CLADDING AND DISPERSION FUEL DEVELOPMENT AT ORNL *

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INTRODUCTION

The presentation from Oak Ridge National Laboratory will be given in three parts. The subject and speaker for each part is as follows: 1) Dispersion Fuel Technology by J. E. Cunningham, 2) Fuel Reprocessing by J. W. Ullmann, and 3) Fuel Cycle Economics and Radiation Effects in Cladding Materials by D. A. Douglas.

UO_2-Stainless Steel Cermet Fuel Experience at ORNL - J. E. Cunningham

In reviewing the technology of the UO_2-stainless steel dispersion fuel, I shall confine my remarks mainly to irradiation experience accummulated over the past 15 years and attempt to point up shortcomings of this fuel type for application in FFTF.

As most of you know, the stainless steel—uranium dioxide dispersion fuel technology for the Army Water Reactor Program was developed in the early 1950's. The product that evolved for startup of the SM-1 reactor at Fort Belvoir, Virginia, was a brazed assembly of thin plates. A representative cross section of such a composite fuel plate is shown in Fig. 1 (LS-4278, Y-25961) for reference. It consists mainly of a 20-mil-thick fuel-bearing section with 5-mil cladding of type 304L stainless steel on each face to give an overall plate thickness of 30 mils. The fuel is incorporated into the fuel-bearing section in the form of 26 wt % or 19 vol % UO_2. The matrix material is type 302B stainless steel powder, which contains 2.5 wt % silicon.

These composite fuel plates were prepared by the conventional roll-bonding technique. Most of the fragmentation and stringering of the oxide that is apparent in Fig. 1 occurred during cold rolling (25% reduction) to final thickness. These composite fuel plates of 1950 design performed quite well in the 10-Mw thermal reactor at Fort Belvoir; in fact, they accummulated a total power output of over 60,000 Mwd (10% greater than the expected design lifetime of 15 Mwyr) before discharge at burnout. The operating conditions were not very severe. At a power level of 10 Mw, for instance, the heat flux was only 55,000 Btu $hr^{-1}ft^{-2}$. The average surface temperature was 450°F, while the maximum centerline temperature of the fuel was 650°F. The elements were subjected to over 3000 thermal cycles during startup and shutdown of the reactor, because the reactor was used primarily as an operator-training facility. The initial core loading accummulated a total reactivity lifetime of 16.4 Mwyr before discharge. The average fuel burnup in terms of depletion of ^{235}U atoms was 26% or 1×10^{21} fissions/cc.

The effect of irradiation on dimensional stability and structural integrity was checked by hot-cell examination of stationary fuel elements that had been removed after various levels of exposure in the reactor. The first fuel component was removed after 10.5 Mwyr life, the second at the end of the core life or 16.5 Mwyr, and the third after 19.5 Mwyr (higher exposure was achieved by reinsertion in Core II). In-cell examination revealed the following:

1) progressively greater tendency toward plate rippling with increasing exposure. Maximum ripple amplitudes of 23, 33, and 45 mils were observed, respectively, for 10.5, 16.5, and 19.5 Mwyr exposures.

2) A maximum plate increase in thickness of 3 mils or 10% occurred at the axial peak burnup position in the fuel element with the greatest neutron exposure. The burnup at this location was 62% depletion of ^{235}U atoms or 2.2×10^{21} fissions/cc.

3) The frequency of intergranular cracks in the cladding varied directly with burnup. In a few instances, cracking was so severe that whole grains were removed at the cladding surface. Some transgranular cracking was noted in the grain structure of the low-cobalt low-carbon stainless steel cladding in the high-burnup fuel element.

No evidence was found of matrix cracking nor sign of gross deterioration in these dispersion fuel plates. Yet, swelling of the order of 10% did occur under the modest operating conditions prevailing in the SM-1 reactor, and such behavior must be factored into the design of the FFTF if the dispersion is used for the driver fuel.

In 1962, we examined 34 dispersion fuel specimens contained in six NaK-filled capsules that had been irradiated to relatively high burnup in the ETR. These irradiation tests were initiated by BMI to evaluate various parameters in connection with their program to develop an improved dispersion fuel for service in the Army SM-2 reactor. The cermet specimens were clad with type 347 stainless steel and contained either UO_2 or UN dispersed in type 347 stainless steel along with small but varying amounts of burnable poison, in the form of B_4C, NbB_2, and ZrB_2. In addition, the behavior of fused and hydrothermal oxides was compared. The highly enriched fuel was incorporated in the fuel-bearing section to a nominal composition of 26 and 38 wt % UO_2 or 34 wt % UN.

Of the 34 specimens examined, 11 showed failure. These specimens varied in degree of failure from surface blistering to complete disintegration, as shown in Fig. 2 (LS-13896, R-9043, R-9045). The six specimens with the high fuel loading operated at a surface temperature of 950°F, and all six failed. Four of these were badly swollen and the other two completely disintegrated. Of the remaining five failed specimens, which contained 24 or 25 wt % UO_2, three were severely damaged and one developed a large blister over the fuel region.

The micrograph in Fig. 3 (LS-8870, R-11235) shows the degree of swelling noted at the edge of a specimen loaded with 26 wt % UO_2 and exposed to a fuel depletion of 74% of the ^{235}U atoms (2.9×10^{21} fissions/cc) at 1000°F. Matrix cracking was observed toward the center of this specimen. A typical picture of severe cracking is shown in Fig. 4 (LS-13897, R-11229). This particular specimen contained 34 wt % UN, operated at a peak temperature of 950°F, and achieved 63 at. % burnup.

Metallographic examination of the 23 undamaged specimens revealed no evidence of matrix cracking or blistering. These specimens all contained 24.2 or 26 wt % UO_2, operated in the temperature region of 420 to 600°F, and achieved burnups of 52 to 67% of the ^{235}U atoms (1.9 to 2.6 x 10^{21} fissions/cc). No significant difference was noted between specimens prepared with spherical (fused) or irregular-shaped (hydrothermal) UO_2 particles nor between specimens containing burnable poison of different chemical form. The microstructure illustrated in Fig. 5 (LS-9601, R-11306) is typical for these specimens and is quite similar in appearance to that found in the high burnup region of the SM-1 fuel elements.

From mid-1959 to 1962, we worked on a cooperative program with APDA to develop an improved UO_2-stainless steel fuel dispersion for use in Core B of the Fermi Reactor. The scope of the program embodied 1) characterization of spheroidal UO_2 particles, 2) fabrication of composite plates, 3) feasibility studies on nondestructive testing, 4) establishment of suitable assembly procedures, 5) measurement of fuel material mechanical properties, and 6) irradiation testing.

Early in the development program, the concentration of fuel was limited to 27.5 vol % (34.5 wt %) UO_2 in order to enhance radiation performance. Spheroidal fuel particles in the 105 to 149-μ size range were chosen, because we felt that this shape would minimize stress concentration and reduce radiation damage in the fuel matrix. Type 347 stainless steel was selected as the structural material because of its superior strength compared to other conventional grades of austenitic steels in the 800 to 1000°F temperature range.

The effect of cold deformation on the integrity and shape of the oxide in the finished plate was investigated and the results are summarized in Fig. 6 (LS-7260, Photo 52670). Cracking of the oxide was noted at all reductions investigated and stringering became pronounced at 20 to 30% reduction in thickness; hence, we decided to hot roll the fuel plates to finish size to minimize oxide fragmentation and stringering. During plate development, we also noticed a wide variation in the behavior during hot working of spheroidal UO_2 particles procured from different vendors, as illustrated in Fig. 7 (LS-7021, Y-38004, Y-38989). Our characterization studies showed that we needed some means to separate good from bad quality starting powder; hence, a new technique based on solid embedment and density was developed to serve this purpose. A specification was also written that allowed us to procure good quality spheroidal particles with a high degree of confidence.

Three instrumented capsules were irradiated in the MTR to assess the in-reactor performance of the Fermi Core B fuel plate. Each capsule contained two miniature plates, 2 x 0.5 x 0.116 in. The fuel-bearing section contained 33 wt % spheroidal UO_2 of 105- to 149-μ particle size, homogeneously dispersed in type 347 stainless steel as shown in Fig. 8 (LS-6986, Y-35413). Two thermocouples were attached to each specimen.

The nominal irradiation condition selected for in-reactor testing were three different burnup levels – 10, 20, and 25% of the ^{235}U atoms – and a surface temperature of 925 to 950°F. These conditions were chosen on the basis that Core B would be used to operate the Fermi reactor at a maximum core thermal power of 270 Mw. At this level, the calculated throughput burnup would be 20.7% of the initial ^{235}U atoms; hence, our burnup targets bracketed this level. Nominal fuel temperatures at the reactor center would be 750°F at the plate surface and 970°F at the fuel centerline. Hot spot and hot channel factors increased the centerline temperature to a maximum of about 1040°F.

The first capsule was discharged after an estimated burnup of 9.5% of the ^{235}U atoms. Subsequent measurement revealed, however, that the actual fission burnup was only 6.3% (average of two samples.) Consequently, an experience factor of 6.3/9.5 or 0.66 was applied in fixing the discharge dates for the remaining capsules. On this basis, the other two capsules were discharged after an estimated fuel depletion of

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16 and 26% of the ^{235}U atoms.

Specimens 1 and 10, which were contained in capsule ORNL-MTR-64-4 were unaffected by the irradiation. The burnup of ^{235}U atoms was 6.3%. The general appearance of specimen 10 is shown in Fig. 9 (LS-8898, R-4549). All four thermocouples remained operational throughout the test and indicated that the surface temperature on these specimens ranged from 800 to 950°F. The appearance of the microstructure was as expected.

Specimen 6 from ORNL-MTR-46-2 showed severe damage, as illustrated in Fig. 10 (LS-8901, R-11567). Two full-penetration holes were noted in the test plate. A major crack also developed at one edge of the specimen, as shown in Fig. 11 (LS-13898, R-11569). Marked swelling was noted in specimen 5 located at the bottom of the capsule. The appearance of specimen 5 in transverse section is depicted in Fig. 12 (LS-13899, R-13091). Gross swelling ($\Delta \ell = -13\%$) and matrix cracking are apparent. In this specimen the surface temperature ranged between 850 and 1150°F throughout the life of test and the measured burnup was 29% of the ^{235}U atoms.

Specimens 8 and 9 in the remaining capsule showed obvious swelling after a burnup of 33% of the ^{235}U atoms. This behavior is illustrated in Fig. 13 (LS-13903, R-14325) which shows specimen 9 in transverse section. Specimen 9 increased 6% in volume. A longitudinal section of specimen 8, which increased 27% in thickness is shown in Fig. 14 (LS-13901, R-14261). Temperature ranged from 950 to 1050°F during test; the average temperature was 1000°F.

These irradiation test results indicate that the temperature-burnup limitation of the improved stainless steel–UO_2 cermet fuel for Fermi Core B was definitely exceeded. Moreover, it should be noted that the specimen size was relatively small, so one would expect better performance due to the restraint offered by the cladding and frame material. In addition, control of particle shape and distribution of fuel appeared to offer only marginal improvement in performance.

During the development of the Fermi Core B fuel element, several advances were made in the development of nondestructive testing techniques to ensure compliance with specifications and as-manufactured product reliability. Among the developments were

1) an ultransonic technique that is capable of detecting core-cladding interface nonbond areas 1/8 in. diameter or larger,

2) an eddy-current technique capable of measuring the fuel plate thickness to within an accuracy of ± 1 mil,

3) a radiographic technique to detect UO_2 concentration inhomogeneities in excess of the specified ± 5% tolerance,

4) a spacing measuring device, based on recently improved eddy-current theory, that measures coolant-channel gaps between plates to ± 0.5 mil, and

5) methods for characterization of UO_2 powder so that particles of the desired shape and integrity can be procured from the vendors with a high degree of confidence that the material is of high quality.

An analysis of pertinent irradiation data on the performance of UO_2–stainless steel dispersions has been compiled at ORNL. These data are plotted in Fig. 15 (ORNL-LR-DWG-75068R3). It should be noted, however, that the temperature points plotted in this figure are based on estimated values since only limited data are available on actual surface temperature measurements during irradiation. The curve indicates that to achieve a burnup of 2×10^{21} fissions/cc the maximum surface temperature of the fuel should be less than 800°F. At a surface temperature of 1200°F, the maximum burnup that can be achieved before uncontrolled swelling occurs is about 0.8×10^{21} fissions/cc.

Ideally, a dispersion fuel should consist of components that are in equilibrium with one another as well as with their environment at all times. Chemical stability of the components, for instance, must be maintained in fabrication and during the lifetime of the fuel in the reactor. Plutonia and urania exhibit notable differences with regard to phase stability. Uranium dioxide is the oxide of lowest oxide content in the uranium-oxygen system whereas PuO_2 is the highest oxide in the plutonium-oxygen system. Silicon, a tramp constituent in stainless steel, will reduce PuO_2.

Finally, I would like to state that in my considered opinion, the PuO_2–stainless steel dispersion fuel is a questionable fuel concept for FFTF operation at 1200°F. At best its development to provide adequate service will be an expensive and uncertain proposition. Radiation-induced swelling of PuO_2 and severe thermal stress under condition of high power density are likely to lead to premature failure of the potentially embrittled stainless steel matrix and cladding. It is true that certain measures can be taken to improve performance at 1200°F, such as built-in void space to accommodate fission gas and reduce swelling. Unfortunately, no one has come up with a practical way of achieving this objective despite the fact that its effect on performance has been suggested for some time. Other parameters that potentially influence or upgrade performance are: 1) higher strength matrix and cladding material, 2) lower concentration of the fissile phase, and 3) lower temperature. Yet when all these factors are considered, the PuO_2–stainless steel cermet fuel concept appears marginal for application in FFTF at temperatures of approximately 1200°F.

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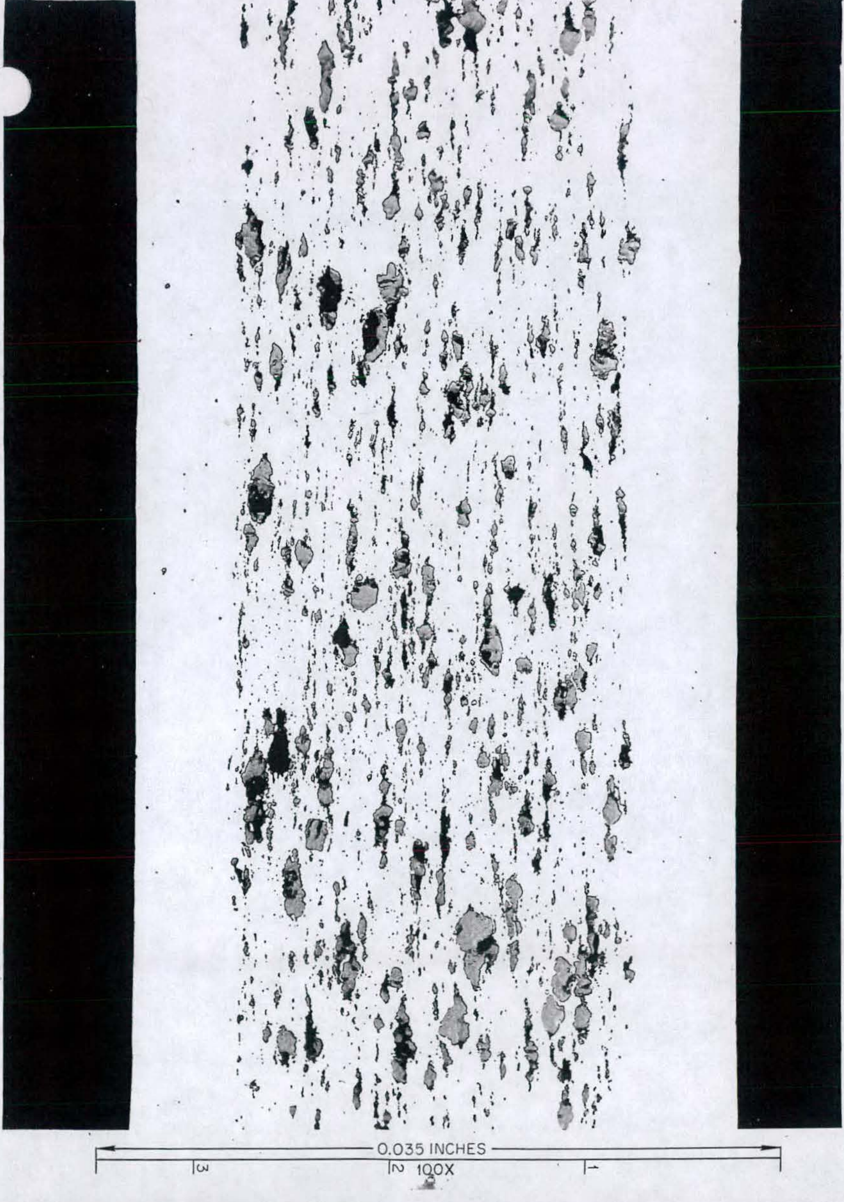
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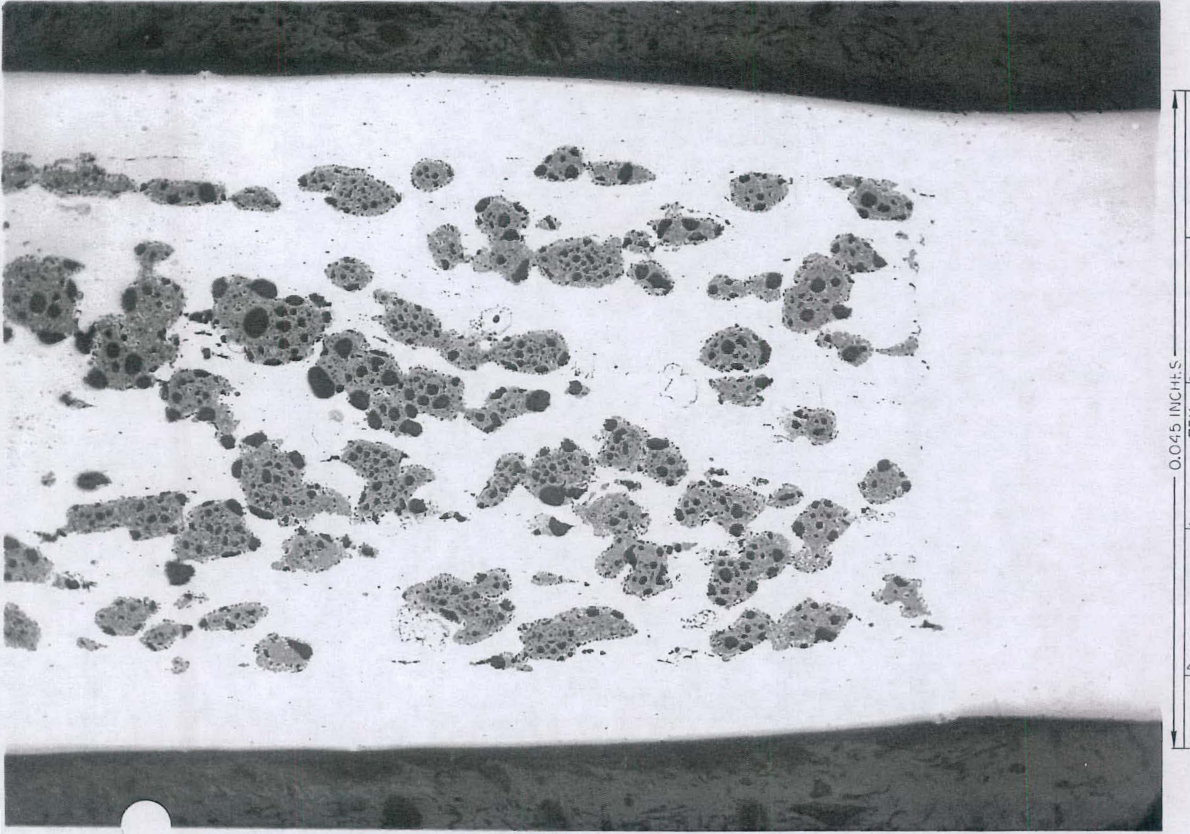
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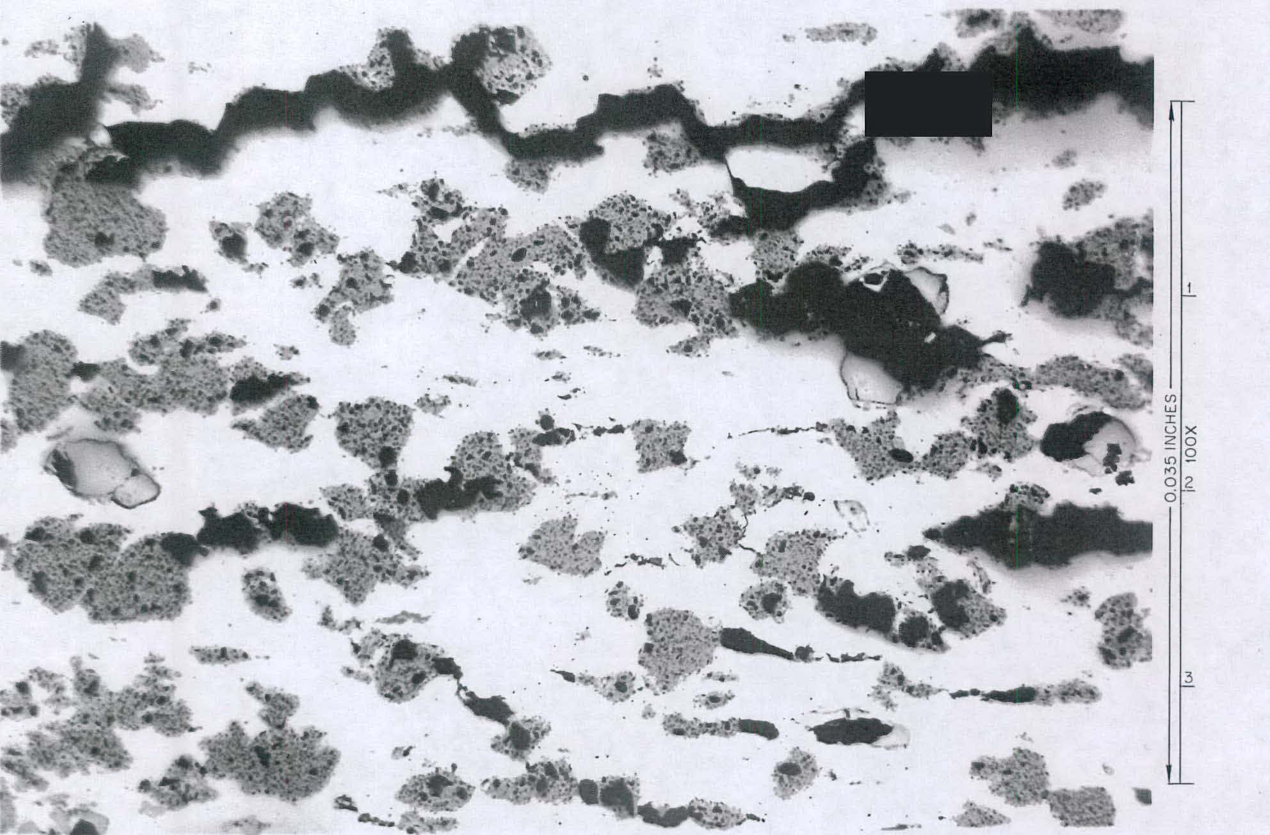
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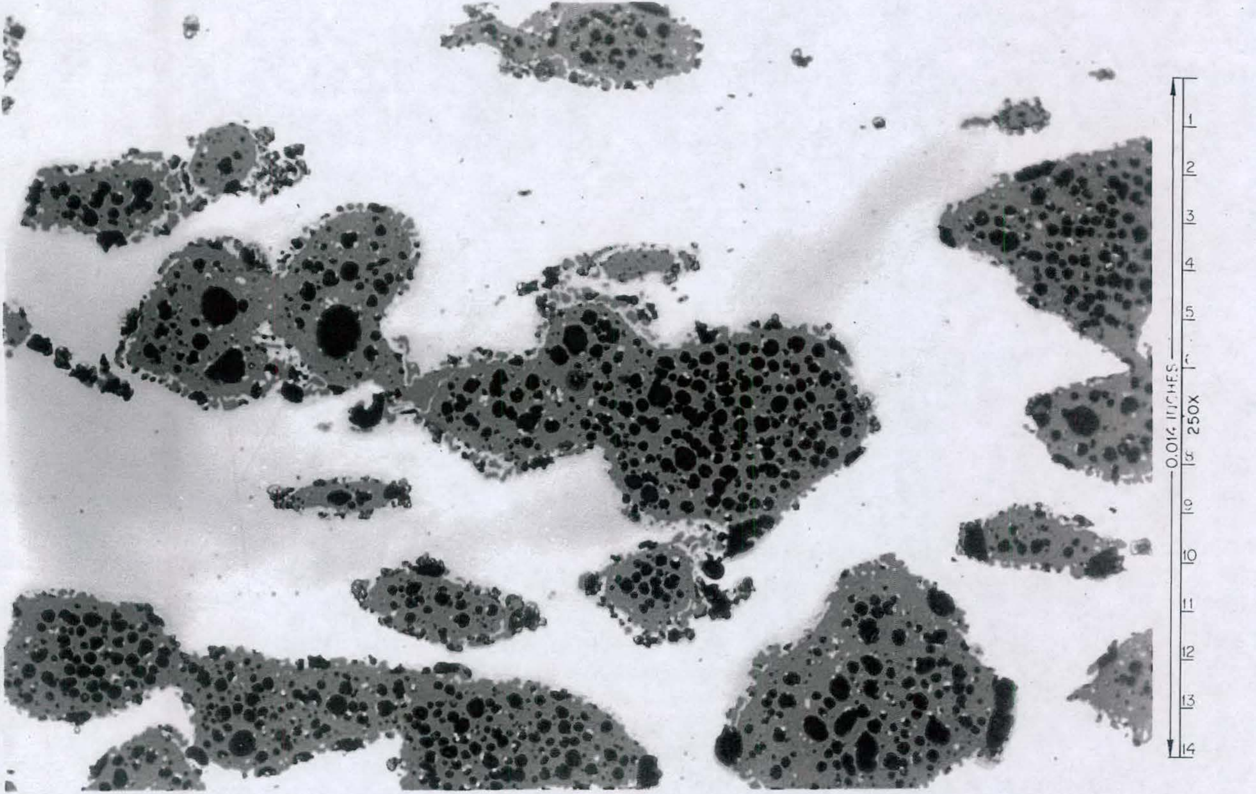
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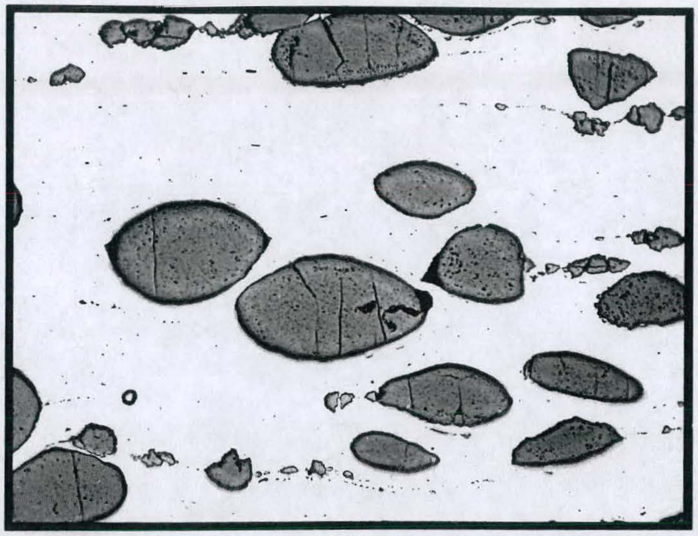

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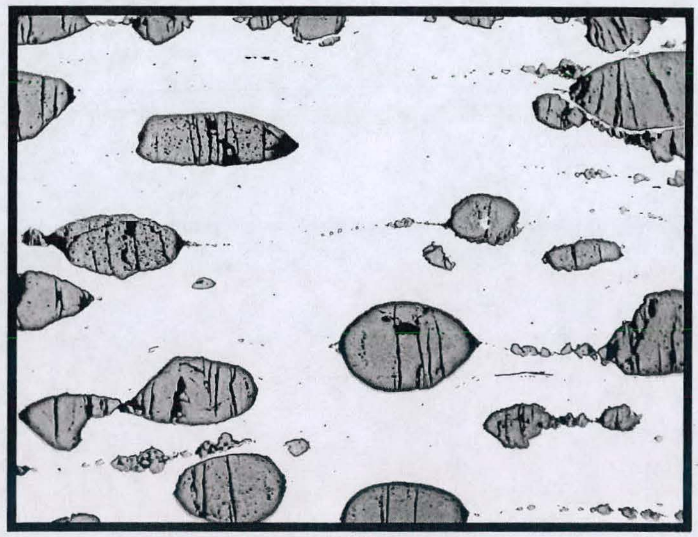

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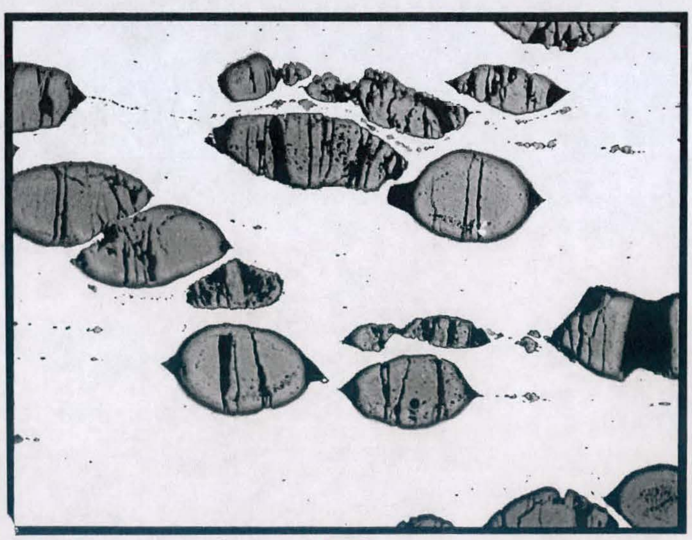

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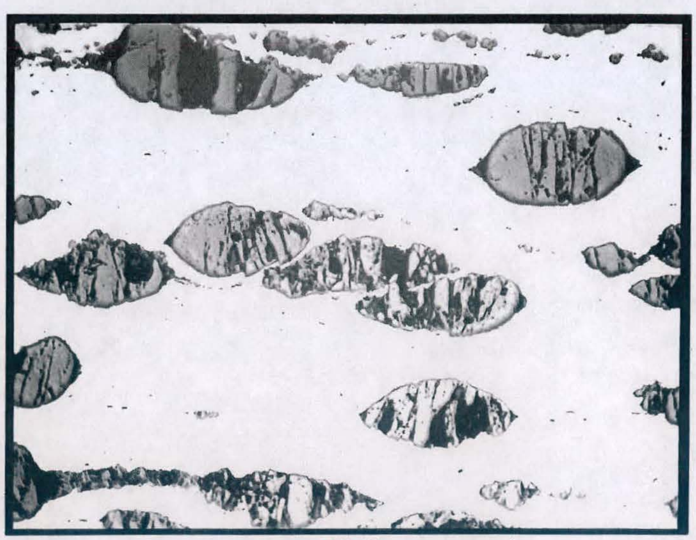

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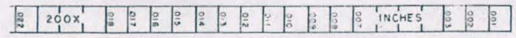
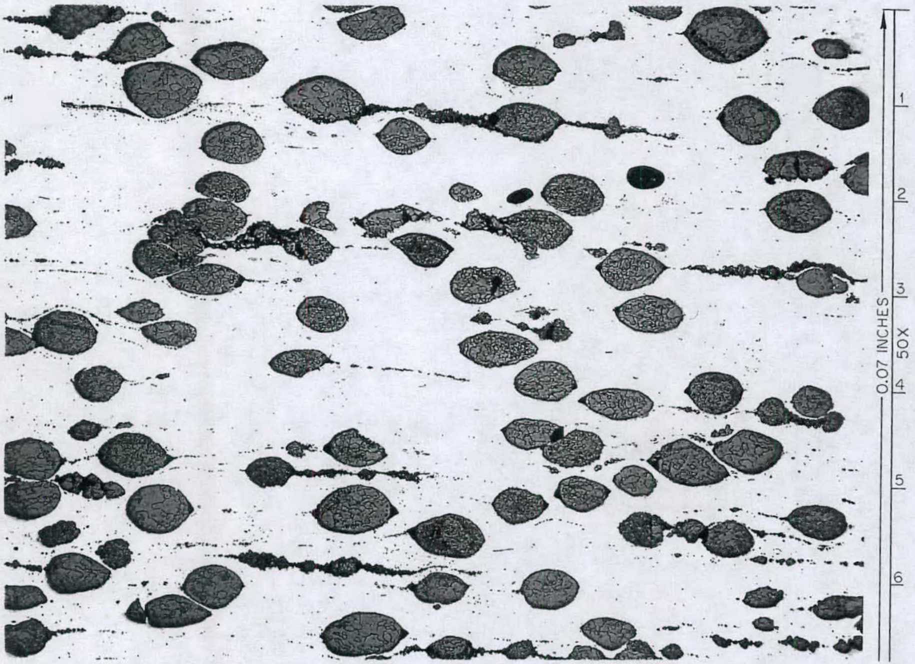


Effect of Total Cold Reduction on UO_2 Particle Geometry (Batch A-534)



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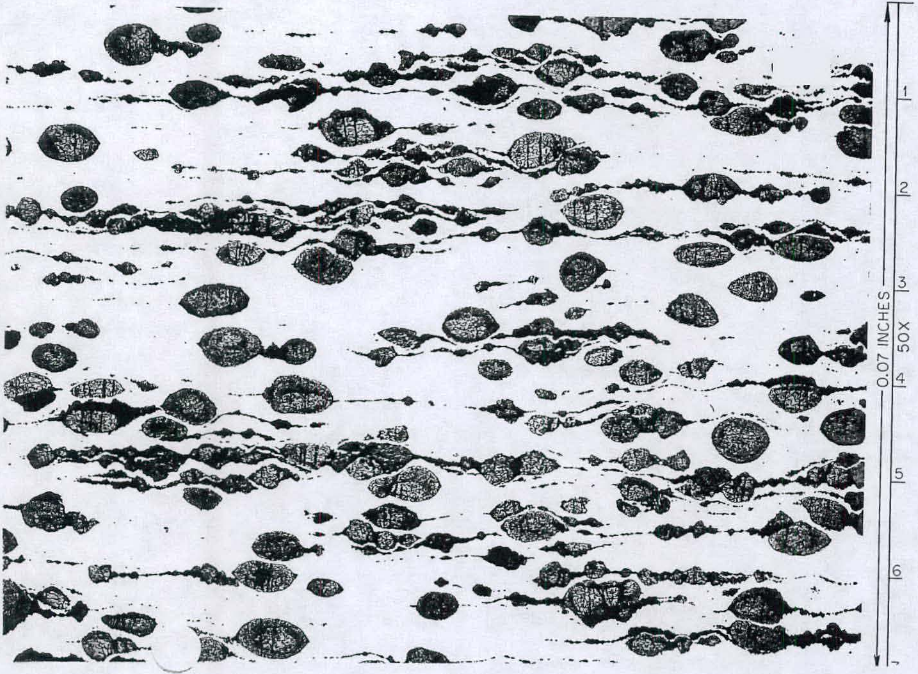
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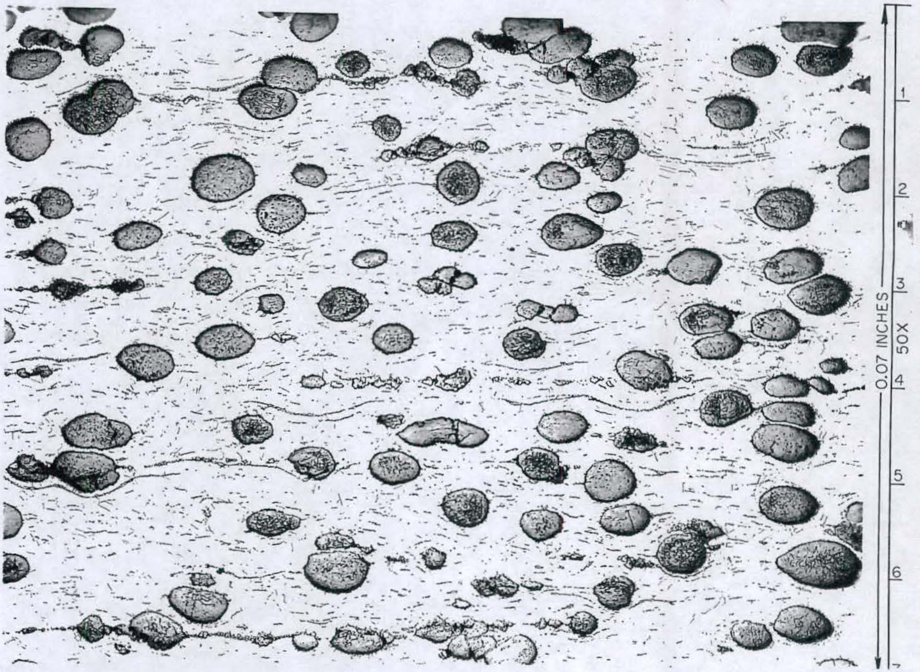
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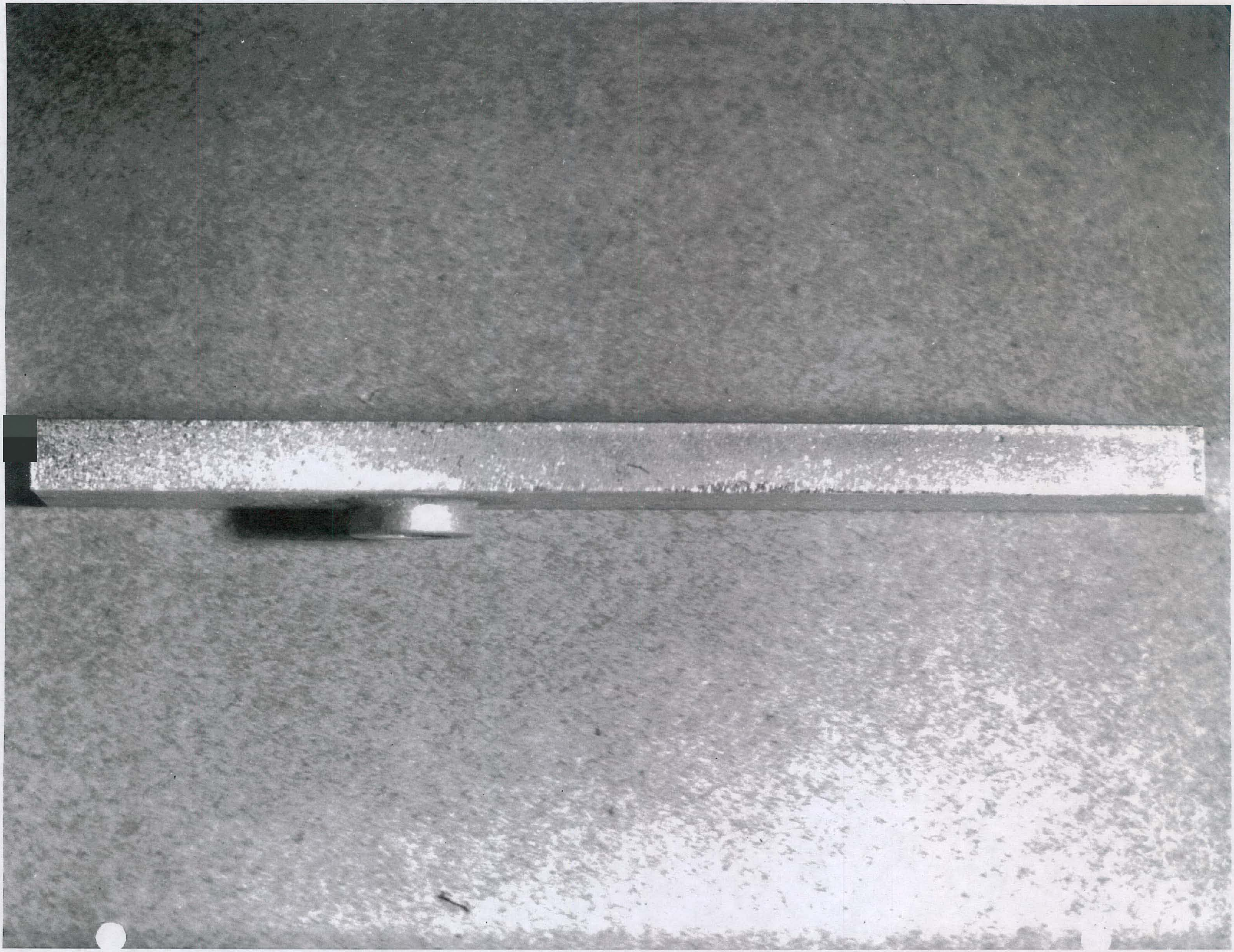

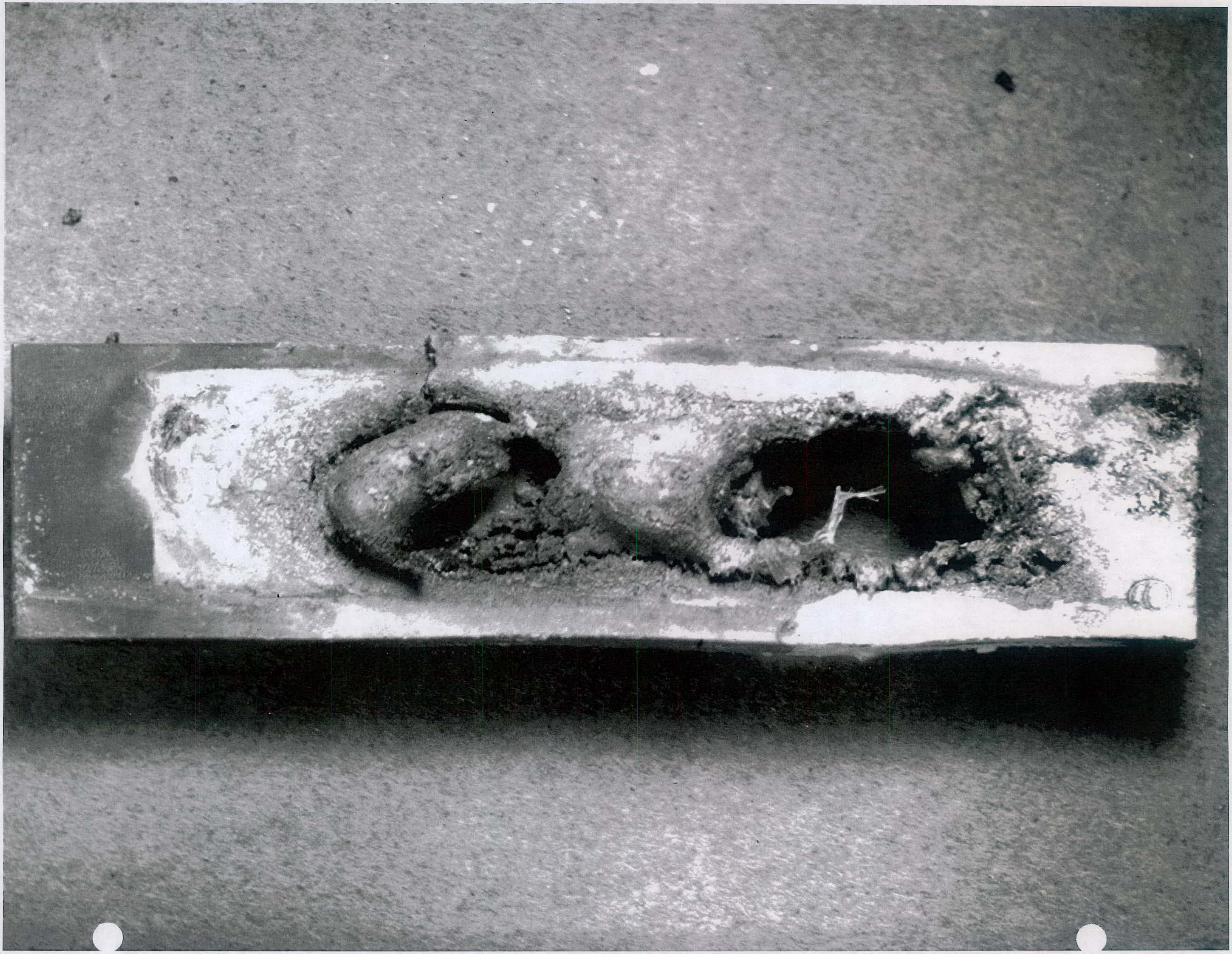

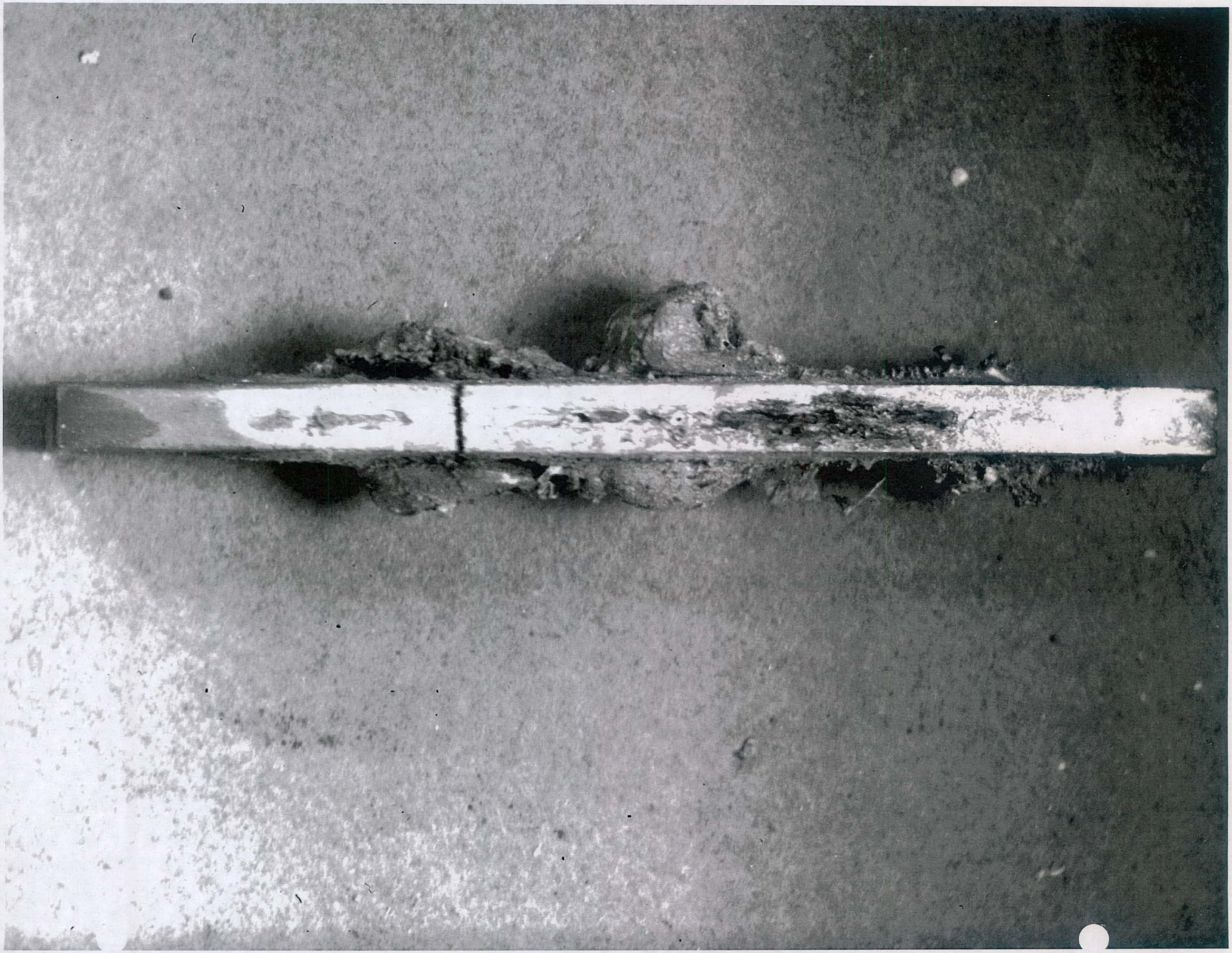

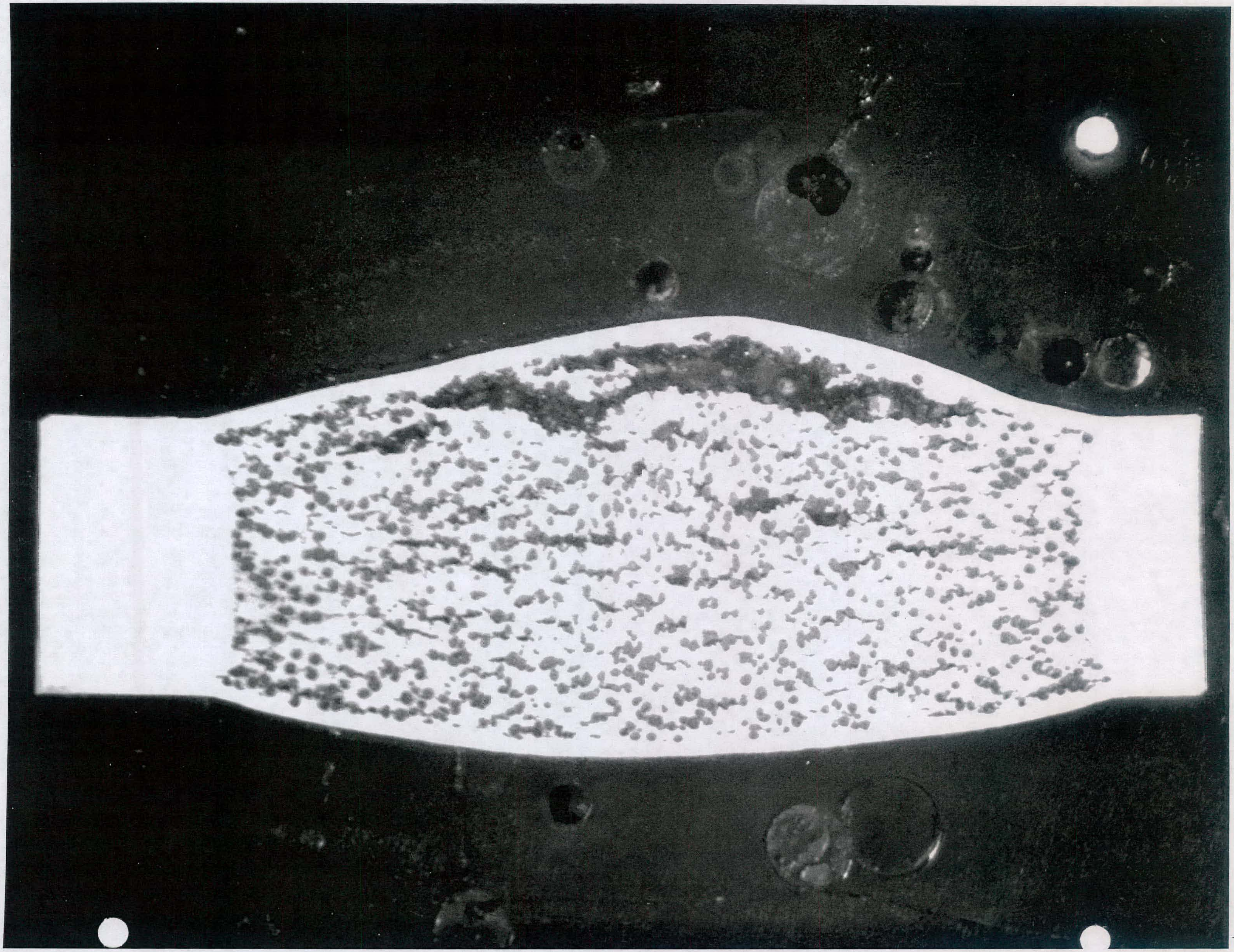

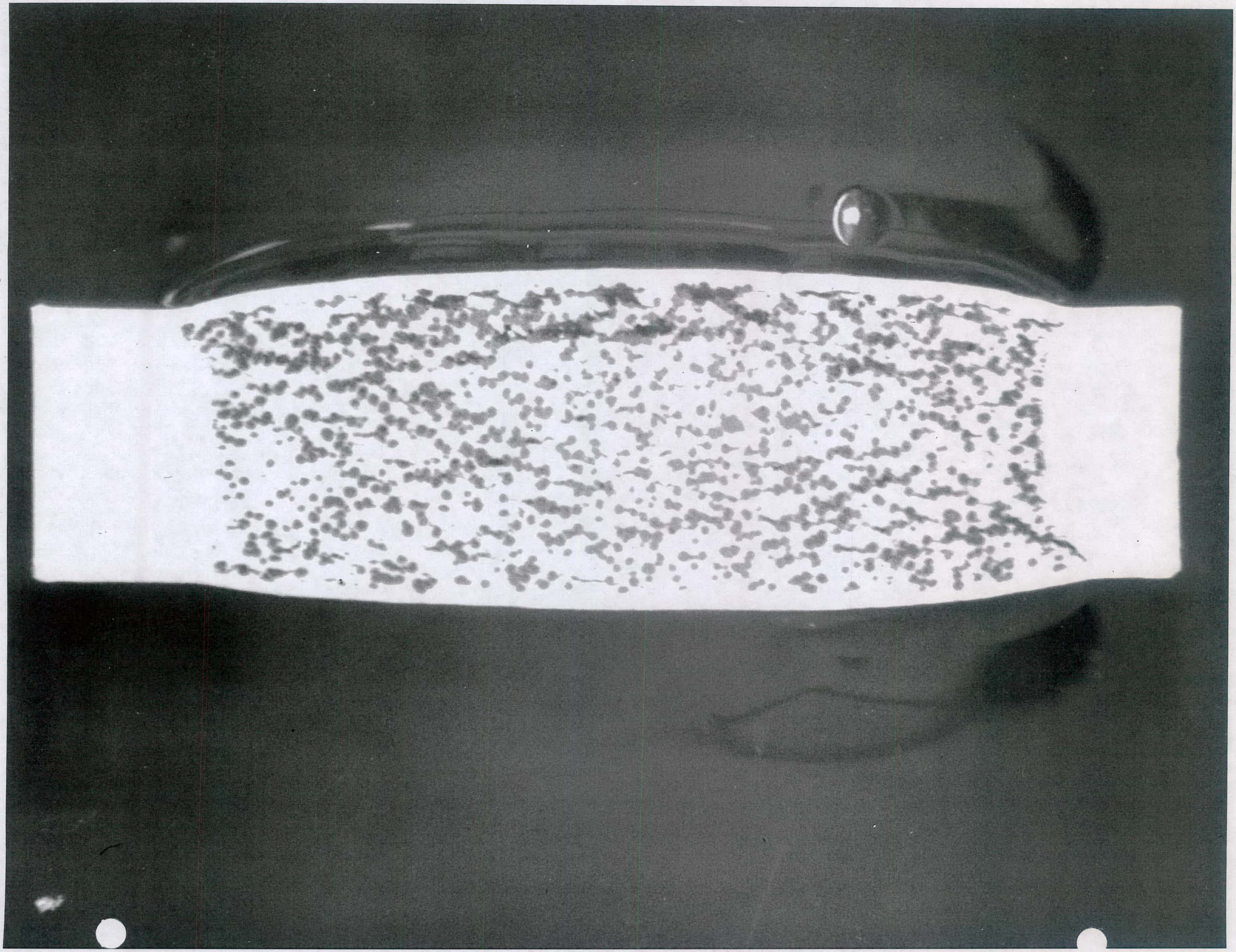

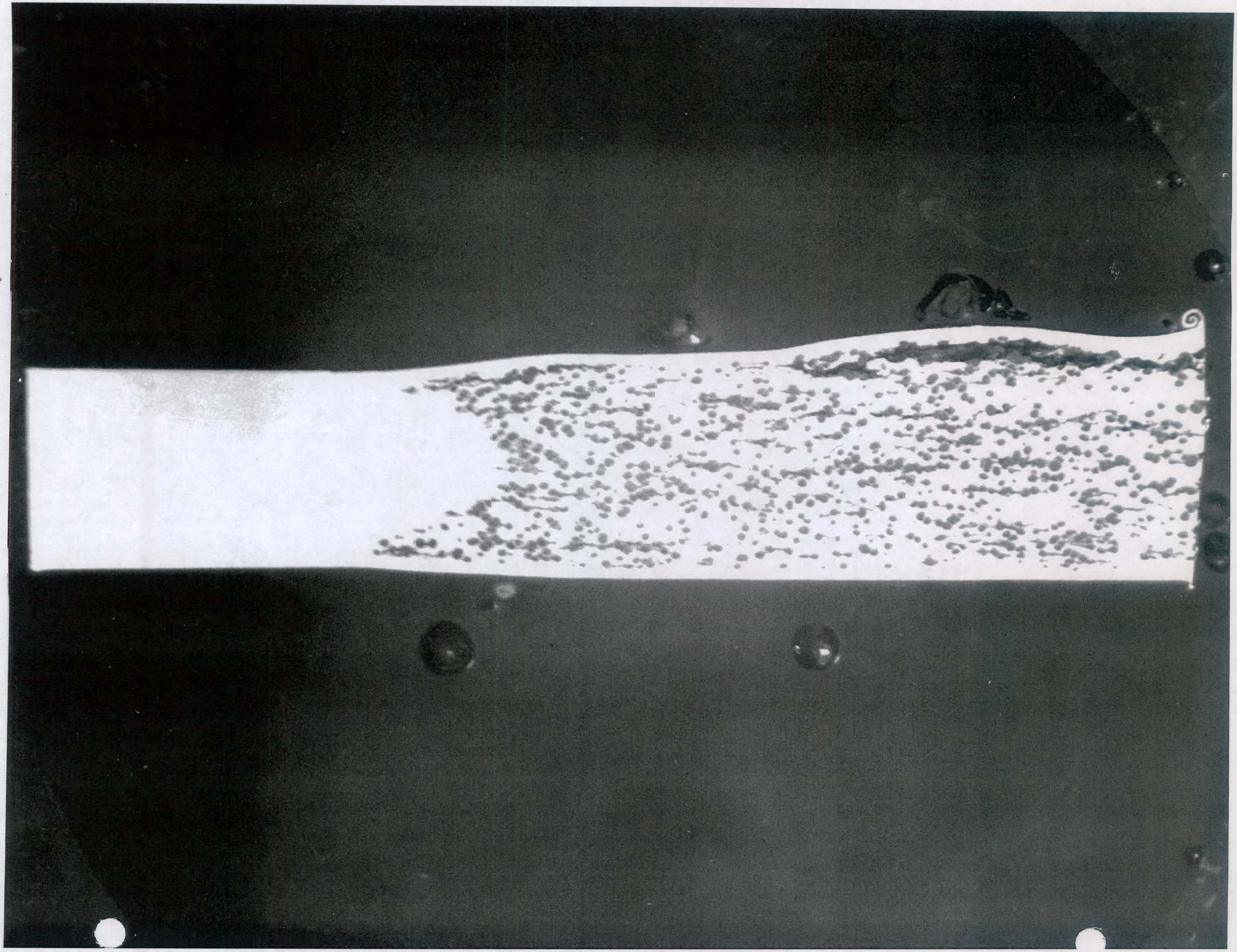

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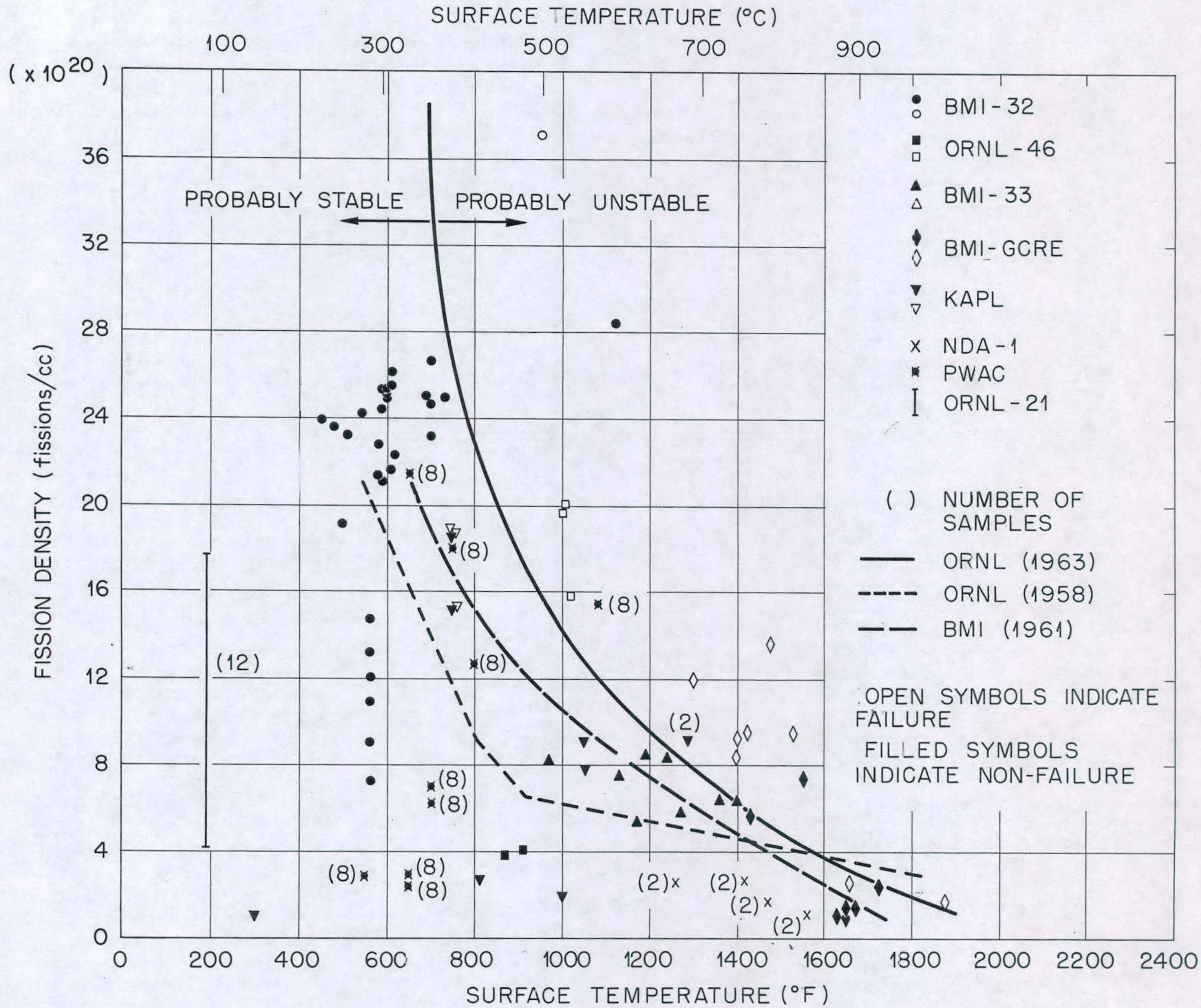

Irradiation Stability of UO_2 - Stainless Steel Dispersions.