OAK RIDGE NATIONAL LABORATORY LIBRARIES

3 4456 0550249 9

ORNL-TM-4561

Cy 39

PLUTONIUM-BERYLLIUM SOURCE SAFETY TESTING PROGRAM

R. G. Niemeyer
N. C. Bradley

OAK RIDGE NATIONAL LABORATORY
CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION
LIBRARY LOAN COPY
DO NOT TRANSFER TO ANOTHER PERSON
If you wish someone else to see this document, send in name with document and the library will arrange a loan.

UCN-7969
(3 3-67)

OAK RIDGE NATIONAL LABORATORY
OPERATED BY UNION CARBIDE CORPORATION • FOR THE U.S. ATOMIC ENERGY COMMISSION

Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22151
Price: Printed Copy $5.45; Microfiche $1.45

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

ORNL-TM-4561
UC-23 — Radioisotope and
Radiation Applications

Contract No. W-7405-eng-26

ISOTOPES DEVELOPMENT CENTER

PLUTONIUM-BERYLLIUM SOURCE SAFETY TESTING PROGRAM

R. G. Niemeyer
N. C. Bradley

MAY 1974

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37830
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

OAK RIDGE NATIONAL LABORATORY LIBRARIES

3 4456 0550249 9

CONTENTS

	Page
ABSTRACT	1
INTRODUCTION	1
SOURCE DESCRIPTION	2
NONDESTRUCTIVE TESTING	2
Photography and Visual Inspection	2
Surface Contamination Test	5
Leak Tests	5
Dimensional Inspection	6
Radiography	8
DESTRUCTIVE TESTING	8
Gas Sampling	9
Sectioning	13
Metallographic Examination	13
SUMMARY OF NONDESTRUCTIVE TESTS	15
SUMMARY OF DESTRUCTIVE TESTS	16
CONCLUSIONS	16
RECOMMENDATIONS	18
REFERENCES	20
APPENDIX A	21

PLUTONIUM-BERYLLIUM SOURCE SAFETY TESTING PROGRAM

R. G. Niemeyer and N. C. Bradley

ABSTRACT

Some representative commercially fabricated Pu-Be neutron sources which had been in service for periods greater than ten years were examined. Three of the five Pu-Be sources which were both destructively and nondestructively tested had weld zone deficiences such as leaks, weld inclusions, excessive roll-over and grain growth, and marginal weld penetration. Due to these deficiencies the tantalum inner capsule cannot be considered a reliable sealed source container. This deficiency does not necessarily reduce the containment reliability of the sources because the outer stainless steel capsule is the primary containment capsule. A recommendation is made to leak test all sources and recall those that leak for reencapsulation or disposal. An improved quality assurance program is recommended for future sources.

INTRODUCTION

Encapsulated neutron sources containing the intermetallic compound $^{239}PuBe_{13}$ are used in a number of applications such as oil well logging and nondestructive testing procedures. Potentially these sources could remain in use for many decades because of the long half-life of plutonium-239; however, their life may be limited by environmental effects which degrade the structural integrity of the source encapsulation. Degradation could occur from external corrosion, reduction of mechanical strength properties due to long-term exposure to neutrons, incompatibility between the source compound and encapsulating materials, and from stresses caused by pressure.

Since some of these sources have been in service nearly 20 years, it was decided to examine in detail a few representative Pu-Be sources which had been in actual use for several years. The objectives of the examination were (1) to evaluate the condition of the source capsules after several years of service, (2) to evaluate the quality of the original fabrication of the source capsules, (3) to evaluate the effectiveness of nondestructive test procedures in detecting potential containment problems in the source capsules, and (4) to determine if the frequency and/or type of inspection should be changed from the presently required 6-month wipe tests.

The six sources tested were selected from AEC listings of Pu-Be sources so as to meet the criteria that the sources be old, from a variety of uses and users, and be fabricated by manufacturers other than USAEC Laboratories. A study of Pu-Be sources fabricated by Mound Laboratory has been published.[1-3]

Five of the sources were subjected to both nondestructive and destructive examination; one source was subjected to nondestructive examination only. The sources selected for testing are listed in Table 1.

Table 1. Pu-Be Neutron Sources Tested

Source Number	Fabrication Date	Pu-Be Weight (g)	Source Use
N-PuBe-292	4-16-62	32	Not used
N-PuBe-293	4-16-62	32	Not used
N-PuBe-294	3-23-62	80	Source of neutrons under water
N-PuBe-295	8-3-62	100	Well logging
N-PuBe-296	11-17-61	80	Well logging
N-PuBe-297	6-21-61	160	Fission counter testing

The authors wish to acknowledge the valuable assistance of B. C. Leslie of the ORNL Metals and Ceramics Division who performed the metalographic examination and participated in interpreting the results.

SOURCE DESCRIPTION

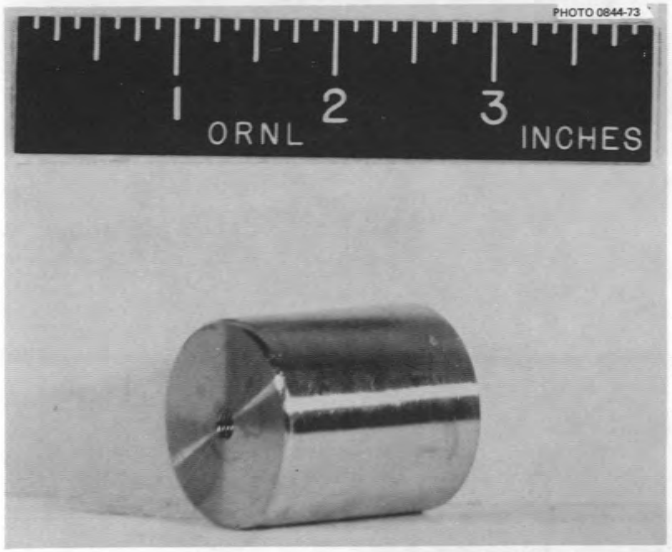
The sources were doubly encapsulated. The inner capsules, containing the intermetallic compound $PuBe_{13}$, were tantalum; the outer capsules were type 304 stainless steel except for one (N-PuBe-296), which was made to special order using a Vega tool-steel outer capsule. Both inner and outer capsules were sealed by inert-gas-shielded, tungsten-electrode arc (GTA) welding. Source contents ranged from 32 to 160 g of ^{239}Pu. Source N-PuBe-294 contained two inner tantalum capsules end-to-end within the outer stainless steel capsule. (The inner tantalum capsule at the threaded tool posthole end of the stainless steel capsule is designated capsule A and the other, B.)

NONDESTRUCTIVE TESTING

The six Pu-Be neutron sources were subjected to the following nondestructive testing: photography and visual examination, surface contamination test, leak test, dimensional inspection, and radiography. The test procedures, equipment, and test results are given below.

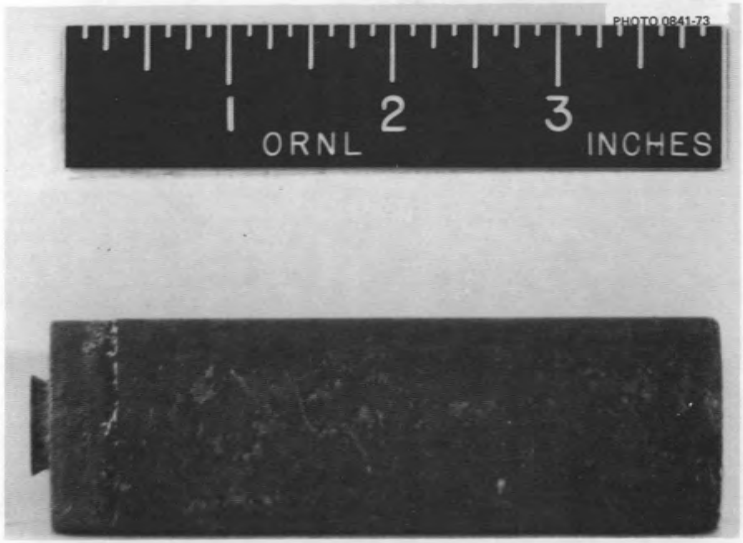
Photography and Visual Inspection

The sources were photographed in the "as-received" condition. They were placed on a lighted table in a hot cell and photographed through the cell window using a 285-mm lens on a Nikon camera which was adapted with a magnifying back for use with 4- by 5-in. film. Photographs of the sources are shown in Figs. 1-2. The sources were initially inspected without magnification and were then inspected with a modified 10X monocular. A crack in the weld of source N-PuBe-294 was viewed using a Questar telescope at 80X.

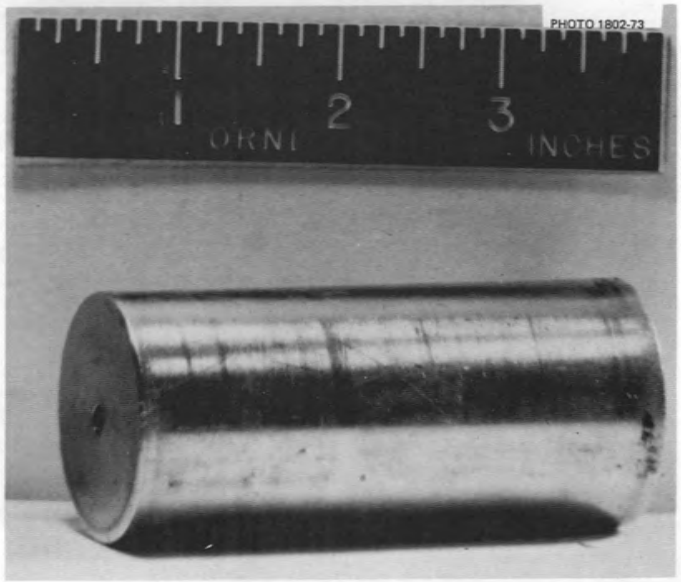


(a) N-PuBe-292 (N-PuBe-293 same)

(b) N-PuBe-295



(c) N-PuBe-296



(d) N-PuBe-297

Fig. 1. Pu-Be Sources in "As-Received Condition.

(a) Enclosed in Lead Shield

(b) With Lead Shield Removed

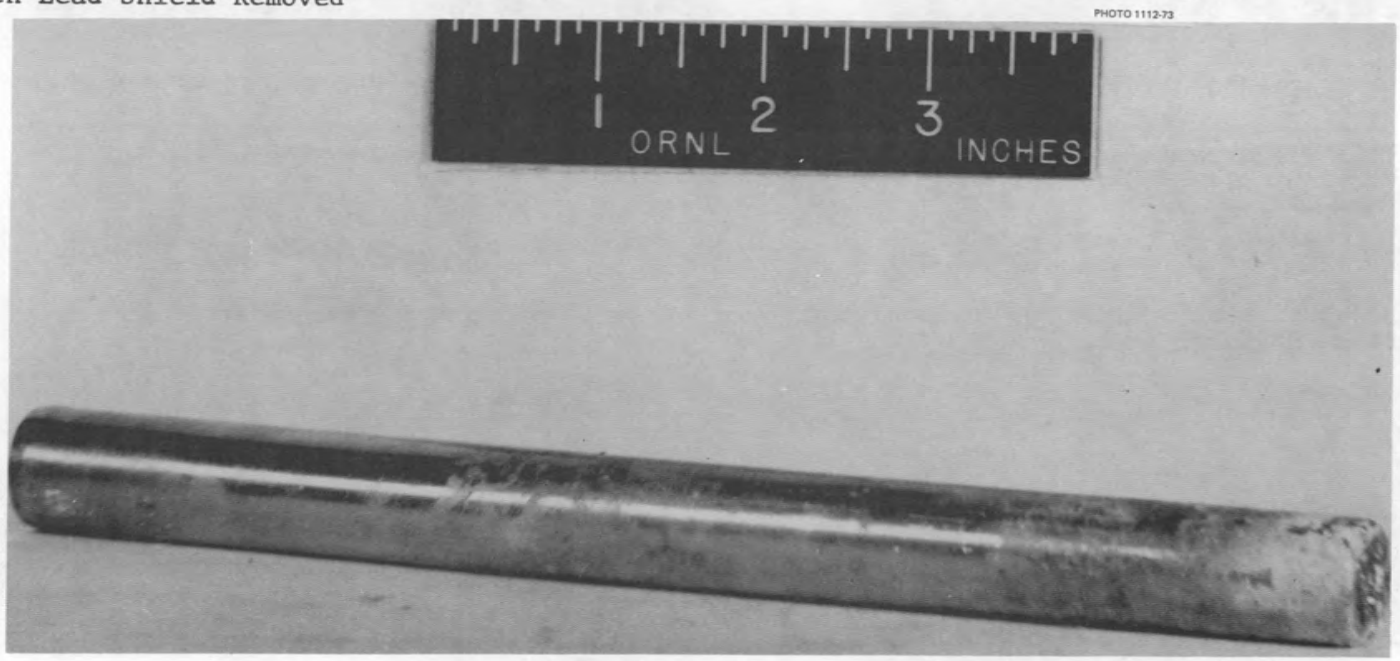


Fig. 2. N-PuBe-294 Source in "As-Received" Condition.

Sources N-PuBe-292 and N-PuBe-293 were bright and shiny with no apparent defects, as might be expected for sources that had been in storage and never used. Source N-PuBe-297 was bright, clean, and lightly scratched in random areas. Source N-PuBe-294 was dull and coated in some areas with a white lead oxide from the lead capsule shield. A crack was noted in the weld at the top end cap in an area that showed welding irregularities. Source N-PuBe-295 appeared dull with minor dents and scratches on the surface. Source N-PuBe-296, which was encapsulated in Vega steel, appeared to be rusted all over with traces of yellow paint still adhering in some areas.

Surface Contamination Test

The sources were tested for surface contamination in the "as-received" condition by wiping all surfaces with dry 4-cm-diam filter paper wipes. Each wipe was then analyzed for alpha and beta-gamma activity by determining the radiation emission rate from the entire surface of the wipe. The radiation emission rates listed in Table 2 show that the surface contamination from the external surfaces of the sources is low.

Table 2. Surface Contamination Test Results

Source Number	Radiation Emission Rates (dis/min)	
	Alpha	Beta-Gamma
N-PuBe-292	<30[a]	<500[a]
N-PuBe-293	<30[a]	<500[a]
N-PuBe-294	3[b]	0
	9[c]	
N-PuBe-295	6	140
N-PuBe-296	0	300
N-PuBe-297	0	260

[a]Indicates no significant surface contamination was found.

[b]Surface contamination on lead shield.

[c]Surface contamination on stainless steel capsule.

Leak Tests

Prior to being leak tested, the sources were cleaned to remove paraffin, grease, and dirt by scrubbing them with wipes wetted with trichloroethylene. This cleaning procedure removed foreign matter that might change the leak detection characteristics or the dimensional measurements.

The sources were leak tested by using the vacuum leak test[4] system shown in Fig. 3. The source is placed in a chamber and submerged in ethylene glycol. The chamber is slowly evacuated to a vacuum of 25 in. Hg as the source is viewed; a stream of bubbles arising from a point on

the surface indicates a positive leak. This test has been shown to be sensitive to a leak rate of 4×10^{-6} atm cm^3/sec.

The leak tests of all sources other than N-PuBe-294 were negative. Source N-PuBe-294 was received enclosed in a two-piece threaded lead shield as shown in Fig. 2(a). The source was removed from the lead shield by sawing through the shield after attempts to disassemble it at the threaded joint failed. An x ray of the source was used to locate a suitable place to cut the lead capsule without damaging the source.

The leak test of source N-PuBe-294 gave an indication of gross leakage from the welded end. Inspection of the leak area using a 10X monocular and an 80X Questar telescope revealed a crack in the weld area on the end cap.

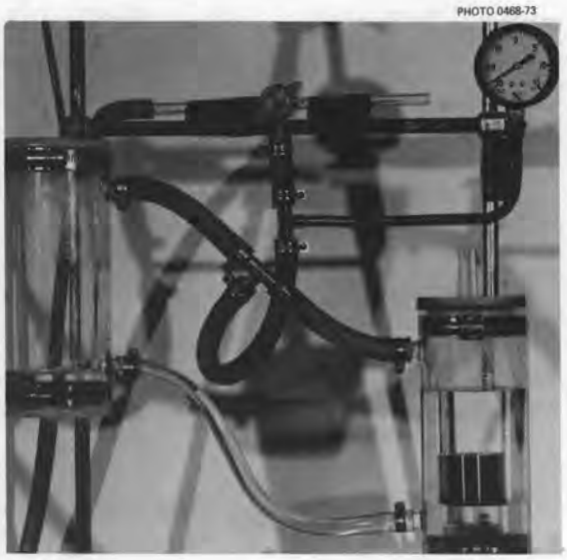


Fig. 3. Vacuum Leak Test System.

Dimensional Inspection

The dimensional inspection system shown in Fig. 4 was used to dimension the sources. A steel inspection table was set up and two dial indicators capable of measuring to 0.001 in. were set at two different heights. Two precision gage blocks, 1 × 2 × 3 in., were provided to preset and test the dial indicators. The positions of measurements made on each source are shown in Fig. 5. The source diameter was measured at the top (welded end cap), middle, and bottom of each source on the x and y planes which are 90° apart. The length of the sources was also measured.

The dimensional measurements of the sources are shown in Table 3. Identical measurements of the capsule diameters at all measurement points on a particular source indicate a uniform circular capsule

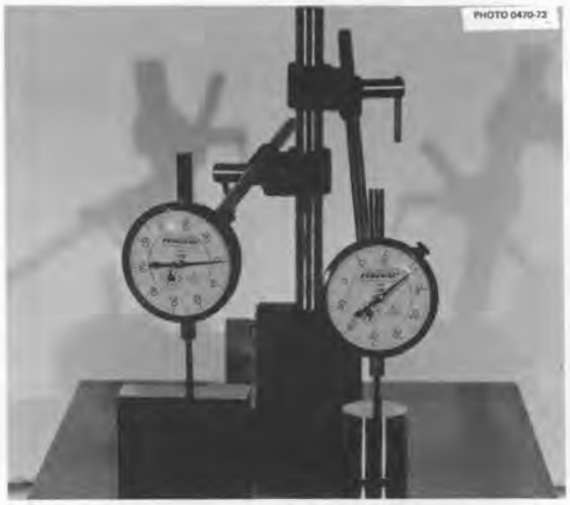


Fig. 4. Dimensional Inspection System.

that has not been deformed by usage and/or internal gas pressure buildup. An increase in gas pressure within a capsule would be expected to cause a swelling of the capsule, with the middle measurements increased a significant amount greater than the end measurements. The measurements in Table 3 show that some sources are not symmetrical. There are no measurements that indicate swelling in any of the sources; however, dents and irregularities from handling and/or fabrication irregularities were visible.

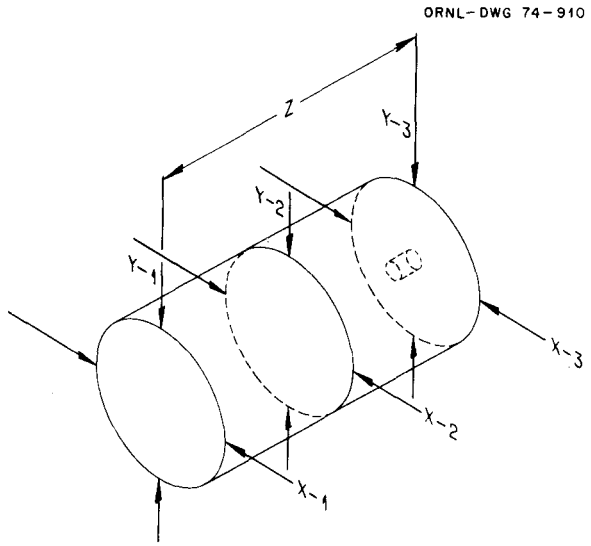


Fig. 5. Schematic of Dimension Measurements

Table 3. Dimensional Measurements

Source Number	x Diameter (in.)			y Diameter (in.)			Length (in.)
	Top	Middle	Bottom	Top	Middle	Bottom	
N-PuBe-292	1.312	1.3135	1.314	1.314	1.3145	1.3145	1.512
N-PuBe-293	1.311	1.311	1.311	1.3115	1.3115	1.3115	1.510
N-PuBe-294	0.755	0.755 0.753[a]	0.752	0.755	0.758 0.752[a]	0.752	8.875
N-PuBe-295	1.307	1.311	1.308	1.310	1.310	1.310	3.317
N-PuBe-296	1.295	1.295	1.295	1.295	1.295	1.295	41.00
N-PuBe-297	1.551	1.552	1.553	1.551	1.553	1.558	

[a]Source contained two inner capsules; middle measurements were taken in the middle of the inner capsules.

In addition to the above sources, eight sources currently in use in a production-line-type nondestructive testing operation were obtained temporarily from the user and were given a detailed dimensional inspection; no other tests were performed on these sources. These sources were returned to the user, and it is planned to periodically reinspect the sources when they can be made available during maintenance shutdowns. The test results are given in Table 4. Only one of the sources had a diameter measurement that was 0.003 in. greater than the corresponding minimum source diameter, indicating that there was no deformation due to internal gas pressure buildup.

Table 4. Dimensions of Neutron Sources Used in NDT Operation

Source No.	Diameter (in.) X-1	X-2	X-3	Y-1	Y-2	Y-3	Length (in.) Z
1120J2	1.552	1.553	1.553	1.551	1.552	1.553	2.585
1120J3	1.550	1.552	1.552	1.550	1.550	1.552	2.590
1120J4	1.551	1.552	1.552	1.550	1.551	1.552	2.589
1120J5	1.550	1.551	1.552	1.549	1.550	1.551	2.581
1120J6	1.550	1.552	1.553	1.549	1.550	1.552	2.587
1120J7	1.550	1.550	1.550	1.551	1.552	1.551	2.588
1120J8	1.551	1.552	1.552	1.552	1.552	1.552	2.585
1120J9	1.552	1.551	1.550	1.549	1.551	1.551	2.586

Radiography

The six sources were placed in contact with type M industrial x-ray film packets and radiographed with a ^{137}Cs source. Two views were made of each source on the x and y planes as shown in Fig. 5. The x rays were viewed on a Picker x-ray viewer using a magnifier.

The radiographs showed some welding insufficiencies which were possibly caused by misalignment of the welding torch and the capsule end-cap joint and/or insufficient penetration. Source N-PuBe-294 shows a broken weld area over most of the welded end cap of the top ("A") tantalum capsule (this source contained two inner tantalum source capsules end-to-end). The radiographs did not detect the crack in the stainless steel-welded end cap of this source where the gross leak was observed in the vacuum leak test. The radiographs of source N-PuBe-295 showed that in one area the weld in the bottom end cap (one without the tool post) of the stainless steel capsule has insufficient penetration (penetration was approximately one-half of the capsule wall thickness).

Source N-PuBe-296 shows an area of the weld zone on the side weld joint of the outer Vega steel capsule that appears to have insufficient weld penetration. The stress-relief groove in the end cap shows up strongly and partially masks the area, making interpretation difficult.

DESTRUCTIVE TESTING

In the destructive testing program, five of the sources were first punctured to release the contained gases for pressure measurements and gas composition. Four of these sources were then sectioned to obtain specimens of the weld areas for metallurgical examination.

Gas Sampling

The quantities of gas contained within the sources were measured and sampled using the equipment shown in Fig. 6. Figure 7 shows a schematic of the equipment. The puncture chamber was constructed to accommodate the various sizes of sources, and solid aluminum holders were drilled to fit each size of source and to hold it in the proper position for puncturing. The holders also reduced the volume of the chamber so that increased sensitivity of the pressure measurements could be obtained. The holders containing the sources were inserted into the chamber from the bottom, and an elevator fabricated from a bellows-sealed vacuum valve stem unit was used to raise and lower the source to the proper level for puncturing. The puncture point was made of hardened tool steel and was fabricated with a small curved groove from the point to its base. The puncture point was fastened to a bellows-sealed vacuum valve stem drive, which was actuated by a ratchet drive handle to insert and withdraw the point.

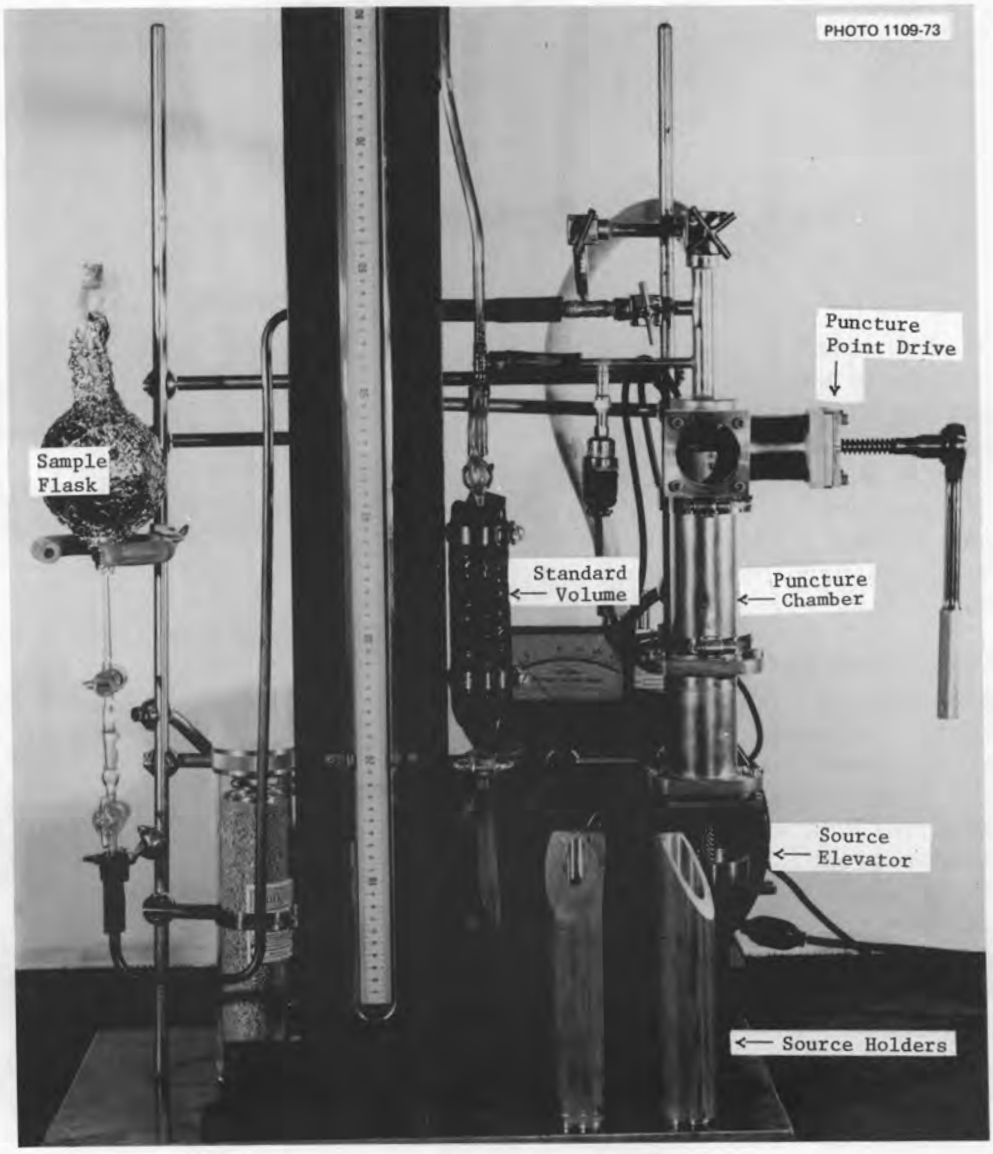


Fig. 6. Equipment for Puncturing Sources and Measuring and Sampling Contained Gas

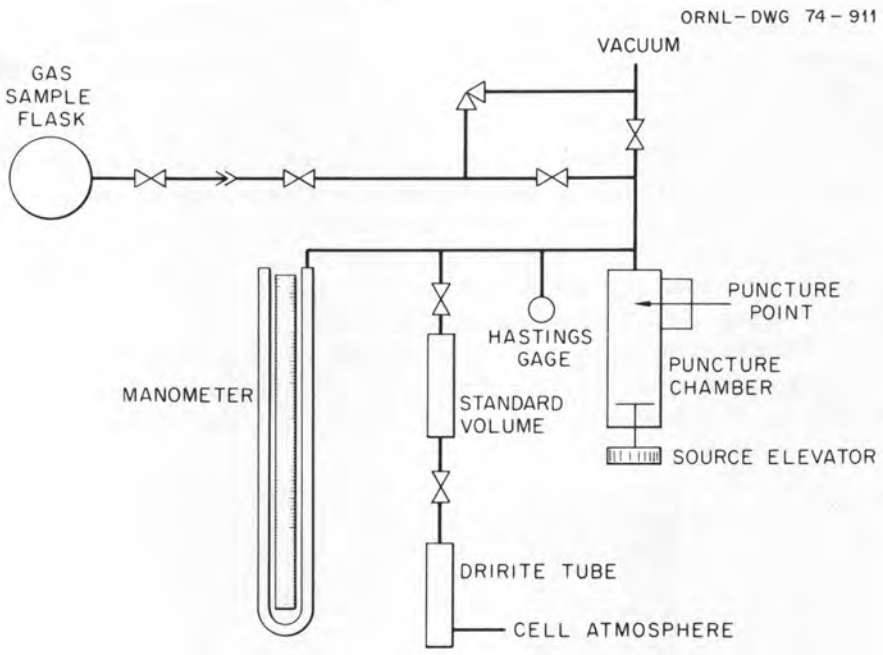


Fig. 7. Schematic of Source Puncturing, Gas Measuring, and Sampling Equipment

The gas contained in the outer source capsule was released by penetrating the outer capsule at the point between the bottom surface of the outer capsule end cap and the top surface of the inner capsule end cap. The inner capsule was punctured by penetrating both capsule walls.

The gas was sampled by allowing the released gas to expand from the puncture chamber into an evacuated 500-ml gas sample flask having a glass connector and breakseal for connection to the mass spectrographic analyzer. The gas sample flask was removed from the puncture equipment and sealed to prevent contamination of the sample due to inleakage (tubing sealer shown in Fig. 8).

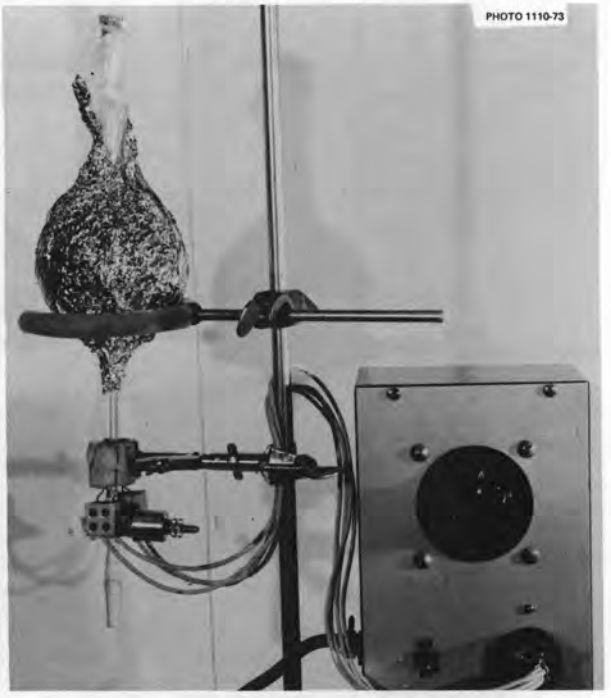


Fig. 8. Electrically Heated Glass Tubing Sealer.

The following procedure was used to measure the source gas volume and pressure in each source and to obtain gas samples:

1. The source was placed into a source holder and then into the puncture chamber.

2. The system was evacuated to <10-μm Hg and leak rate tested.

3. The system was vented to cell atmosphere through a tube containing a desiccant, and the pressure was recorded.

4. The standard volume chamber was isolated and the system evacuated to 10-μm Hg.

5. The puncture point was fully withdrawn and the capsule holder-elevator completely lowered to a zero point for all manometer readings below atmosphere. The gas in the standard volume chamber was released into the puncture chamber and the manometer reading was recorded.

6. The standard volume chamber was isolated, and the puncture chamber evacuated and isolated. The capsule was positioned and punctured. The puncture point and elevator were withdrawn to zero position and the manometer pressure recorded.

7. The released gas in the puncture chamber was valved to the evacuated sample flask, which was then isolated, removed, and sealed.

8. The puncture system was then vented to cell atmosphere through the standard volume chamber, and the pressure was recorded.

9. The standard volume chamber was isolated, and the puncture chamber evacuated and isolated. The standard volume of gas was released into the puncture chamber and the manometer reading recorded.

10. The gas volume in the source was determined by difference in system pressure measurements before and after puncture. By using the released gas pressure and source volume, the pressure of the contained gas in the source was calculated.

11. The inner capsule gas pressure and sample were obtained by returning to step 6 and repeating the procedure.

Five sources were punctured for gas pressure determinations, and samples of gas were taken where sufficient amounts of gas were present. The exceptions were the outer stainless steel capsule of N-PuBe-294, which had a gross leak, and the Vega steel capsule of N-PuBe-296, which could not be punctured. The inner tantalum capsule of N-PuBe-296 was removed from the outer Vega steel capsule and punctured, and the gas pressure was measured. Table 5 lists the calculated internal gas pressures in the source capsules. All pressures were found to be less than atmospheric.

Table 5. Source Gas Pressures

Source Number	Capsule Material	Pressure (psia)
N-PuBe-292	Stainless steel	11.9
	Tantalum	0.7
N-PuBe-293	Stainless steel	0.2
	Tantalum	0.1
N-PuBe-294	Stainless steel	Leaker
	Tantalum, upper	1.4
	Tantalum, lower	0.16
N-PuBe-295	Stainless steel	8.4
	Tantalum	None detected
N-PuBe-296	Vega steel	Not punctured
	Tantalum	0.12

Sufficient gas was available in three of the sources for determining gas composition; the results are given in Table 6.

Table 6. Mass Spectrometer Analysis

Gas	N-PuBe-292		N-PuBe-295	N-PuBe-294	
	Outer (%)	Inner (%)	Outer (%)	Inner A (%)	Inner B (%)
Hydrogen	7.9	2.6	0.1	1.4	15.8
Methane		2.0		0.5	7.8
Hydrocarbon		3.0	<0.1	0.2	6.1
Nitrogen	81.8	61.7	77.9	77.1	54.8
Oxygen	1.4	25.6	20.7	18.9	10.2
Argon	7.1	2.6	1.1	1.2	0.7
Carbon dioxide	1.8	2.3	0.1	1.0	4.5

The analyses indicate that the major portion of all the sampled gases was air. Source N-PuBe-294 inner capsule B (lower capsule) contained quantities of hydrogen, methane, hydrocarbon, and carbon dioxide, which totaled 34% of the measured gases. This composition indicates that a small quantity of some hydrocarbon was present as a contaminant. The gas analysis for source N-PuBe-292 inner and outer capsules showed indications of 10% hydrocarbon gas products per sample.

Sectioning

The manipulator cell used in sectioning the sources was equipped for handling high levels of alpha contamination and for inert gas operation to preclude the possibility of burning plutonium dust during the sawing operations. Shielding against the neutrons emitted from the sources was provided by 2-ft-thick water shielding tanks.

The cell was made inert with argon to a level below 500 ppm of oxygen prior to sectioning the sources. The rotary saw shown in Fig. 9 was then used to cut off the end caps of the sources. These end caps were then cut in half, and in some cases quartered, to obtain the metallographic specimens. Nine specimens taken from four sources were chosen for examination. The following is a list of the samples submitted for metallographic examination.

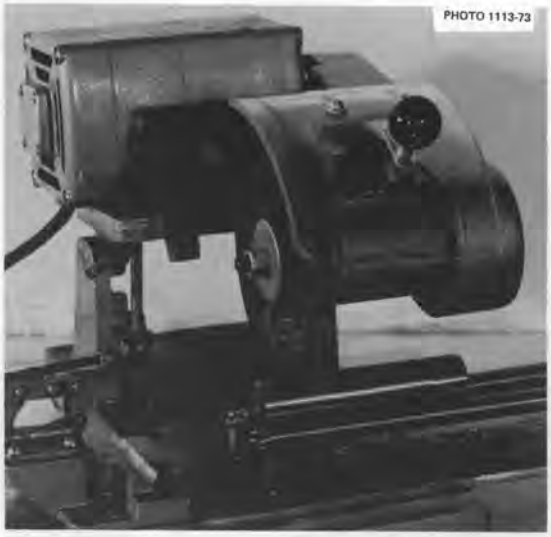


Fig. 9. Rotary Saw in Alpha Cell

Source Number	Sample Section	Material
N-PuBe-292	Weld end-cap inner capsule	Tantalum
N-PuBe-294	Bottom end, inner and outer capsules	Tantalum and stainless steel
N-PuBe-294	Top end, outer capsule	Stainless steel
N-PuBe-295	Top end, outer capsule	Stainless steel
N-PuBe-295	Bottom end, inner and outer capsules	Tantalum and stainless steel
N-PuBe-296	Top end, inner capsule	Tantalum
N-PuBe-296	Top end, outer capsule	Vega steel

Metallographic Examination

The sectioned samples of the source capsules were prepared, examined, and photographed. The details of the inspection test results are presented in Appendix A. A summary of the examination relating to capsule integrity for each source is presented below, including a list of possible losses in source integrity caused by various defects.

Defects Which Reduce Source Integrity

Defects	Potential results of defect
1) Grain boundary oxidation	Weakens affected area and tends to cause embrittlement

Defects	Potential results of defect
2) Insufficient weld penetration	Weld does not have the design strength
3) Capsule leaks	Result in possible leak of activity especially if vibrated or immersed in liquid
4) Excessive grain growth	Weakens weld zone metal and tends to cause embrittlement
5) Weld roll-over	Frequently results in low weld penetration (See defect 2 above)
6) External corrosion	Reduces strength of capsule in area affected and may result in a leaking capsule
7) Weld cracks, voids, and occlusions	Cause weak areas in the weld zone

Source N-PuBe-292, Welded Tantalum End Cap, Sample 1

This sample had a nearly satisfactory weld although the weld penetration* (∿70% of the capsule wall thickness) was somewhat less than desirable. The grain size was not as large as in several of the samples described below. The weld zone appears to be free of cracks and voids.

Source N-PuBe-294, Sample 1, Bottom Section, Inner and Outer Capsules

The sample of the outer stainless steel capsule weld zone indicates a satisfactory weld. There were no visible cracks or voids in the weld zone and weld penetration was ∿95% of the capsule wall thickness.

The sample of the inner tantalum weld zone showed a crack penetrating through the weld zone, making it possible for fuel to leak out of the inner capsule. Excessive grain growth, which weakens the weld zone metal and tends to cause embrittlement, occurred during welding. It appears that the original weld penetration was only one grain thick (∿30% of the capsule wall thickness). Low penetration such as this drastically reduces the strength of the weld metal. Amorphous material observed between the inner capsule wall and the end cap may have contributed to the failure of this weld.

Source N-PuBe-294, Sample 2, Stainless Steel Outer Capsule

This sample of the outer stainless steel weld zone indicates that an apparent welding alignment problem occurred in which the weld was not centered over the end cap-capsule interface. This resulted in excessive weld roll-over and low penetration (∿35%) and, therefore, in a weld with poor strength. An unidentified inclusion was trapped between the end

*Weld penetration is defined as the least distance from the weld joint pocket to the capsule wall (80% penetration is desired for capsule wall thicknesses up to 0.0625 in.).

cap and the weld metal which can also reduce weld joint strength by creating void areas in the weld zone.

Source N-PuBe-295, Sample 1, Outer Capsule Stainless Steel Weld

This sample indicates a satisfactory weld, free of cracks and voids, and having good penetration (∿100%).

Source N-PuBe-295, Sample 2, Bottom Tantalum and Stainless Steel Outer Capsules

The stainless steel outer capsule weld sample showed less than desirable weld penetration (∿60%), which means that the weld is below the desired strength.

The tantalum capsule weld had nearly satisfactory penetration (∿75%). The grains in the weld zone were quite large (a condition which weakens the weld zone metal and tends to cause embrittlement). There appears to be an oxide layer adhering to the outer surface of the capsule and internal oxide precipitation platelets, both of which can weaken the weld joint and cause embrittlement of the metal.

Source N-PuBe-296, Top Weld, Tantalum Inner Capsule, Sample 1

This sample indicates that a nearly satisfactory weld was obtained. The grains are larger than desirable and porous metallic material was present on the inside surface of the end cap (conditions which can cause low strength and embrittlement in the weld zone). Weld penetration was 100%.

Source N-PuBe-296, Sample 2, Top Vega Steel Weld, Outer Capsule

In this sample the weld nugget is not centered over the capsule end-cap interface, giving much less than the desired weld penetration (∿65%) and desired weld strength.

SUMMARY OF NONDESTRUCTIVE TESTS

The visual inspection of the sources in the "as received" condition revealed that source N-PuBe-294, which had been used under water, was coated in some areas with white lead oxide which probably came from the lead capsule shielding. This source also had a visible crack in the top end-cap weld in an area that had visible welding irregularities. Source N-PuBe-296, which had been used in well logging had a Vega steel outer capsule which appeared to be rusted all over and to have traces of yellow paint. The remaining four sources had essentially clean surfaces with no visible defects.

The wipe tests of the sources in the "as received" condition were near background and did not identify any source as having a leak. The vacuum leak test was used for leak testing the sources. For doubly encapsulated sources, this test will detect leaks in the outer capsule only. A gross

leak in the outer capsule of source N-PuBe-294 was observed. The leak tests of the other five sources were negative.

The dimensional inspection indicated that the sources were only slightly out-of-round and that swelling had not occurred. This was also indicated in measurements of the eight sources obtained for inspection from a source user.

The radiographs showed some welding deficiencies, possibly caused by misalignment of the welding torch and the capsule end-cap joints and/or insufficient penetration. Source N-PuBe-294 had a broken weld area over most of the welded end cap of the "A" tantalum capsule (this source had two inner tantalum source capsules stacked end-to-end). The radiographs did not detect the crack in the stainless steel welded end cap that contained the gross leak observed in the vacuum leak test of source N-PuBe-294. Insufficient weld penetration was noted in some areas of sources N-PuBe-295 and -296 outer capsules.

SUMMARY OF DESTRUCTIVE TESTS

Internal pressures were observed to be less than atmospheric in the gas pressure measurements of the eight capsules which contained sufficient amounts of gas for measuring. Capsules for which the internal pressure could not be measured were the outer capsule of source N-PuBe-294, which had a gross leak, and the outer Vega steel capsule of source N-PuBe-296, which could not be punctured due to the hardness of the metal.

The metallography results show deficiencies in all of the tantalum weld zones--in one case, a crack was observed which penetrated completely through the weld zone. In most of the samples the tantalum showed excessive grain growth and oxidation near the weld zones, and weld penetration was somewhat less than desired. Alignment problems occurred in two cases where the bead was not centered over the end cap-capsule interface, causing excessive roll-over and reduction in weld penetration.

CONCLUSIONS

1.0 The quality control programs used in the fabrication of the six sources examined in this study were not adequate to detect defects which may result in the release of radioactive contamination. The supporting data for this conclusion are:

a. One out of six of the outer capsules (16.6%) had a leak in the weld area which was not detected by a wipe test but was detected by a vacuum leak test with the capsule submerged in ethylene glycol.

b. The location of the weld zone in relation to the end cap-capsule interface showed that the welding electrode was not centered in the case of two outer capsule welds.

c. Sufficient weld penetration was not obtained in two outer capsules.

d. Insufficient weld penetration, cracks, and large grain structure were observed in the weld zones of four inner capsules. Although a fully sealed inner capsule is not required by regulation, these defects indicate that welding procedures need to be improved.

e. One of six inner capsules contained a gas composition indicating the presence of a hydrocarbon contaminant.

f. The percentage of sources having defects (60%) in this study is in agreement with the results of a study of 53 similar sources by Mound Laboratory[1,2,3], in which 62% of the sources were found to have defects.

2.0 The present requirement for periodic (6 month) status reports of leak test results for sources in use appears to be adequate. The wipe test, consisting of smearing the source with a dry wipe and assaying the radioactivity removed on the wipe in a suitable radiation counting instrument, has some inherent potential inadequacies as follows:

a. Of most importance, the wipe test tells that release of contamination has already occurred; it cannot predict a potential release of contamination, but it can help prevent the contamination from becoming widespread.

b. The mechanical action of wiping the source capsule may not be adequate because it must be done remotely by wiping all areas which may have contamination.

c. The radiation counting instrumentation used by the source owner to determine the amount of contamination in the wipe may not remain in proper calibration during the periods of non-use between wipe tests.

3.0 The results of this study indicate that some form of periodic nondestructive examination in addition to the 6-month wipe test is needed to detect defects not found by the manufacturer's quality control procedures and to serve as an independent check on the source containment integrity. This conclusion is based on the following:

a. The smear test did not detect a leak in the outer capsule of one source. Evidently the inner capsule provided sufficient containment to prevent the release of contamination in this case; however, the design of the capsule closure, the weld quality, and the welding deficiencies of the inner capsule indicate that it cannot be relied upon as a contamination containment barrier.

b. The wipe test has inherent potential inadequacies which could allow a source having leak holes to go undetected. An example of this was observed while examining source N-PuBe-294 in which

both the inner and outer capsules were found to have leak holes in the weld zones which were not detected by the wipe test. The PuBe appeared to be in the form of a solid mass, and the likelihood of PuBe migration through both the inner and outer end-cap weld leaks would be quite low unless some of the Pu were present as a powder, such as $^{239}PuO_2$, having particle sizes small enough to pass through the leak holes.

Periodic destructive testing of sources is not indicated by this study; however, the thorough inspection of a few sources from a given generation of sources (defining a generation as those sources produced in a five-year period) at 10, 15, and 20 years after manufacture as defined by the median age of the generation can be used to detect the effects of aging, compatibility effects, or external environmental attack.

Sources which show significant damage or corrosion should be recalled immediately for thorough nondestructive and/or destructive inspection and possible reencapsulation or removal from service.

4.0 The results of this study do not indicate a maximum age for a Pu-Be source beyond which the continued integrity is suspect. The development of an accelerated aging program, originally planned for this study, was deleted due to funding problems.

Accelerated aging tests were conducted on 316L stainless steel encapsulated BNL strip-type CsCl sources in which four sources were subjected to thermal cycling between -75°C and 176°C for a minimum of 757 to a maximum of 2457 cycles. There were no detectable leaks, swelling or corrosion.[5]

RECOMMENDATIONS

1.1 The examination of these sources has shown that the major problem with the sources is weld defects, with the greater portion of the defects found in the tantalum capsules. Some of the reasons for the defects are: (a) excessive grain growth in tantalum weld joints, (b) embrittlement of the tantalum weld joint due to impurities, (c) misalignment of the welding torch and weld joint, (d) insufficient weld penetration, (e) a weld joint design joining a thin wall and thick end cap which leads to unequal heating, which in turn causes poor penetration.

From the evidence of the number of weld defects found in these tests of the sources manufactured during the 1961 and 1962 period, the integrity of encapsulation of the PuBe is questionable. Leak-proof, double encapsulation is a desirable goal for the PuBe sources because of the extremely high toxicity level of the Pu and Be. Based on this goal, the rate of defects (∿60%) found indicates that half of the sources of this period are defective. However, these tests showed a rate of outer capsule leaks to be ∿17% and ∿10% for those tested by Mound Laboratory.

The quality of source encapsulation could be improved by: (a) redesign of the weld joints to provide metal of equal thickness for fusion weld joints, (b) use of radiography to inspect each weld joint, (c) use of a reliable leak test which is both sensitive and capable of measuring gross leaks, such as a vacuum leak test or a hot water bubble test,[4] (d) careful cleaning and handling of source capsules prior to welding to ensure that no organic or foreign material is inadvertently encapsulated which could cause welding problems and/or the buildup of internal gas pressure by radiolytic decomposition of the material.

2.1 The wipe testing of sources every six months as a means of detecting leaking sources should be continued, even though it is inadequate in detecting sources with leaks unless the sources are leaking a detectable quantity of plutonium. This leak test should be supplemented with a hot water bubble test at one-year intervals or following an accident that would deform the capsule and, in particular, the weld area. A source giving a positive indication of a leak should be returned for reencapsulation.

An additional test that should be used for all newly fabricated sources would be for the fabricator to supply with each source a low-cost simple go no-go gage to be used to test for source swelling and/or accident deformation. The go no-go test of the source would be made at six-month intervals for a period of two or three years, after which time this could be discontinued except in the case of an accident, as the possibility of swelling from gas formation of sealed-in foreign matter would be expected to have ceased.

3.1 Based upon the ∿10% rate of leaks found in the sources tested for the period of 1958 thru 1962 (Mound Laboratory), it is recommended that two groups of sources be selected, one group of approximately five year-old sources and one less than one year old for nondestructive testing. A small fraction of these sources should be selected for destructive testing. This test program would indicate the probable number of defective sources in use and whether there is a need for a more stringent user-testing program for detection of defective sources.

4.1 There is no apparent reason why a well-manufactured leak-tight, doubly encapsulated source should not have an essentially unlimited life span when used in mild environments. Sources used in well logging operations can be subjected to corrosive and erosive environment, and their life will be dependent upon a minimum dimensional measurement that should be specified in the original design.

Leak tests of sources have shown that ∿10% of the old (10 years and older) sources would be expected to have leaks. These leaking sources should be located and removed from service. This can be accomplished by: (a) either recalling all old sources for leak testing and reencapsulating the leaking sources or (b) requiring a user to perform a hot-water bubble test and wipe test, and to submit a sample of the water used in the hot-water bubble test for determination of the

contamination removed from the source. If there is a positive test indication of a leak, the source should be returned for reencapsulating or disposal.

REFERENCES

1. R. D. Madding, Jr., and R. E. Vallee, *Plutonium-239/Beryllium Neutron Source Surveillance and Testing*, MLM-1927, Mound Laboratory (February 15, 1973).

2. K. L. Breakall, *Destructive Analysis of Neutron Sources M-71 and 1053S*, Mound Laboratory (April 5, 1972).

3. R. D. Madding, Jr., and R. E. Vallee, *Plutonium-239/Beryllium Neutron Source Surveillance and Testing*, Mound Laboratory (June 1, 1972).

4. R. G. Niemeyer, *Leak Testing Encapsulated Radioactive Sources*, ORNL-4529, Oak Ridge National Laboratory (July 1972).

5. Letter from A. F. Rupp, Oak Ridge National Laboratory, to E. E. Fowler, D.I.D.-Atomic Energy Commission (August 11, 1970).

APPENDIX A

Metallographic Inspection Results

The metallographic examination consisted of mounting the samples in an epoxy resin metallographic mount and grinding the sample flat on a lapping machine, using a silicon carbide abrasive slurry. After being ground, the samples were polished using vibratory polishing units. The final polishing step employed a 1/2-μm diamond paste-water slurry on a nylon polishing cloth. After being polished, the samples were examined using a metallurgical microscope and photographed in the as-polished condition. The samples were then etched to reveal the grain structure. An etchant of five parts HCl and one part HNO_3 was used to etch the stainless steel samples, while an etchant of 30 g NH_4F-HF, 50 ml HNO_3, and 20 ml H_2O was used to reveal the grain structure of the tantalum samples. A solution of 2 ml HNO_3 and 98 ml methanol was used to etch the Vega steel sample. After being etched, the samples were examined using a microscope, and photomicrographs were made to record the grain structure.

Source N-PuBe-294, Sample 1, Bottom Section, Inner and Outer Capsules

Figure 10 shows the weld zone of the stainless steel outer capsule in the as-polished condition. This photograph shows there are no visible cracks in the weld and that the weld joint penetration measures only slightly less than the tube wall thickness of the capsule. Figure 11(a) shows this capsule in the etched condition. This photomicrograph shows that part of the outer surface of the end cap material has been either torn or ground away (probably during the sectioning of the capsule), but most of the weld nugget was left intact. Figure 11(b) shows the weld zone microstructure at a high magnification (100X). Figure 12 shows the weld zone of the inner tantalum capsule in the as-polished condition. Note the crack penetrating through the weld zone. Figure 13 shows the same area in the etched condition. Excessive grain growth is visible around the weld zone. At this plane of polish it appears that the original weld penetration was only one grain thick and that the capsule end-cap separation occurred at the boundary of a large grain. Figure 14(a) shows amorphous material between the inner capsule wall and the end cap. This material was observed in the as-polished condition only. When this sample was etched to reveal the grain structure of the tantalum, this feathery-appearing material was dissolved along with the unidentified layer marked by arrows in Fig. 14(b). This photomicrograph shows the bottom of the end cap on the left and the inside of the capsule on the right of the picture.

Source N-PuBe-294, Sample 2, Stainless Steel Outer Capsule

Figures 15 and 16 show the end-cap weld zone in the as-polished and etched conditions. It is evident that this weld penetration is less than half the thickness of the capsule tube wall. Judging from these photographs, it

appears that a welding alignment problem was encountered during the capsule welding process. Figure 16 shows the first weld bead (arrow) was not centered over the end cap-capsule interface. In the final welding operation it appears that the wall of the capsule was heated to a molten stage extending 0.120 in. down the side of the capsule wall. This molten zone collapsed (thickened area near top of tube wall), which caused excessive roll-over at the corner of the weld zone. Figure 17(a) shows the weld zone at a higher magnification (100X) in the as-polished condition, and Fig. 17(b) shows the etched condition. The arrow points to an unidentified inclusion trapped between the end cap and the weld metal. Figure 18 shows a void area at the tube wall-collapsed tube section interface. This void area penetrated 0.004 in. into the tube wall.

Source N-PuBe-295, Sample 1, Stainless Steel, Top Weld, Outer Capsule

Figure 19 is the as-polished photomacrograph of the weld zone. Figures 20(a) and 20(b) show the weld zone in the etched condition. This weld joint appeared to be sound and free of cracks and voids.

Source N-PuBe-295, Sample 2, Bottom Section
Tantalum Inner and Stainless Steel Outer Capsules

Both the inner and outer capsules are shown in the as-polished condition in Fig. 21. Higher magnification photomicrographs of the outer stainless steel weld zone are shown in Figs. 22(a) and 22(b). In Fig. 22(b) a small crack (less than 0.002 in. in length) is visible at the capsule wall-end-cap-weld metal interface; this figure shows the weld zone in the etched condition. The inner tantalum capsule is shown in Figs. 23(a) and 23(b) in the etched condition. As in the previously discussed tantalum capsule, the grains are quite large near the weld zone. An oxide-appearing layer adheres to the outside surface of this capsule. Also noteworthy are the parallel platelets near the outside surface of the capsule (arrows); these could be internal oxide precipitation platelets.

Source N-PuBe-296, Sample 1, Top Weld, Tantalum Inner Capsule

Figure 24 shows the end-cap weld zone along with an unidentified porous metallic material which was found near the inside surface of the end cap. Although this material does not appear to be attached to either the end cap or the capsule wall at this plane of observation, it could not be dislodged during or after the sectioning of this material. The dark line diagonally spanning the photograph is a scratch caused by dislodged segments of this porous material. No cracks were visible in the weld zone of this capsule. Figure 25(a) shows this porous material at a higher magnification. Also shown in this photograph is the inner surface of the end cap. Note the light colored (arrow) layer adhering to this surface. When this sample was etched to reveal the grain structure of the tantalum, this porous material reacted violently with the etchant. This material was dissolved by the etchant along with the layer adhering to the surface

of the end cap. Figure 25(b) shows a two-phase oxide layer adhering to the crown of the weld. Also observed in this etched condition, near the surface of the capsule, were Widmanstätten patterned platelets. These could be internal oxide precipitating out as platelets; Fig. 26 shows the weld zone in the etched condition.

Source N-PuBe-296, Sample 2, Top Weld, Vega Steel Outer Capsule

The configuration of this capsule was different from the other capsules, as the end cap was "T" shaped; therefore, the weld zone attaching the end cap to the capsule was located on the side of the capsule. This is shown in Figs. 27 and 28 in both the polished and etched conditions. Seen near the top right corner of Fig. 27 is a discontinuous oxide or sulfide stringer spanning three-fourths the thickness of the end cap. Also noted in Fig. 28 is that the weld nugget is not centered at the capsule-end cap interface. Figures 29(a) and 29(b) show this weld zone at a high magnification.

Source N-PuBe-292, Sample 1, Welded Tantalum End Cap

Figures 30 thru 32 show the microstructure of this tantalum end cap. In Fig. 31 note that the grains around the weld area are not as large as in the previous tantalum weld zones that were examined. As shown, this weld zone appears free of cracks and voids.

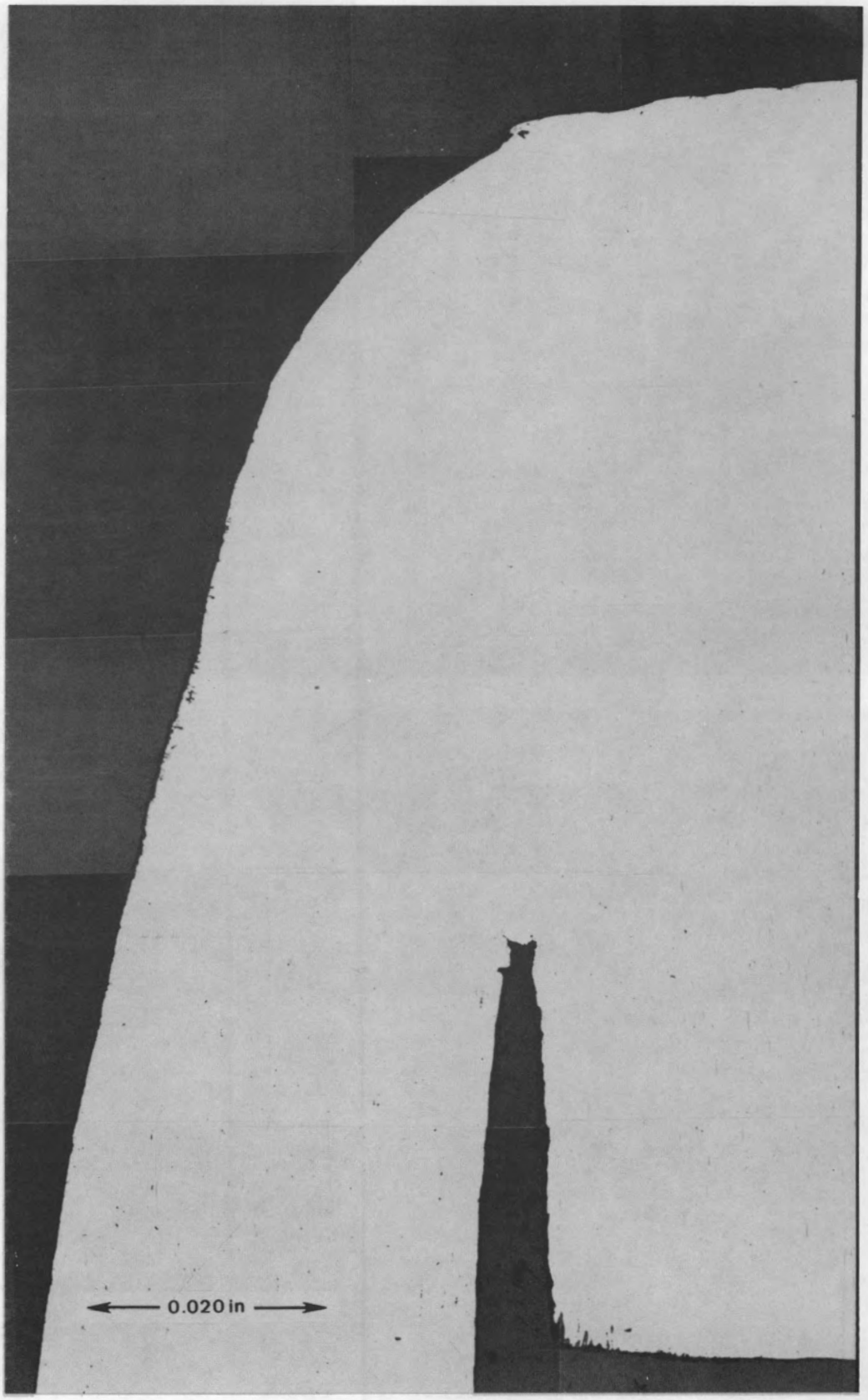


Fig. 10. Source N-PuBe-294, Bottom Section, Outer Stainless Steel Capsule, Sample 1. As Polished.

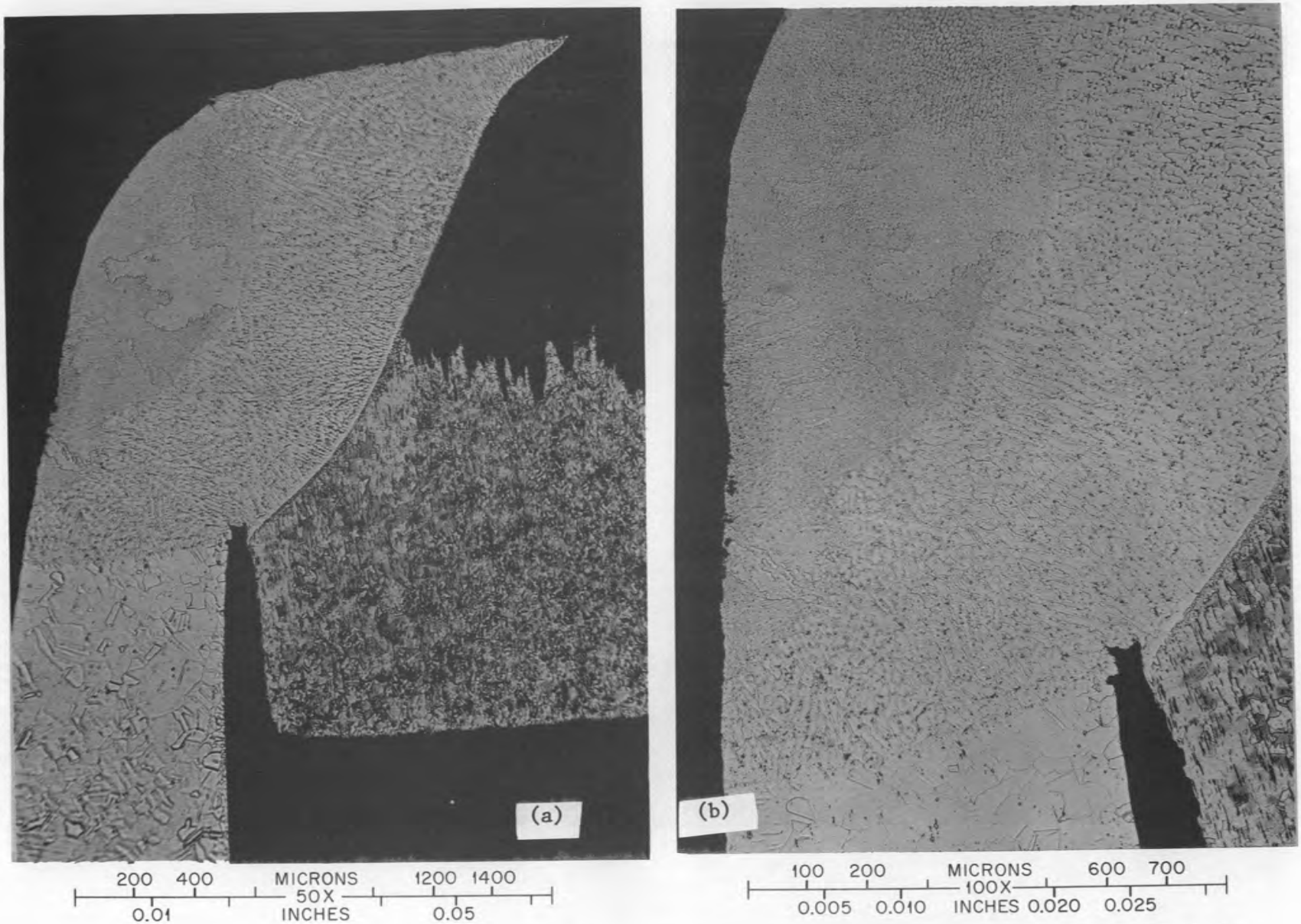


Fig. 11. Source N-PuBe-294, Bottom Section, Outer Stainless Steel Capsule, Sample 1. Etched.

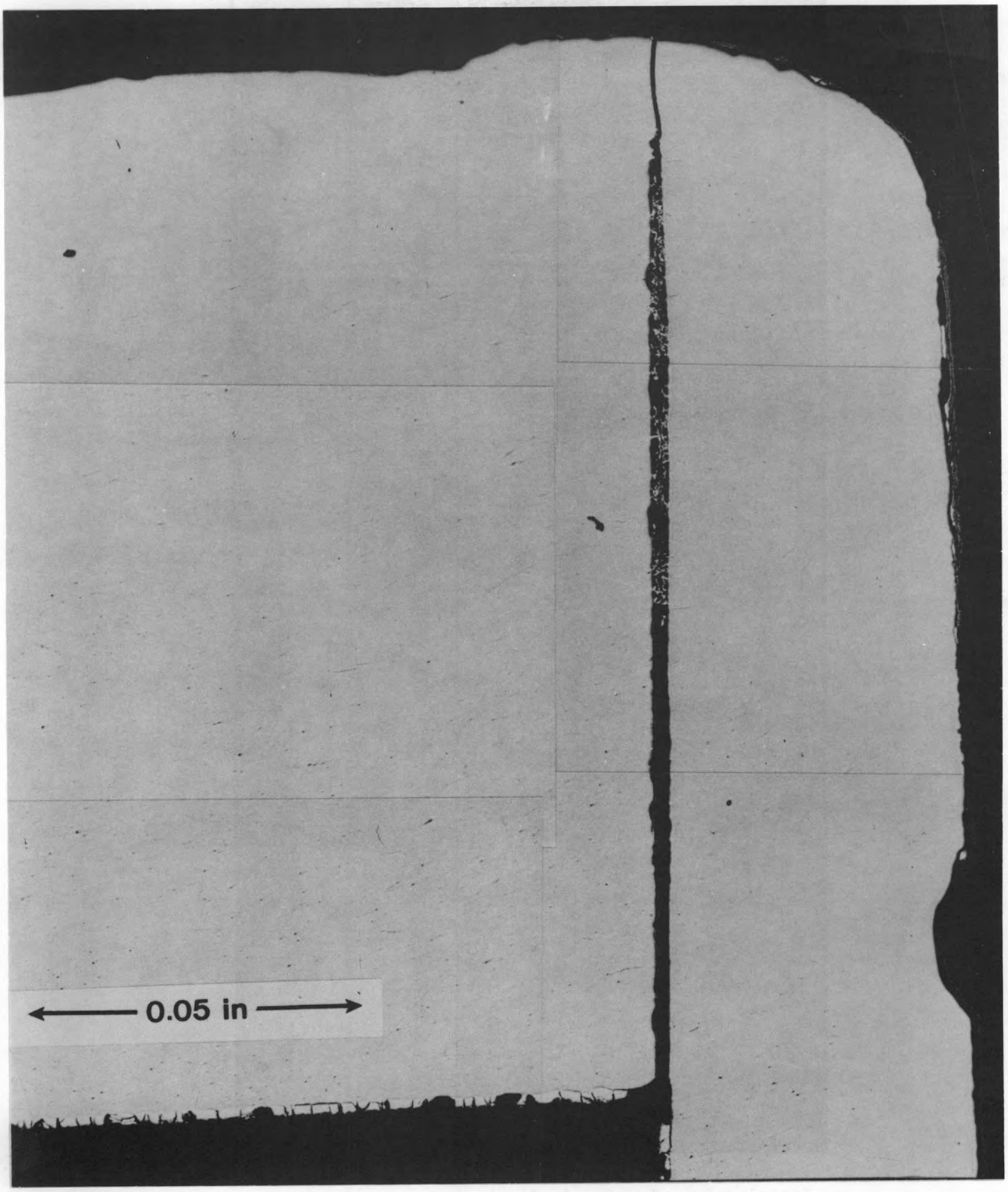


Fig. 12. Source N-PuBe-294, Bottom Section, Inner Tantalum Capsule, Sample 1. As Polished.

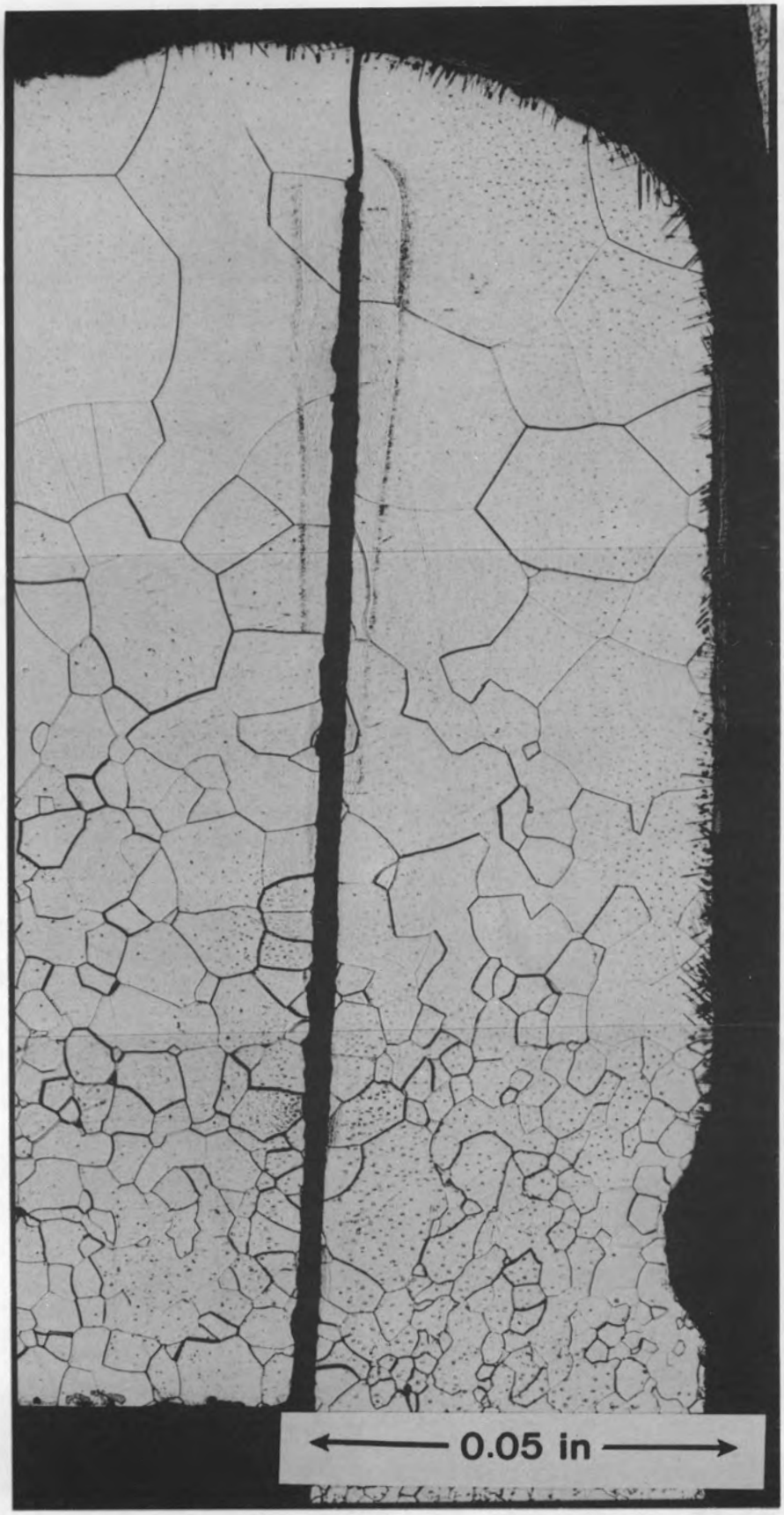


Fig. 13. Source N-PuBe-294, Bottom Section, Inner Tantalum Capsule, Sample 1. Etched.

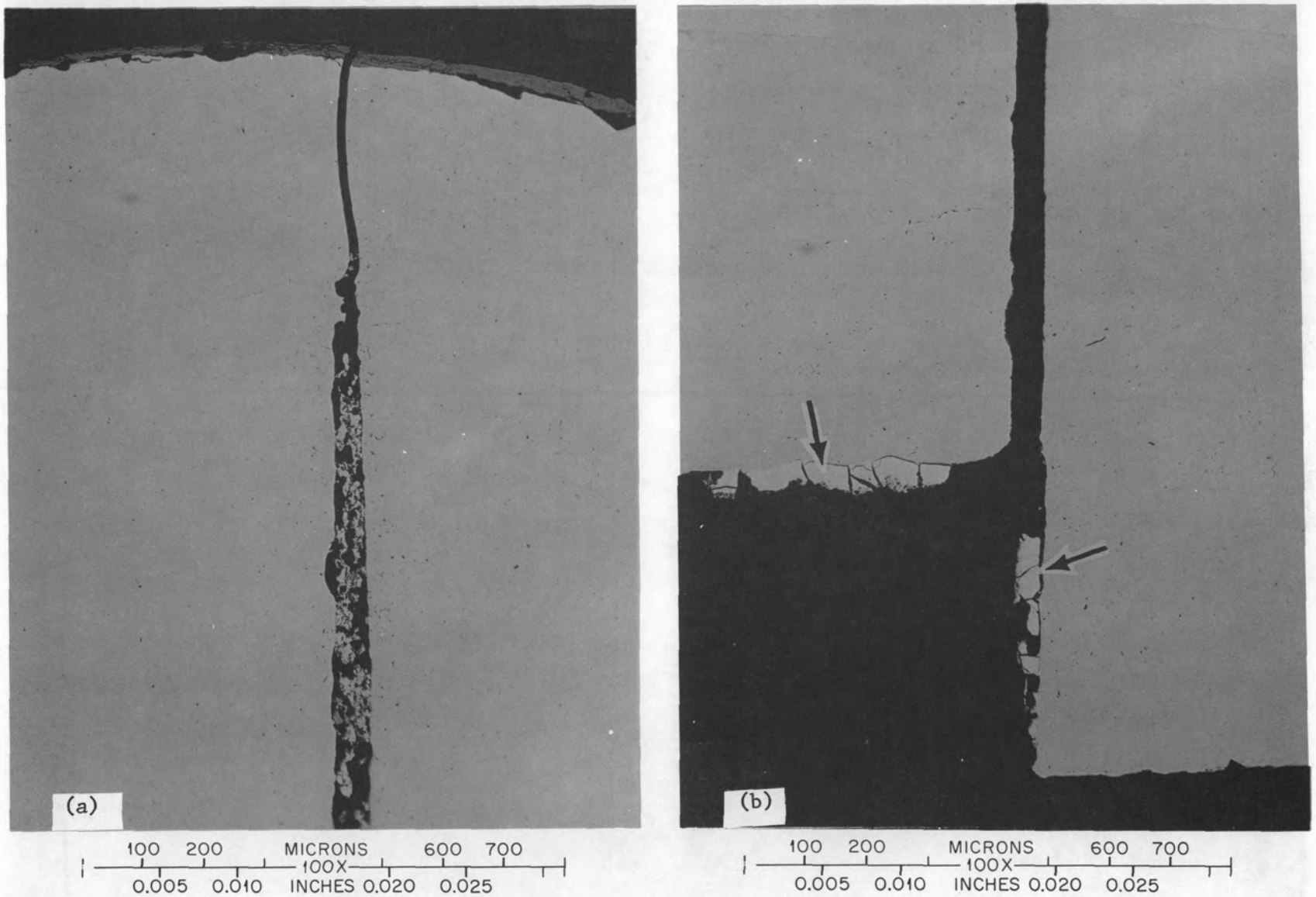


Fig. 14. Source N-PuBe-294, Bottom Section, Inner Tantalum Capsule, Sample 1. As Polished.

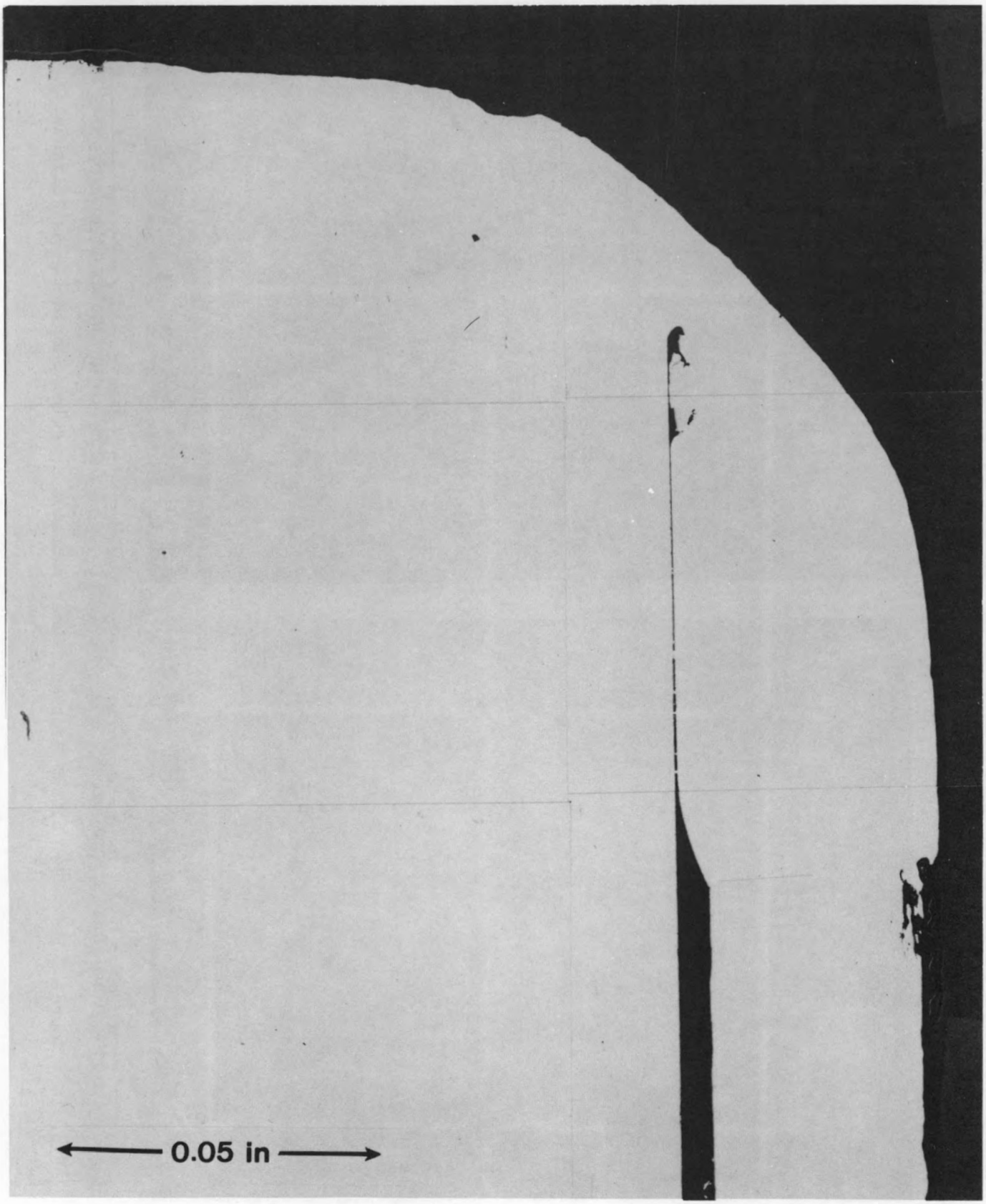


Fig. 15. Source N-PuBe-294, Top Weld, Outer Stainless Steel Capsule, Sample 2. As Polished.

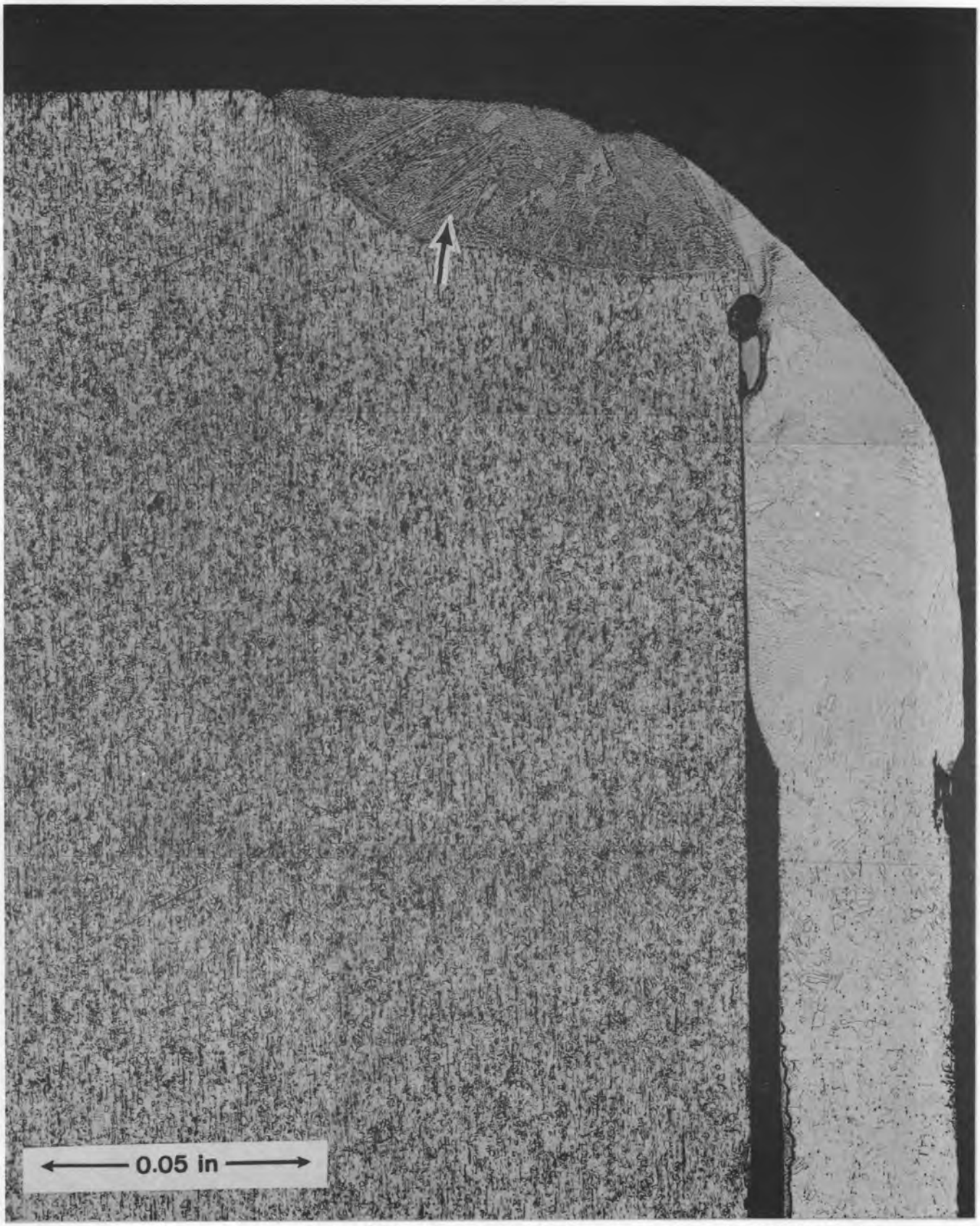


Fig. 16. Source N-PuBe-294, Top Weld, Outer Stainless Steel Capsule, Sample 2. Etched.

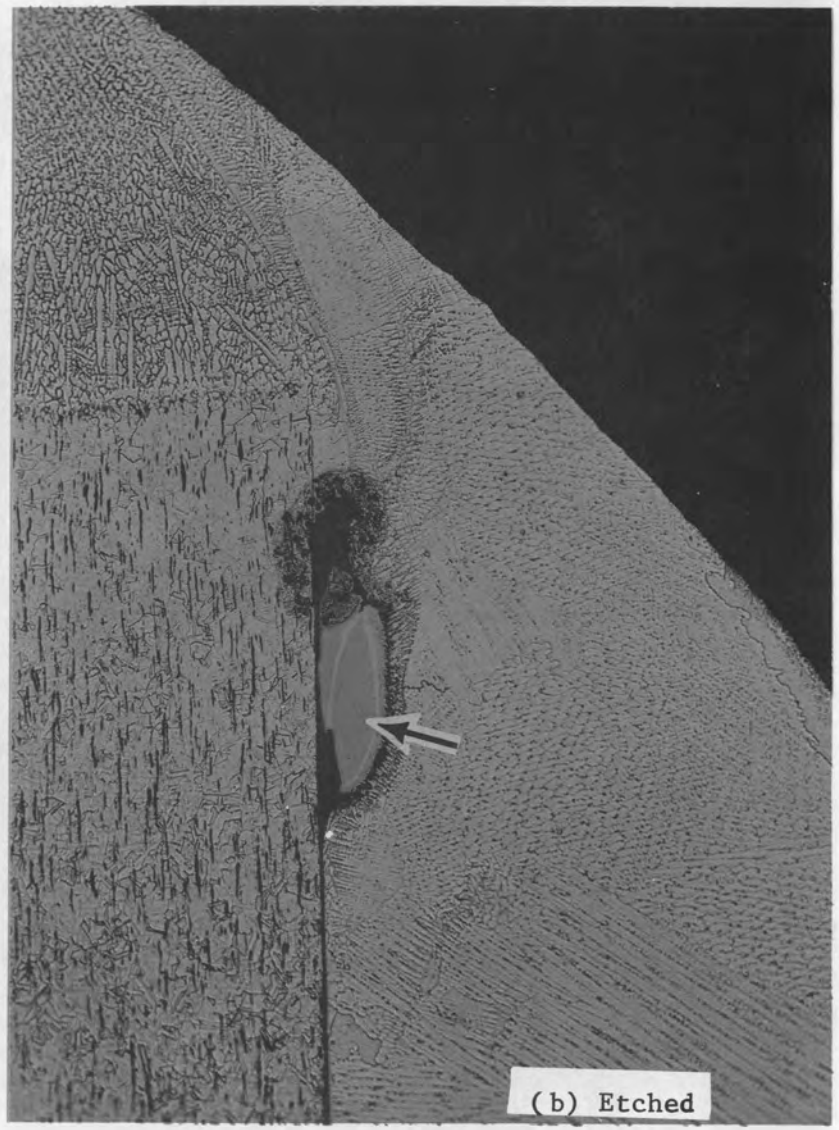


Fig. 17. Source N-PuBe-294, Top Weld Outer Stainless Steel Capsule, Sample 2.

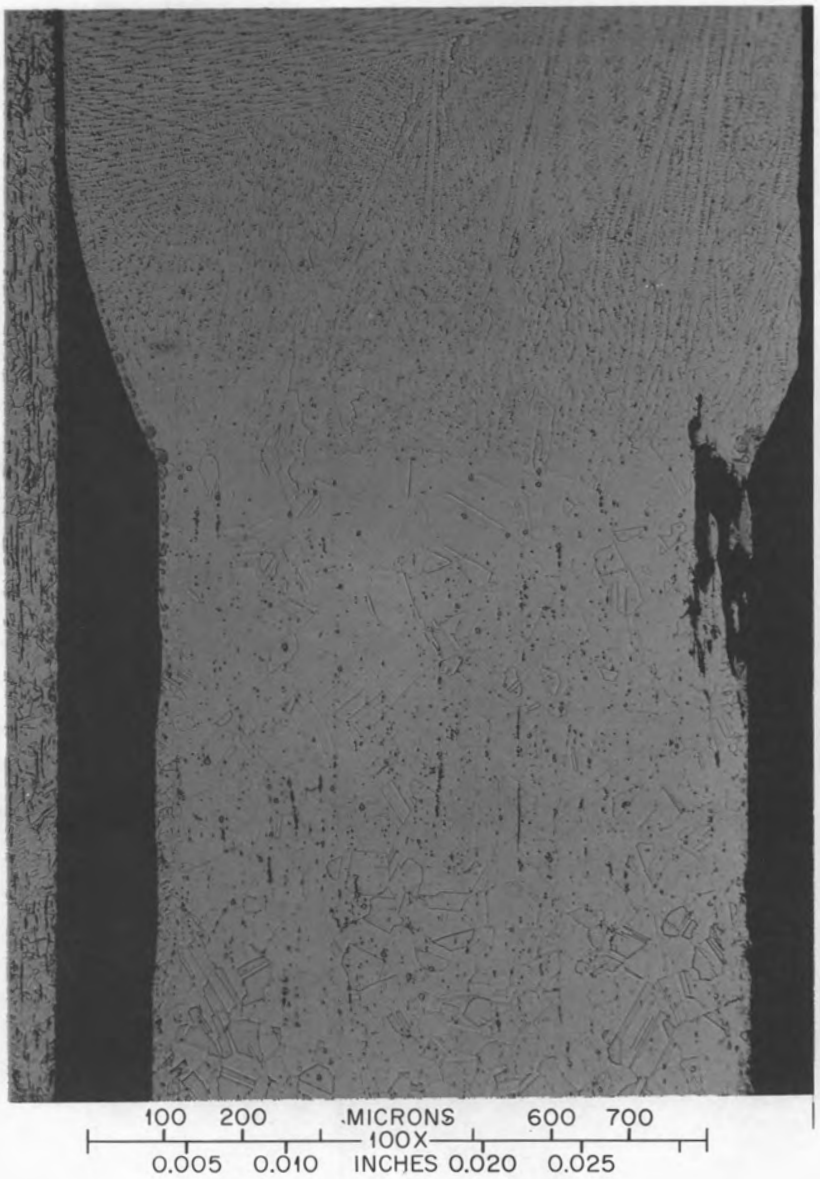


Fig. 18. Source N-PuBe-294, Top Weld, Outer Stainless Steel Capsule, Sample 2. Etched.

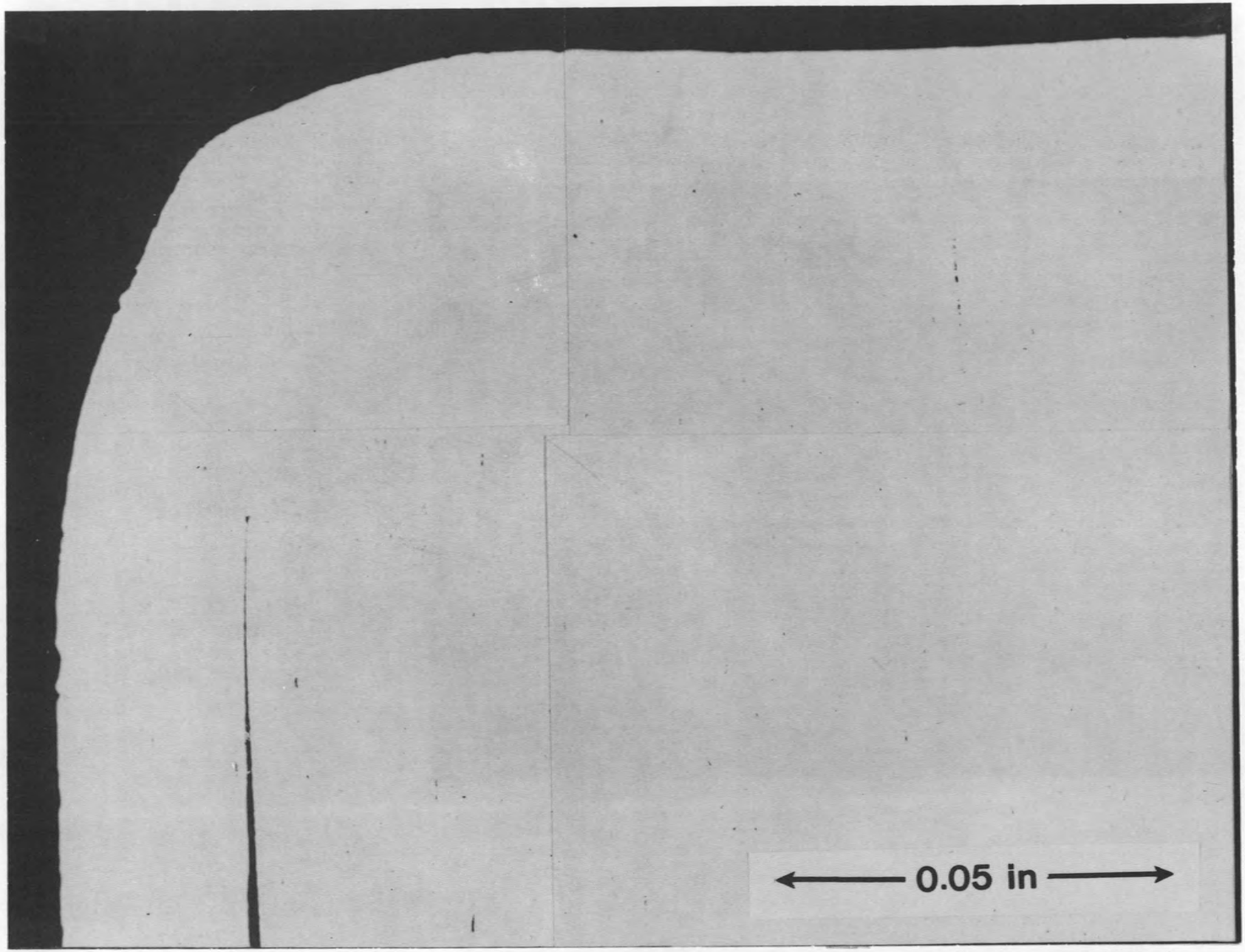


Fig. 19. Source N-PuBe-295, Top Weld, Outer Stainless Steel Capsule, Sample 1. As Polished.

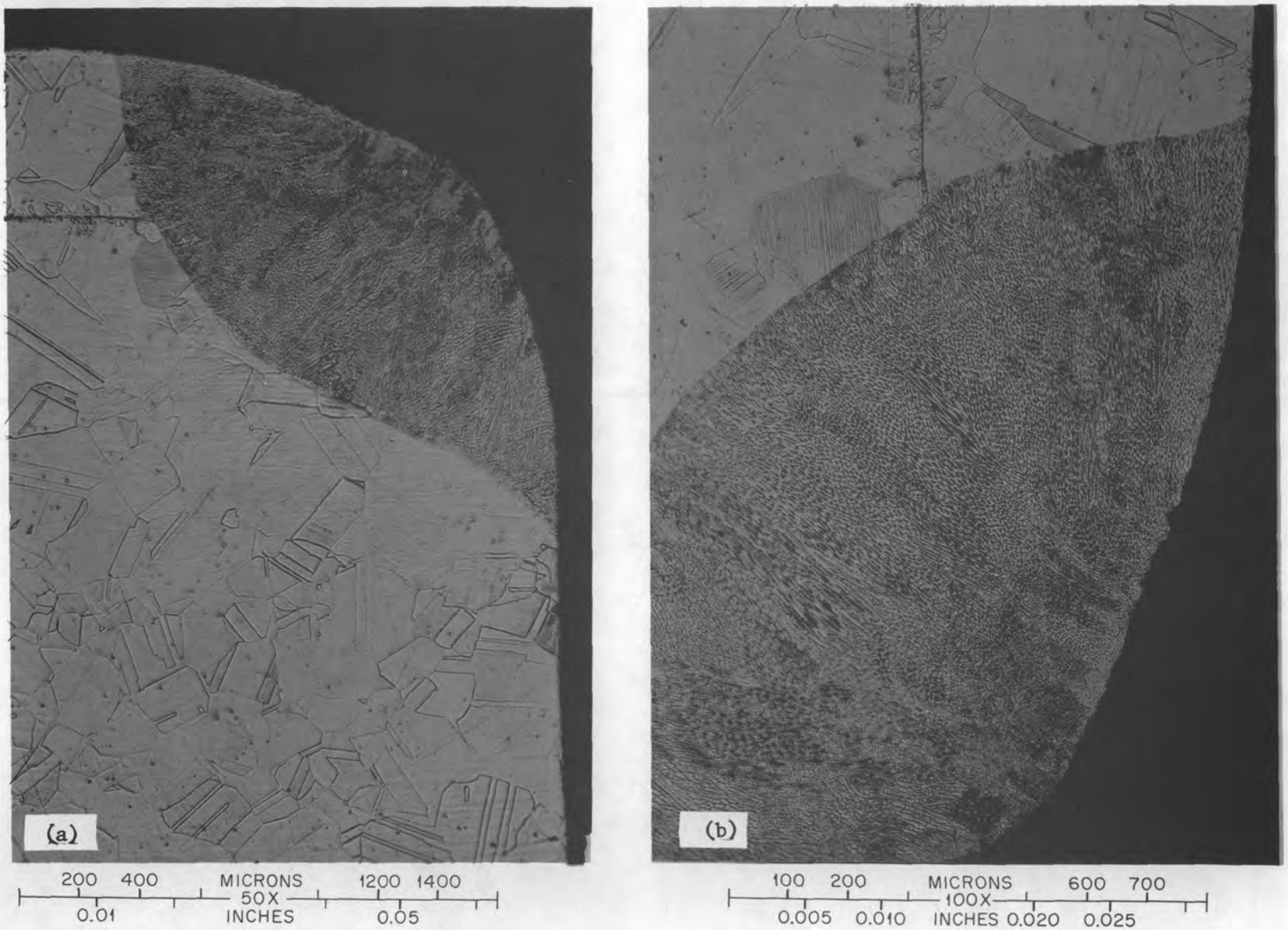


Fig. 20. Source N-PuBe-295, Top Weld, Outer Stainless Steel Capsule, Sample 1. Etched.

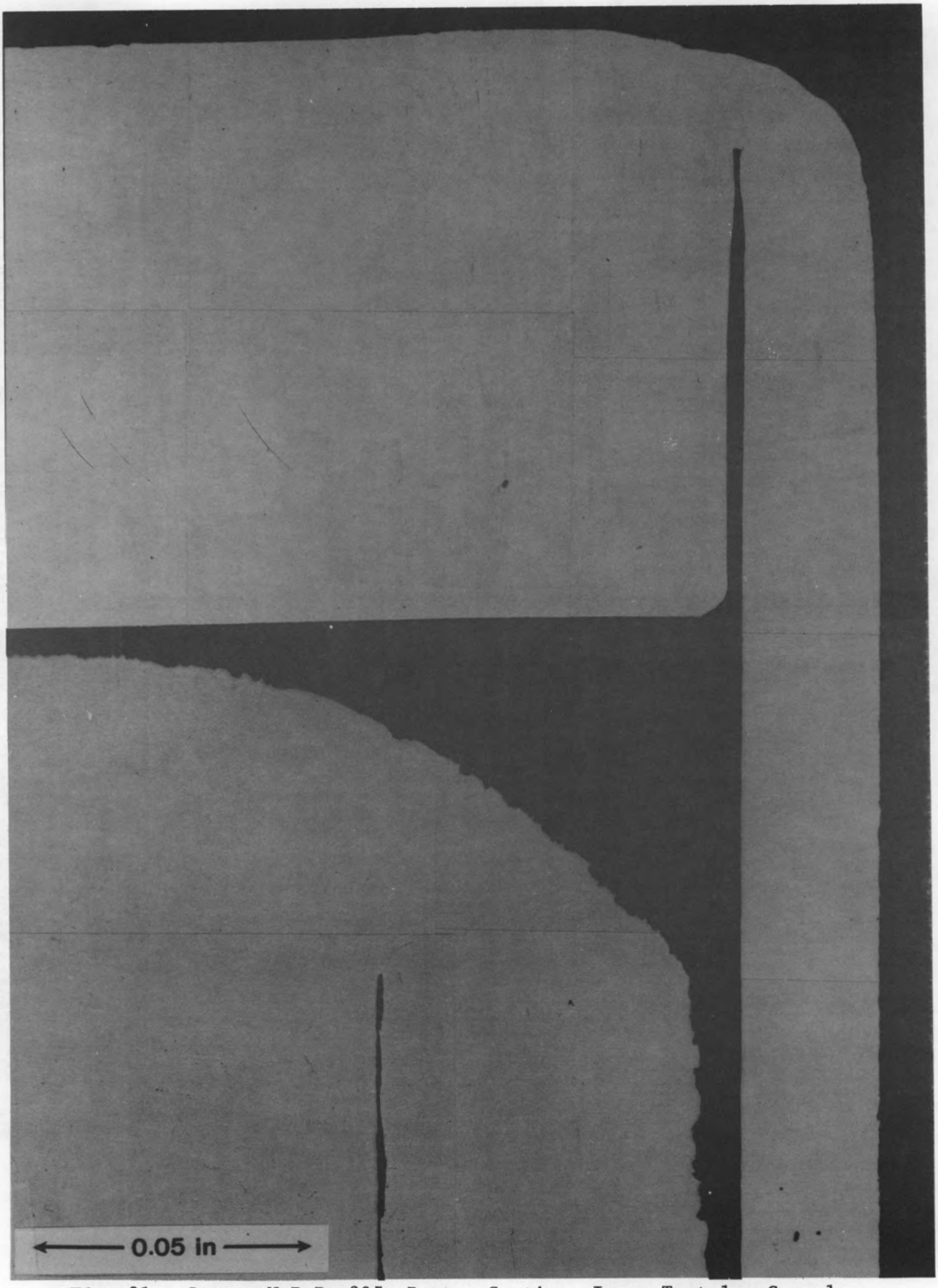


Fig. 21. Source N-PuBe-295, Bottom Section, Inner Tantalum Capsule and Outer Stainless Steel Capsule, Sample 2. As Polished.

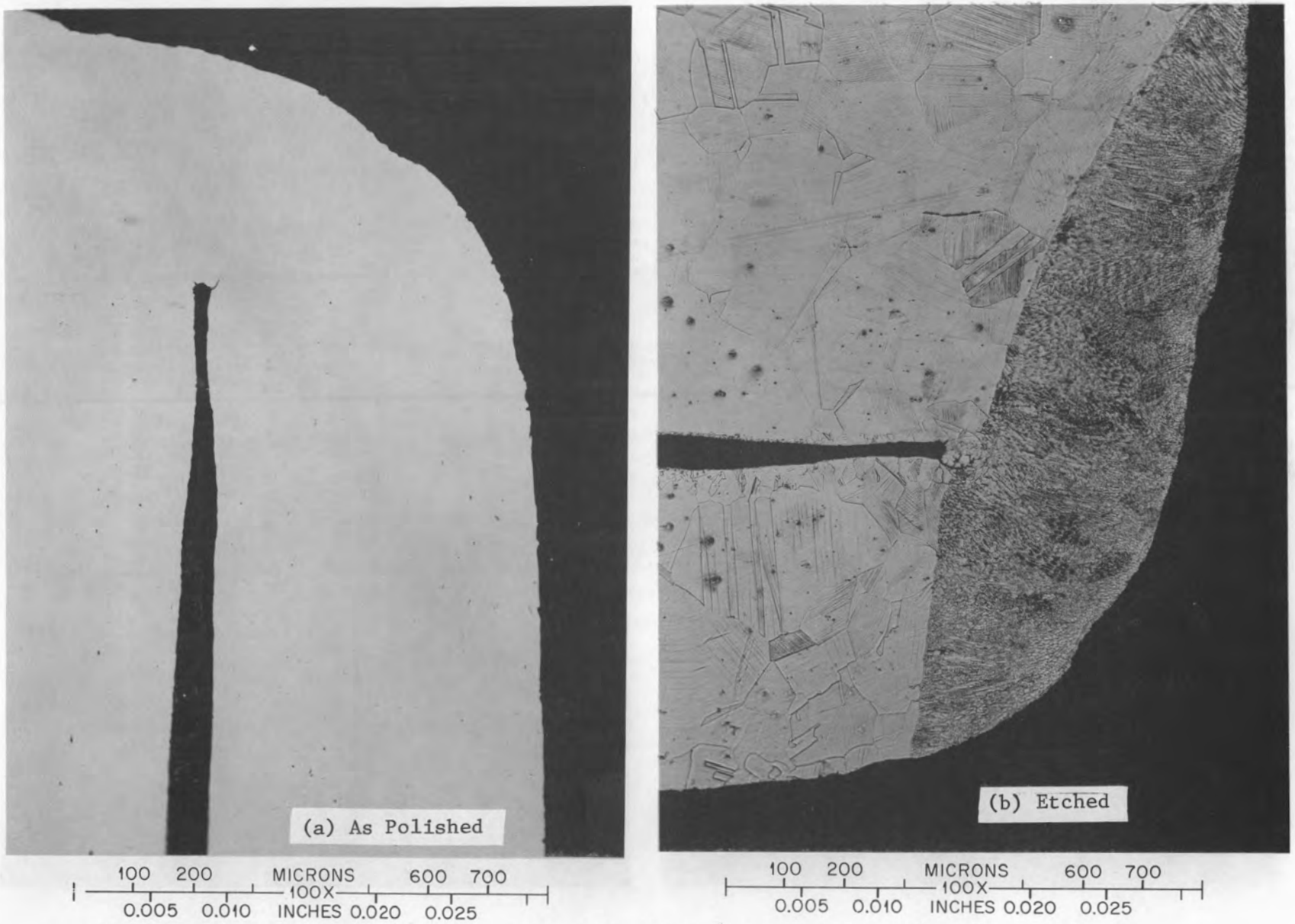


Fig. 22. Source N-PuBe-295, Bottom Section, Outer Stainless Steel Capsule, Sample 2.

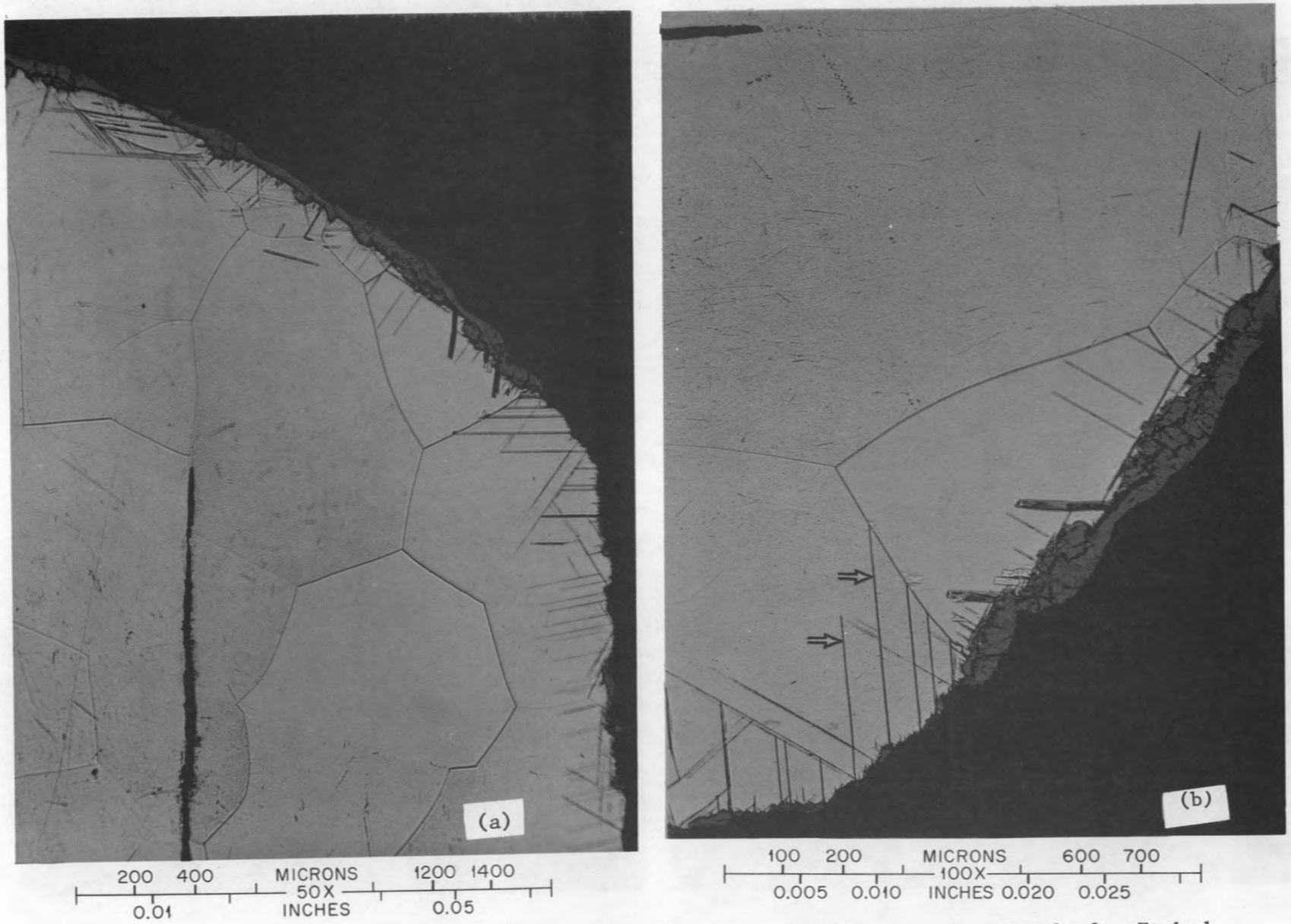


Fig. 23. Source N-PuBe-295, Bottom Section, Inner Tantalum Capsule, Sample 2. Etched.

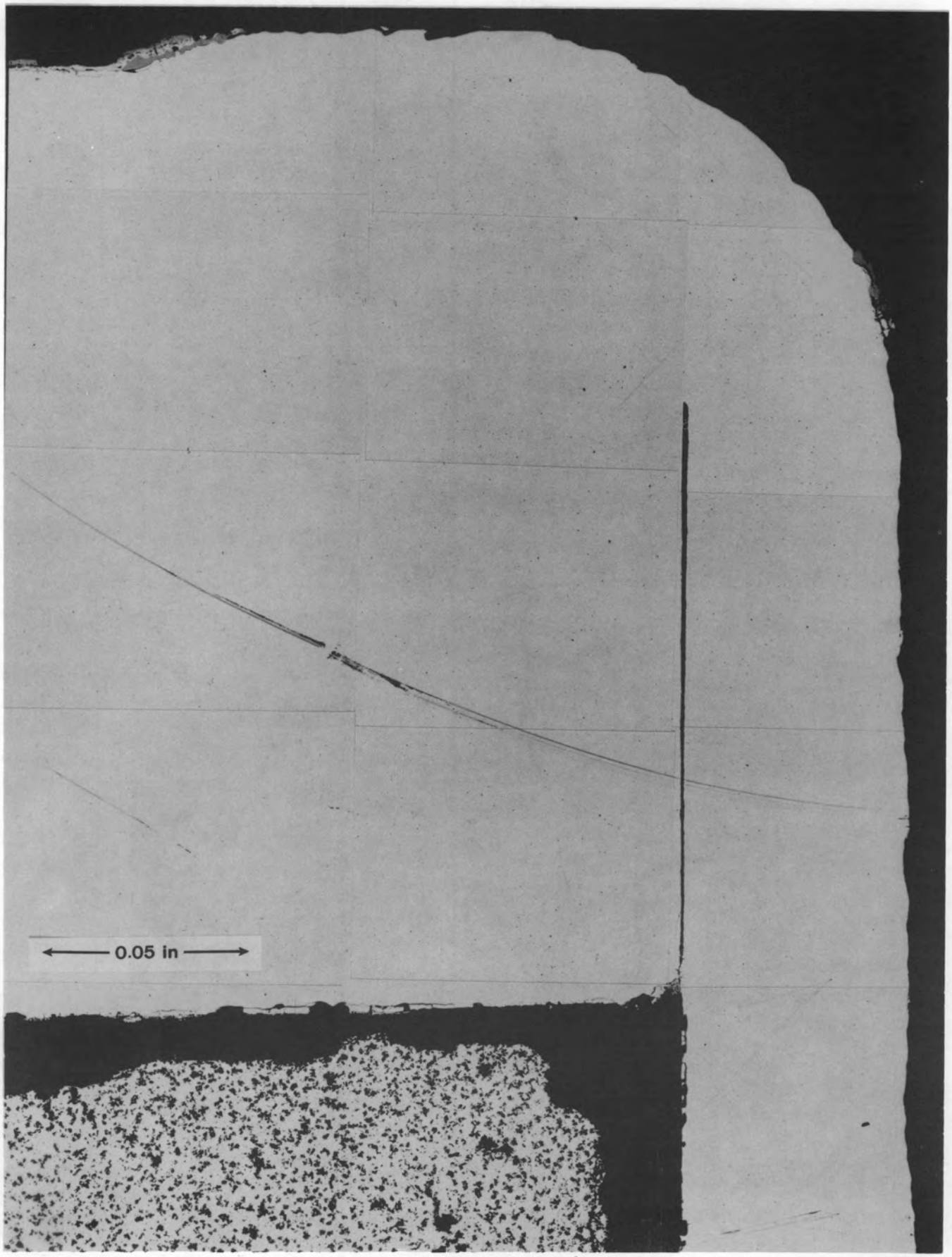
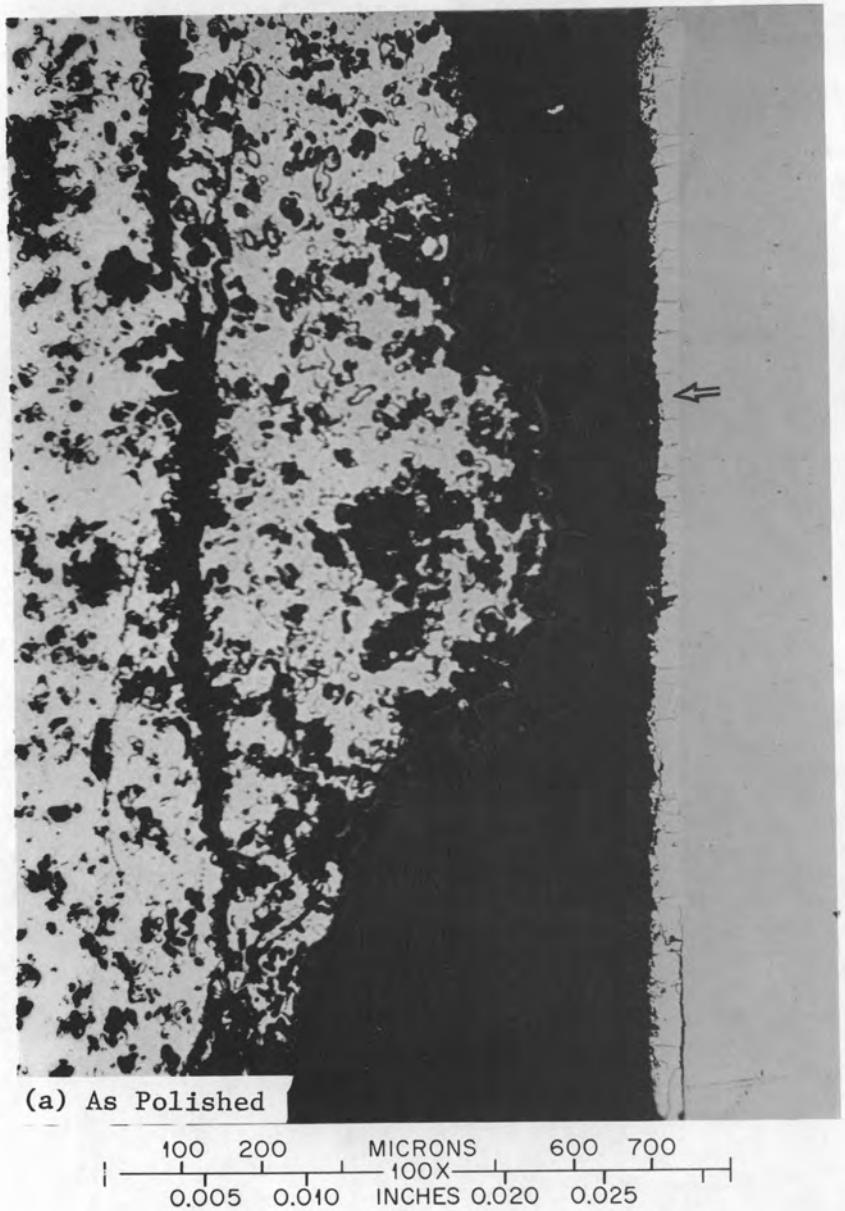


Fig. 24. Source N-PuBe-296, Top Weld, Inner Tantalum Capsule, Sample 1. As Polished.



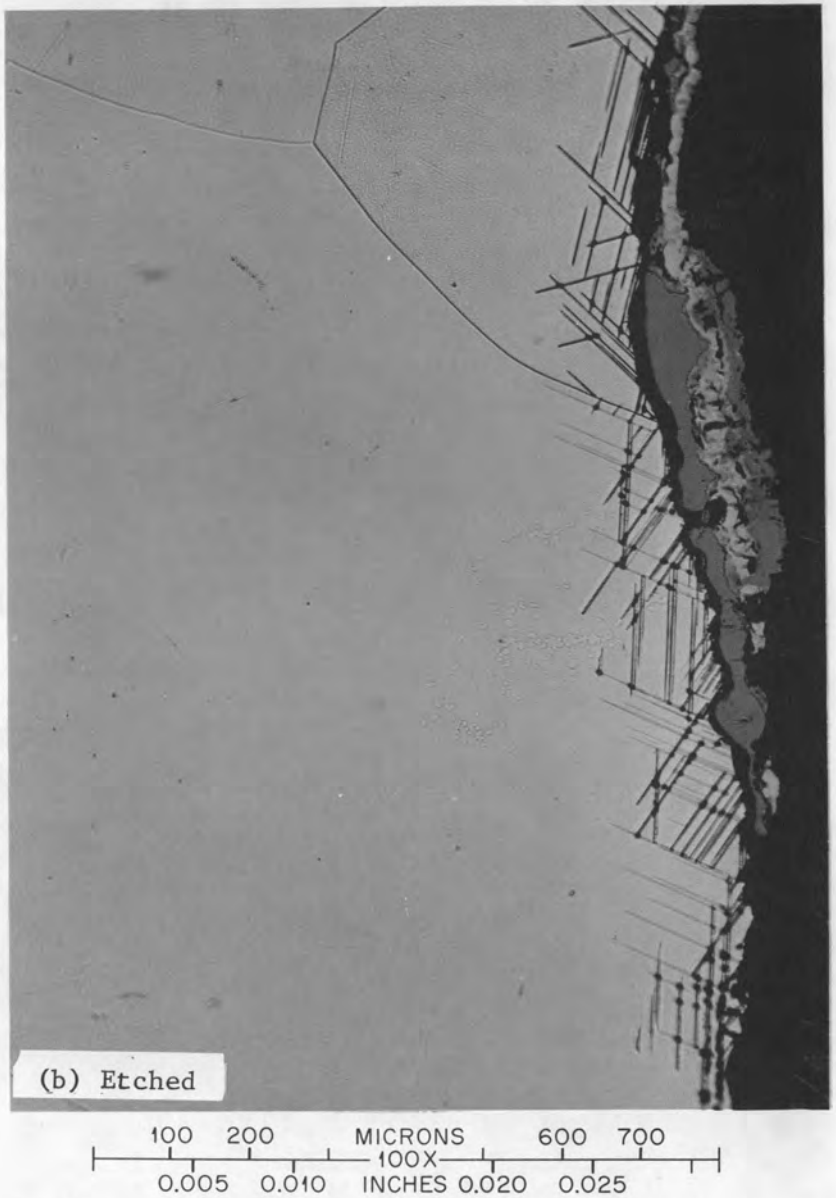


Fig. 25. Source N-PuBe-296, Top Weld, Inner Tantalum Capsule, Sample 1.

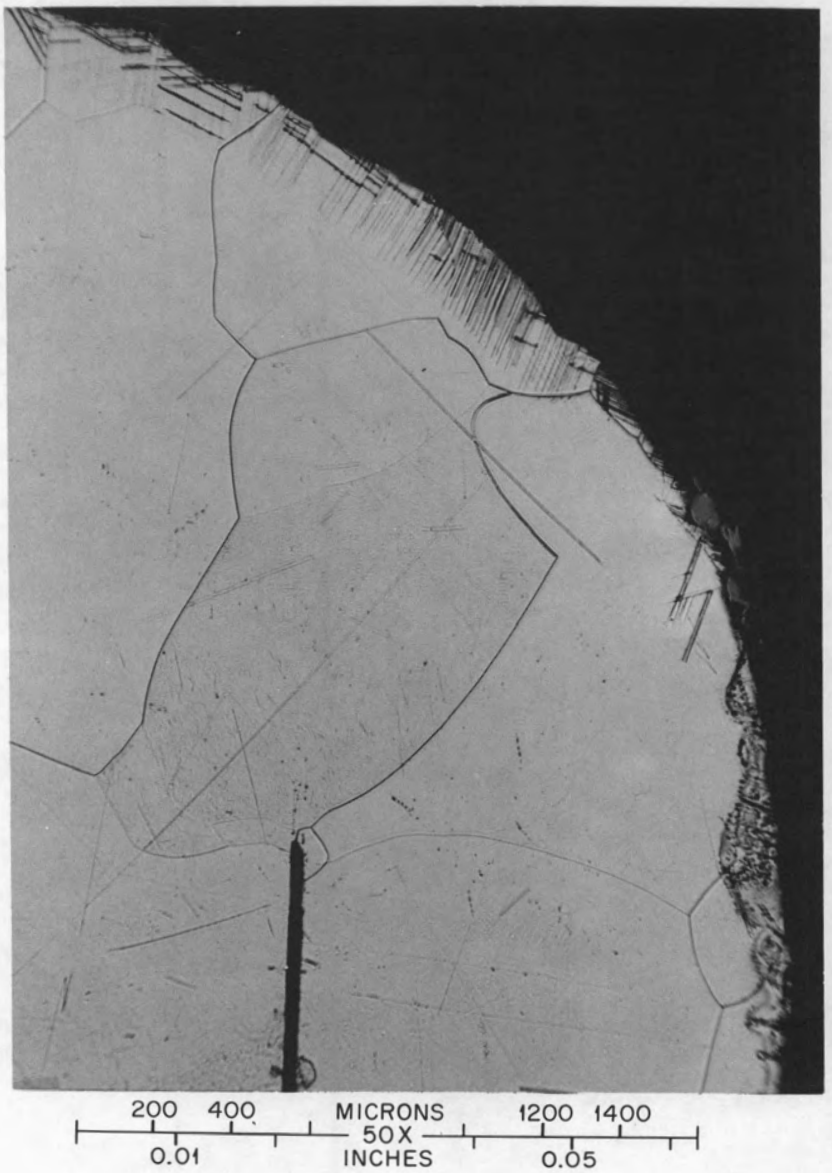


Fig. 26. Source N-PuBe-296, Top Weld, Inner Tantalum Capsule, Sample 1. Etched.

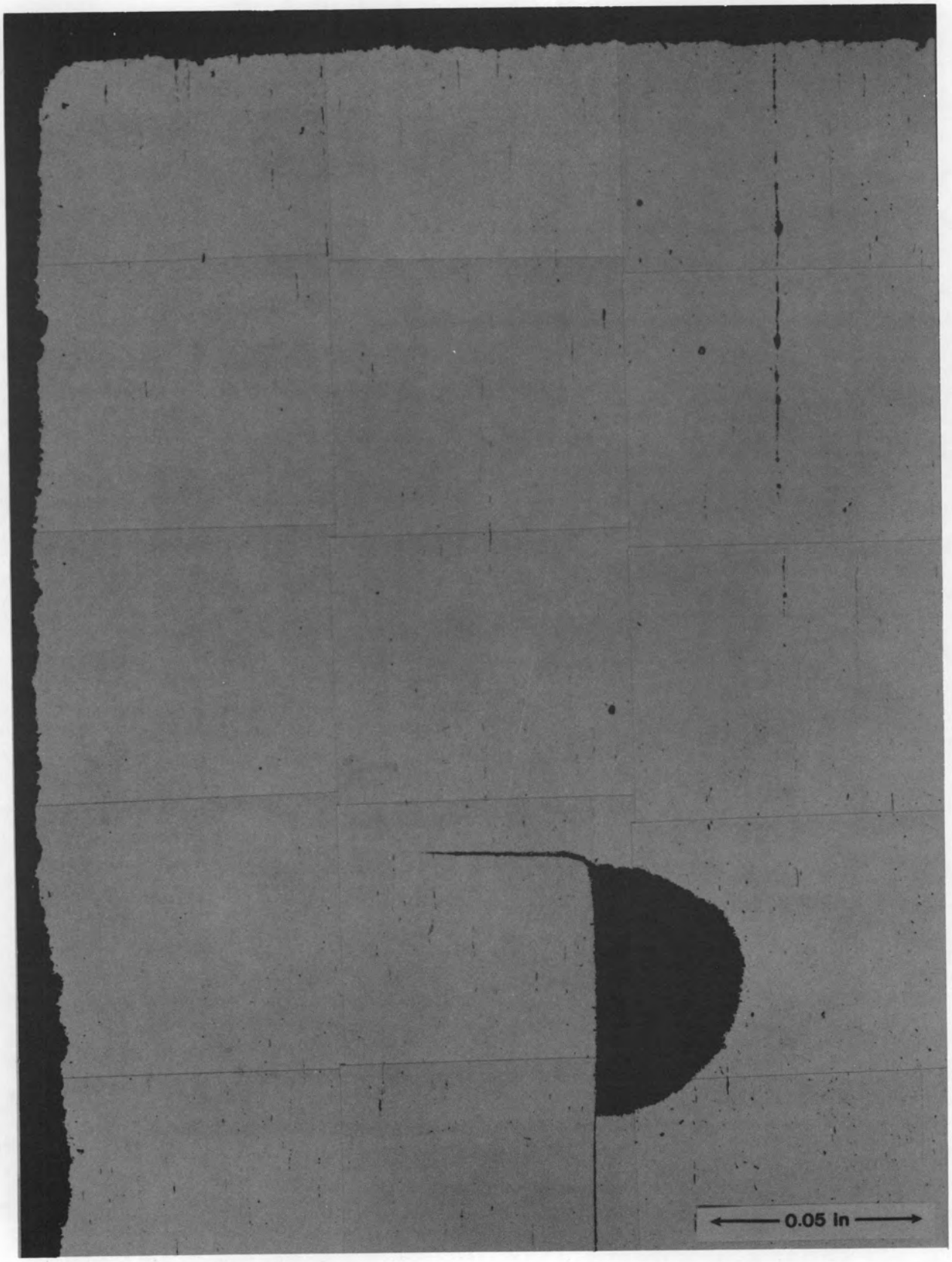


Fig. 27. Source N-PuBe-296, Top Weld, Outer Vega Steel Capsule, Sample 2. As Polished.

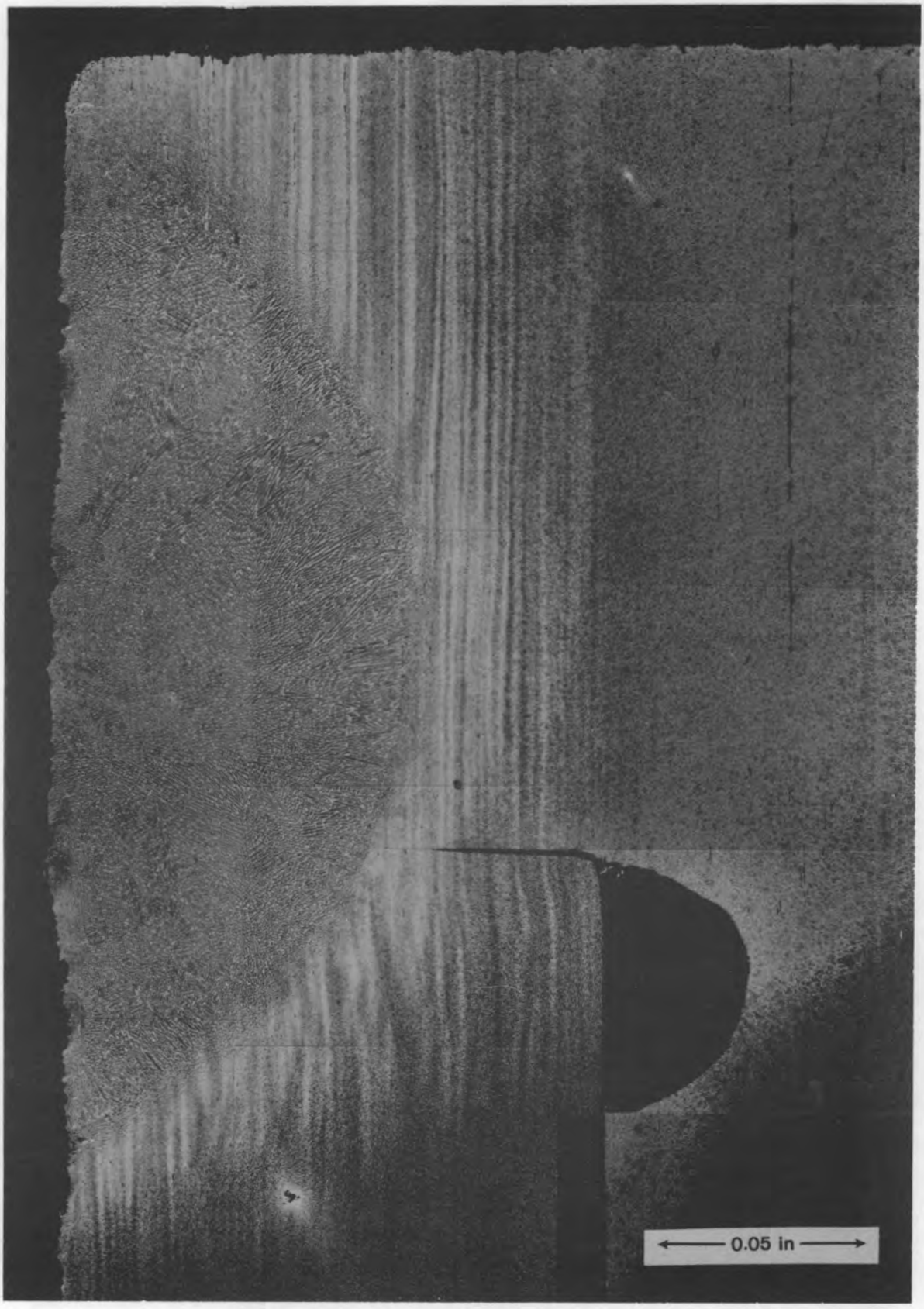
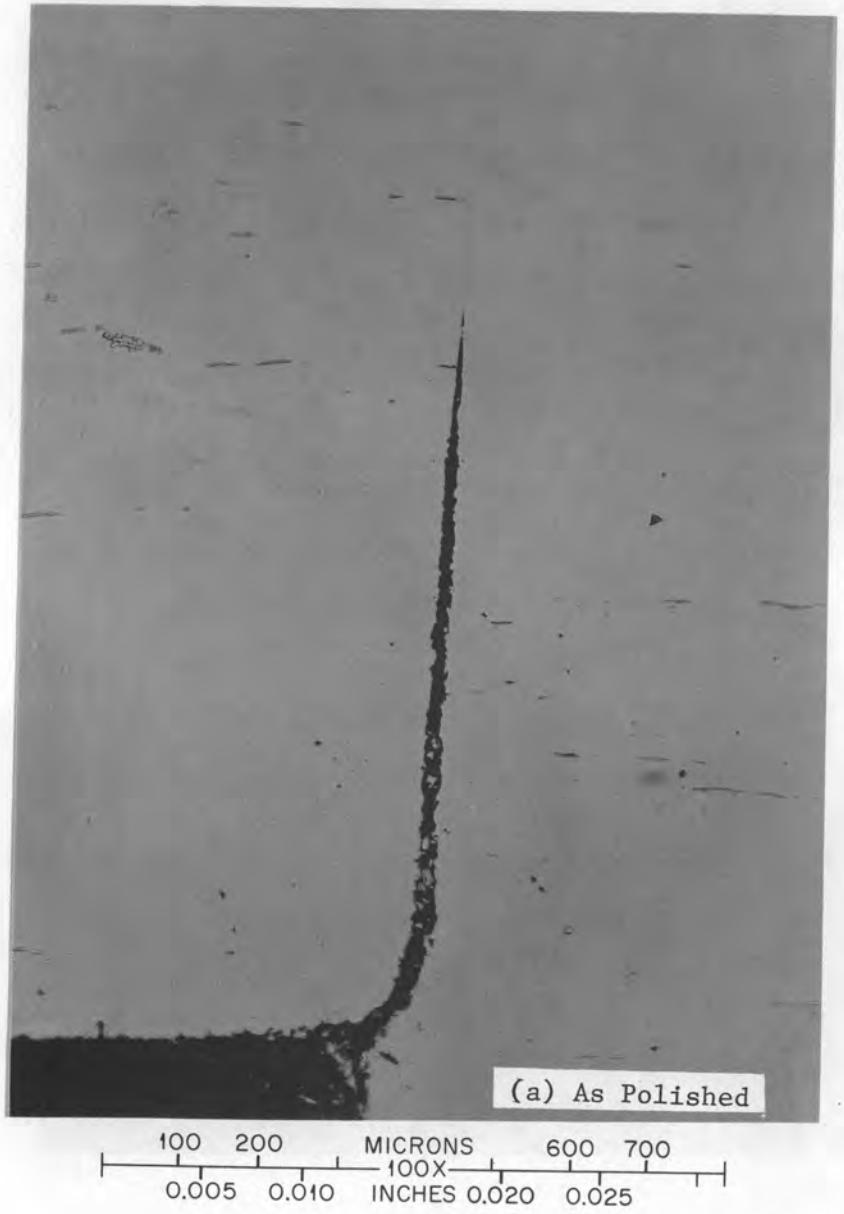


Fig. 28. Source N-PuBe-296, Top Weld, Outer Vega Steel Capsule, Sample 2. Etched.



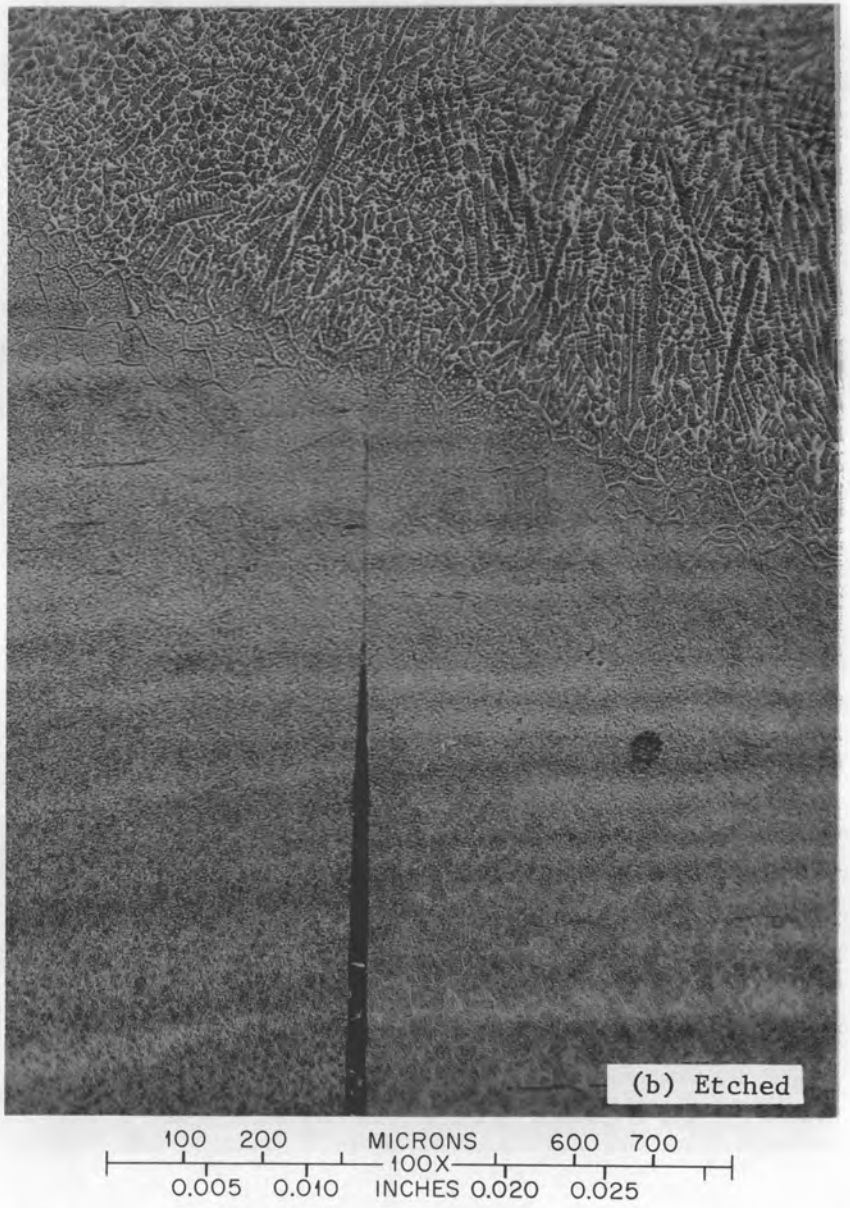


Fig. 29. Source N-PuBe-296, Top Weld, Outer Vega Steel Capsule, Sample 2.

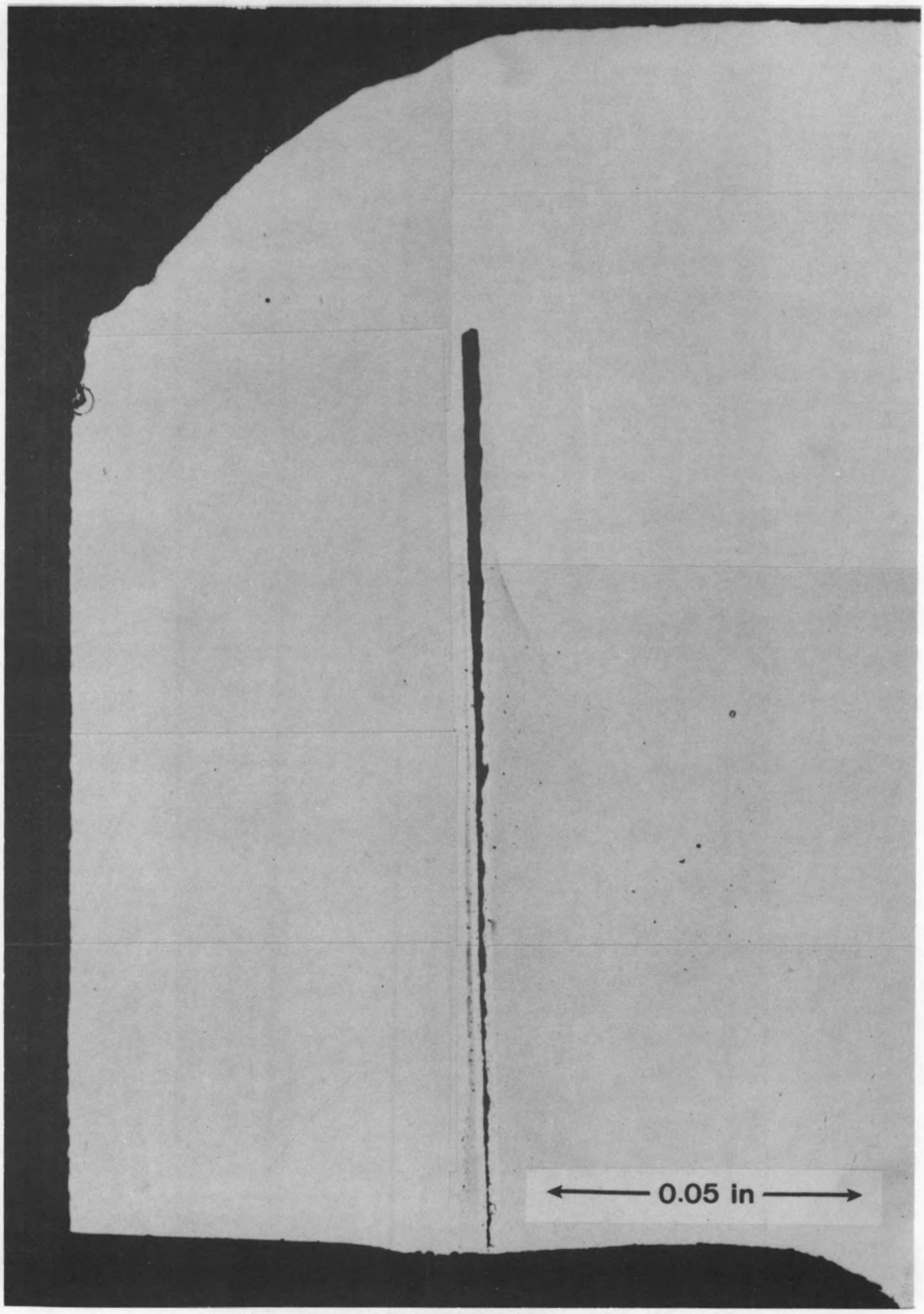


Fig. 30. Source N-PuBe-292, Welded Tantalum End Cap, Sample 1. As Polished.

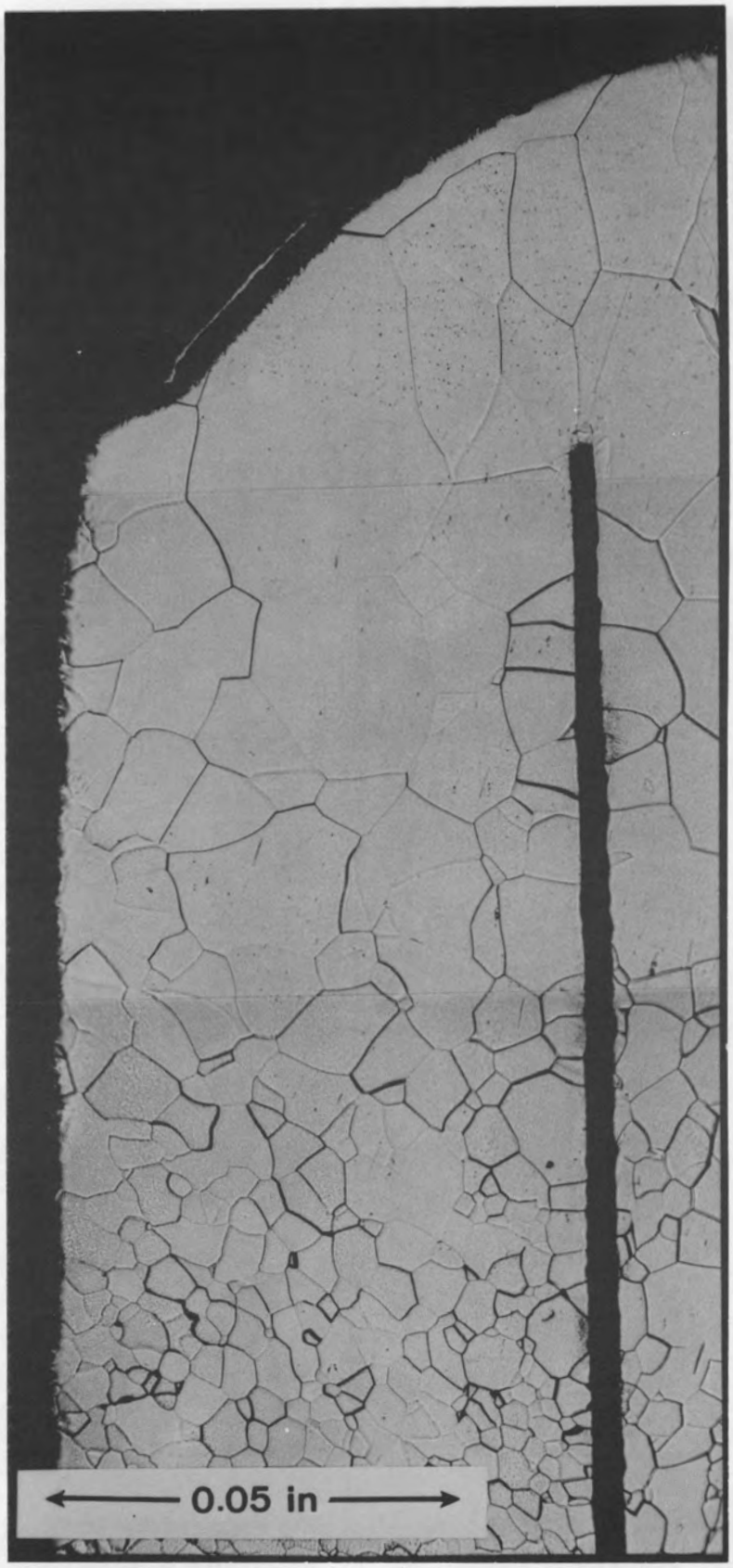


Fig. 31. Source N-Pu-292, Welded Tantalum End Cap, Sample 1. Etched.

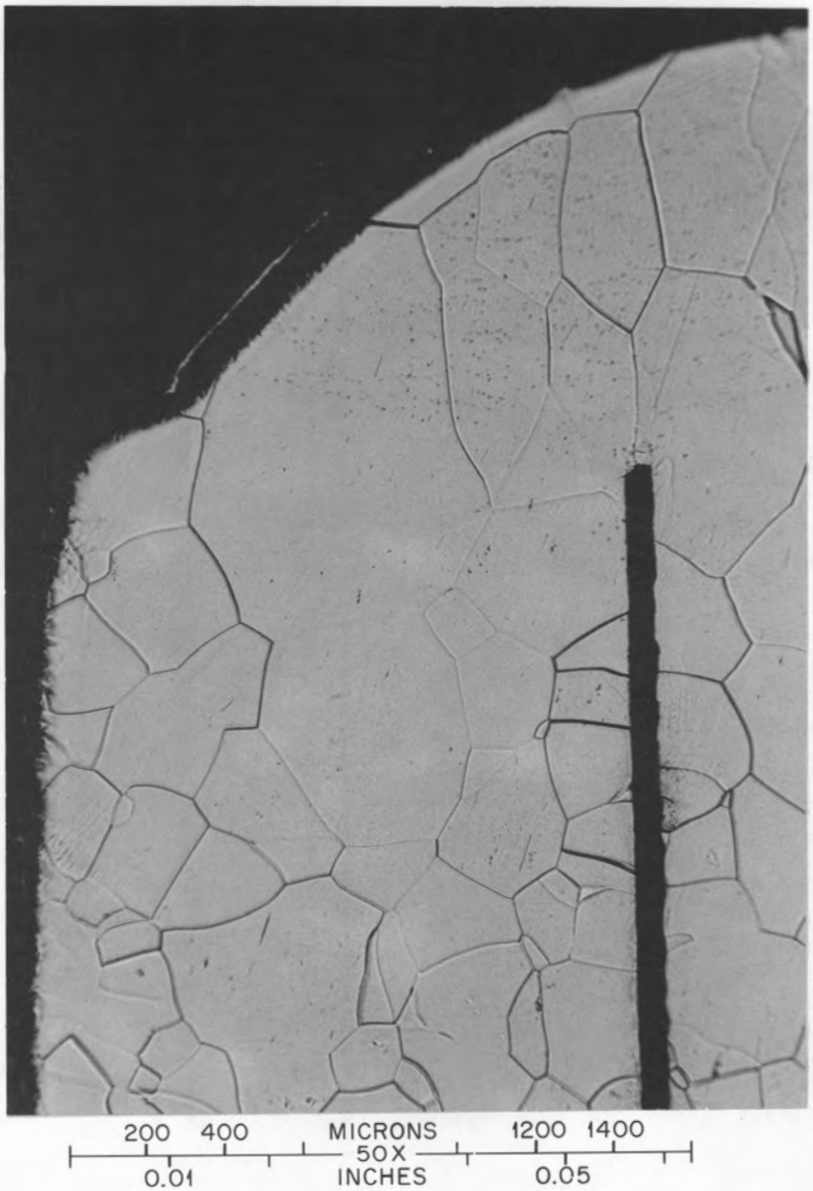


Fig. 32. Source N-PuBe-292, Welded Tantalum End Cap, Sample 1. Etched.

ORNL-TM-4561
UC-23 — Radioisotope and
Radiation Applications

INTERNAL DISTRIBUTION

1. P. S. Baker
2. E. E. Beauchamp
3-12. N. C. Bradley
13. F. N. Case
14. D. F. Cope
15. F. L. Culler
16. J. H. Gillette
17. K. W. Haff
18. R. F. Hibbs
19. E. H. Kobisk
20. E. Lamb
21-30. R. G. Niemeyer
31. C. L. Ottinger
32. H. Postma
33. M. E. Ramsey
34. R. A. Robinson
35. R. W. Schaich
36. R. D. Seagren
37. M. J. Skinner
38. A. H. Snell
39-40. Central Research Library
41. ORNL - Y-12 Technical Library
Document Reference Section
42-46. Laboratory Records Department
47. Laboratory Records - RC

EXTERNAL DISTRIBUTION

48-59. G. R. Grove, Office of Regulations, U. S. Atomic Energy Commission, Washington, DC 20545
60. N. F. Barr, Office of General Manager, AEC, Washington, DC 20545
61. D. C. Davis, Research & Technical Support Division, AEC-ORO
62. W. D. Sandberg, Deputy Director, Technical & Production Division, Savannah River Operations Office, AEC, P. O. Box A, Aiken, South Carolina 29801
63. S. H. Smiley, AEC, Washington, DC 20545
64. D. H. Turno, Savannah River Laboratory, E. I. DuPont de Nemours and Company, Aiken, South Charolina 29801
65. Research and Technical Support Division, AEC-ORO
66. Patent Office, AEC-ORO
67. Richland Operations Office, AEC, P. O. Box 550, Richland, Washington 99352
68-233. Given distribution as shown in TID-4500 under Radioisotope and Radiation Applications (25 copies - NTIS)