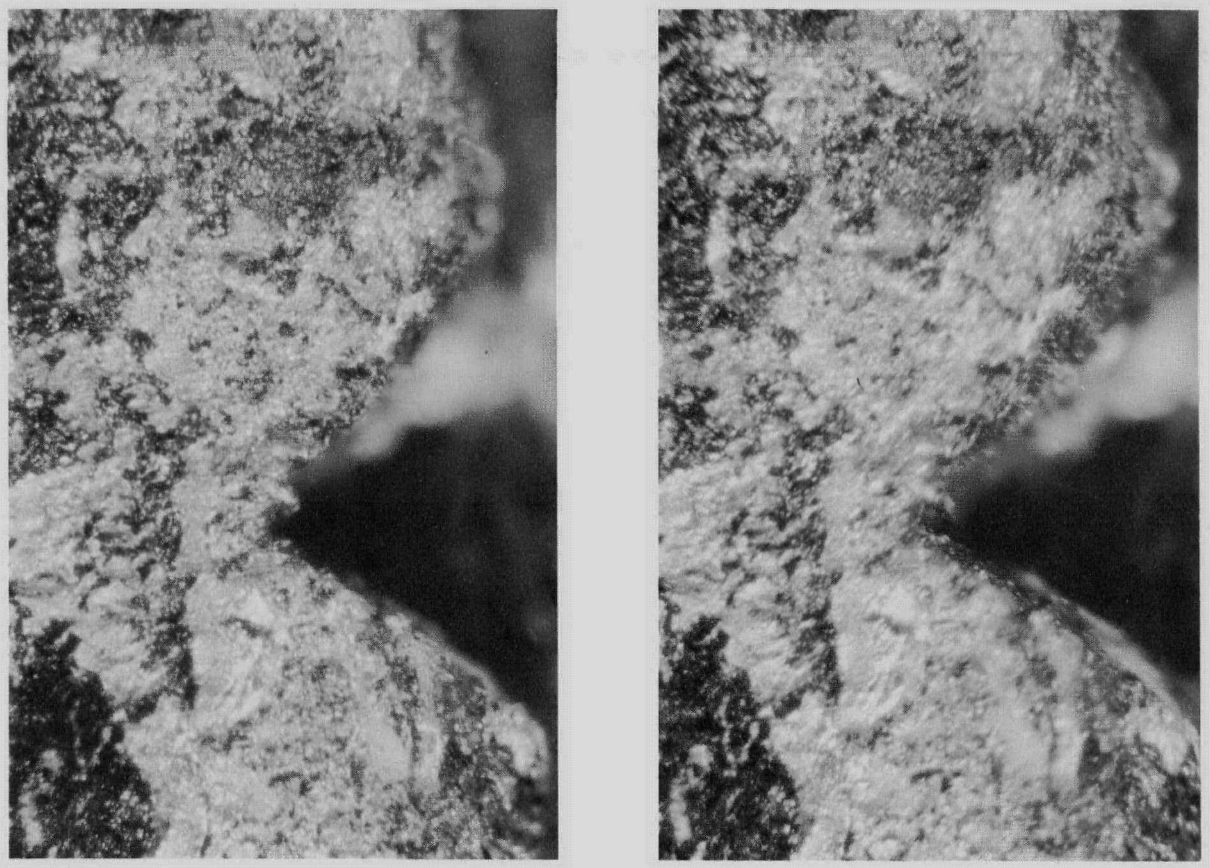
Figure 8. Magnified View of Plate No. 5 at Edge. Raised edge, rough surface, and adhering aluminum are seen in this stereo.



17251

12X

Figure 9. Typical View of Surface near Middle of Plate No. 5. Note evidence of poor contact between sheath and core.

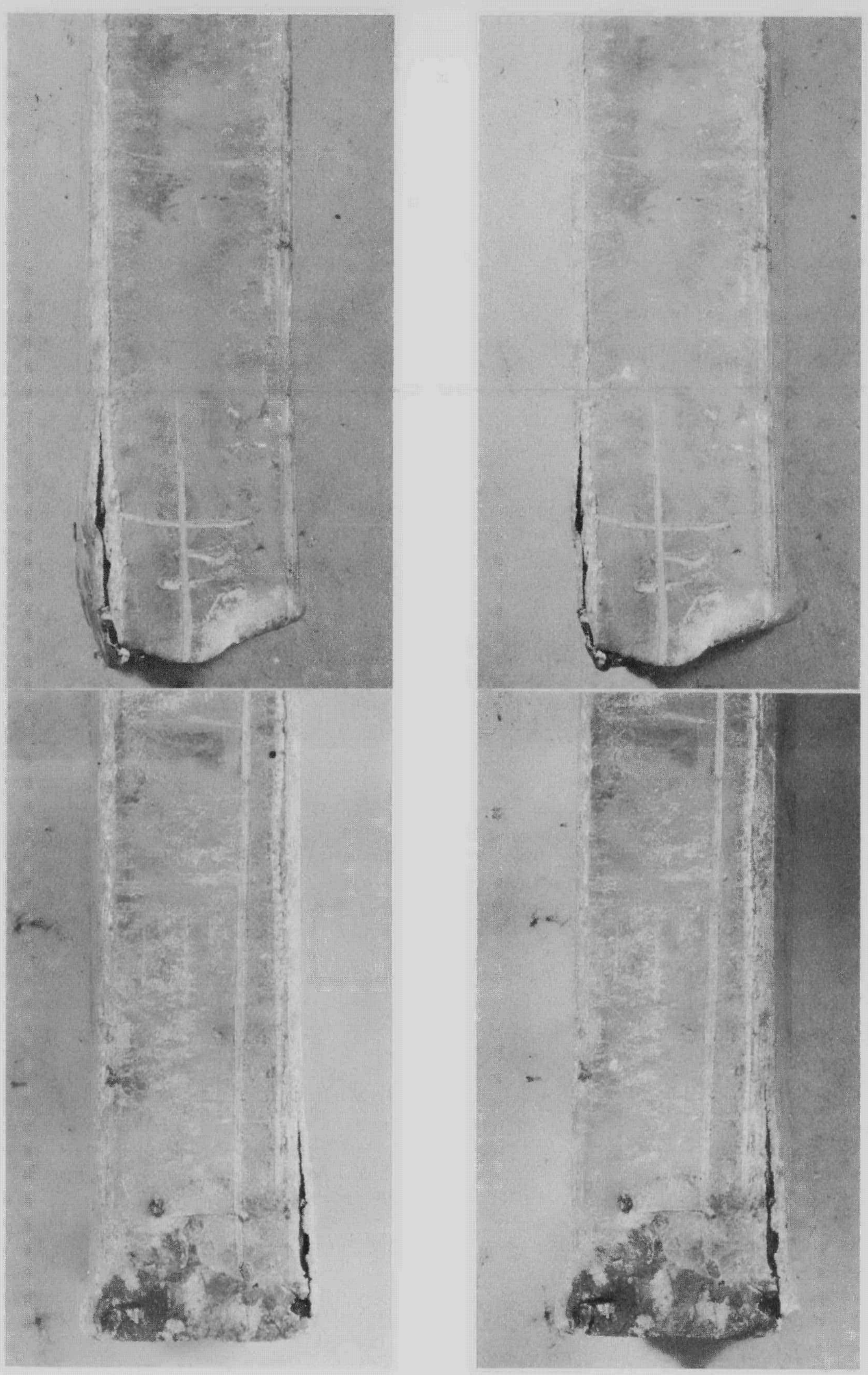


17253 34X

Figure 10. Greatly Enlarged View of Aluminum Fused to Edge of Plate No. 5 at Fiducial Notch

a)

b)

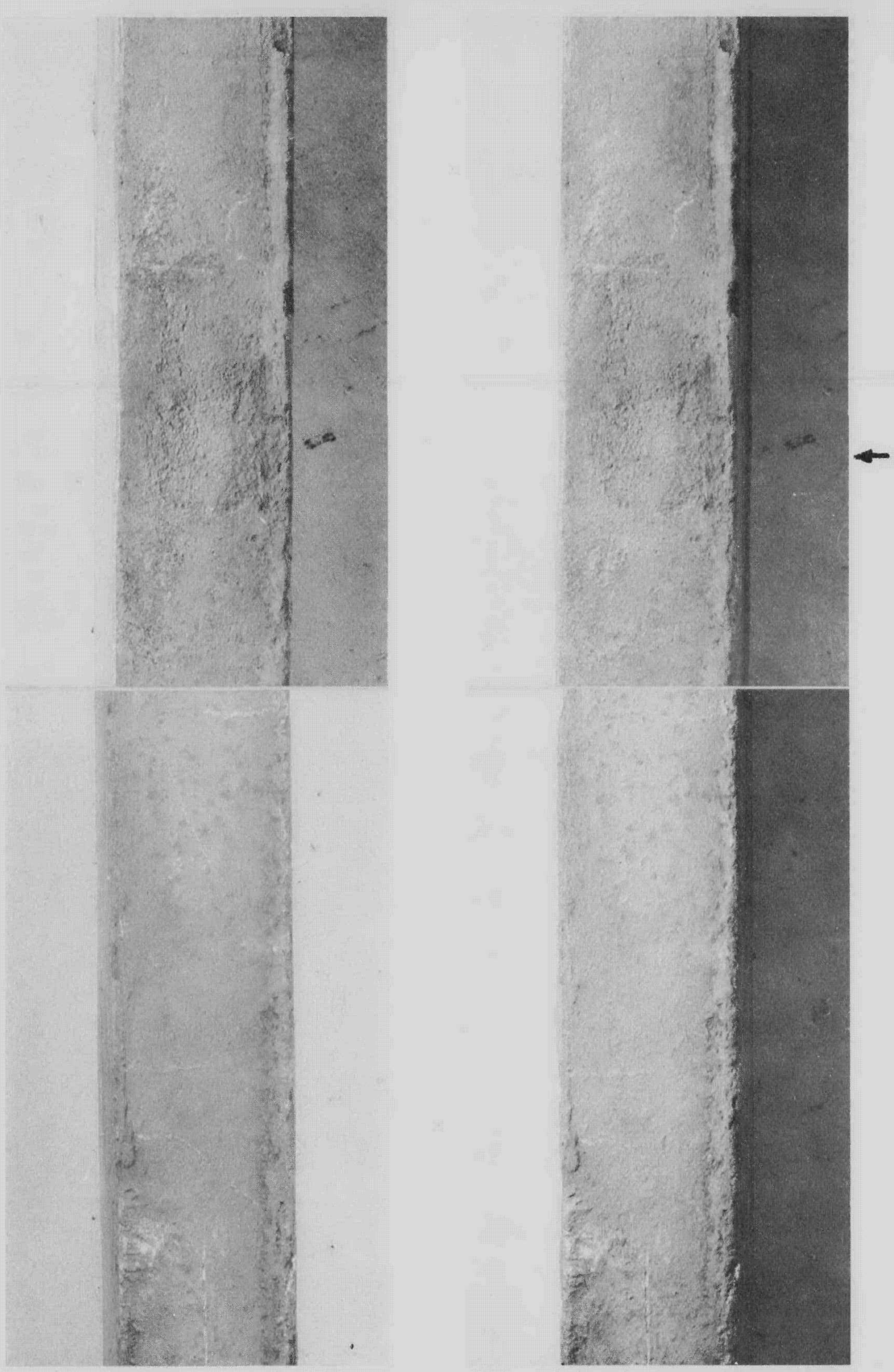


16320, 16321 1X

Figure 11. Views of Sheathed Square Rod at Sheared End (As received)

a)

b)

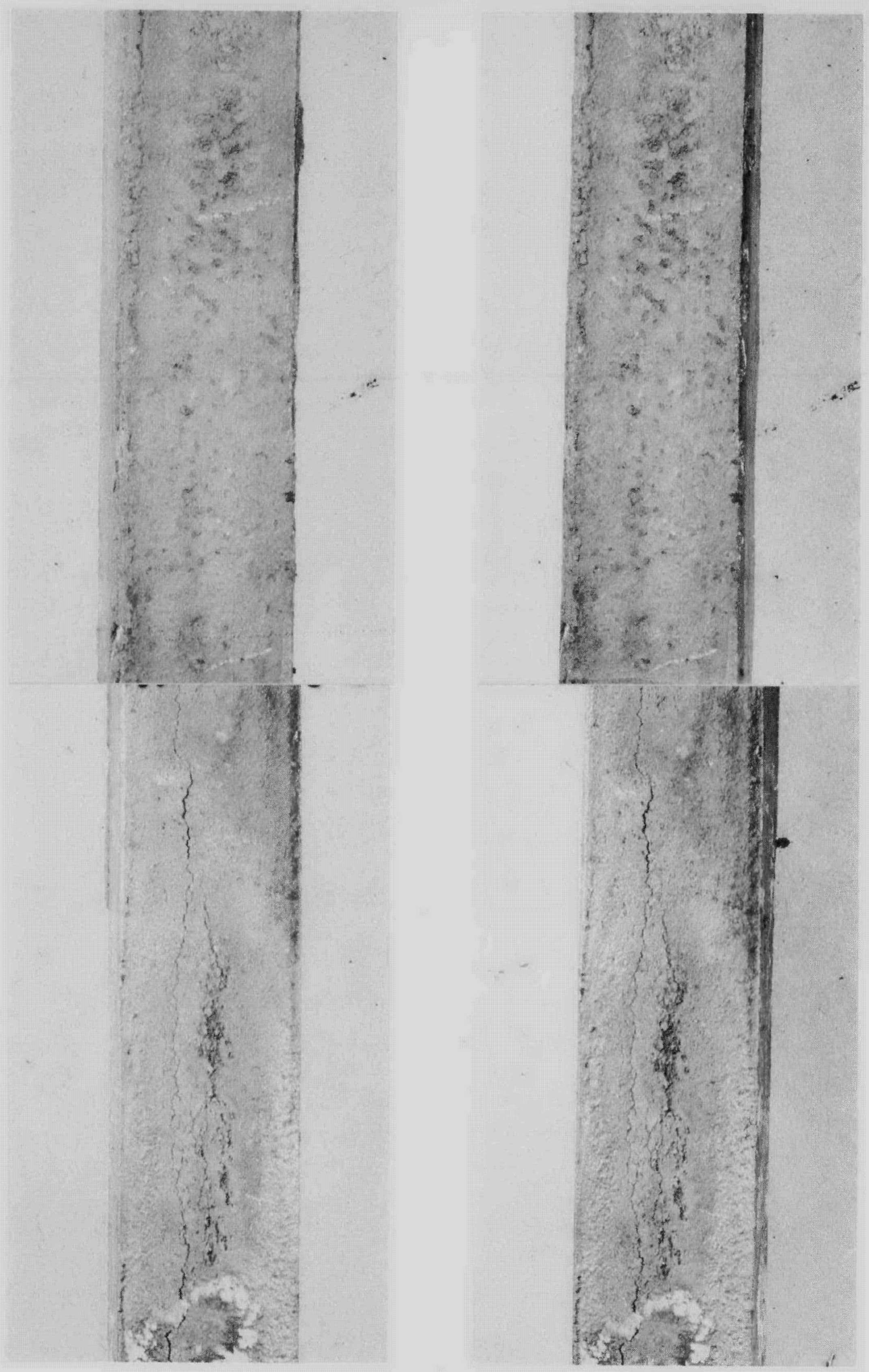


16318, 16322 1X

Figure 12. Views of Half the Perimeter of Square Rod after Removal of Sheath. Note corrosion damage to edges and surfaces.

c)

d)



16326, 16323 1X

Figure 13. Views of Other Half Perimeter of Square Rod. Note jagged longitudinal cracks along one surface.