1949

Indium replica for metallurgical examination

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Annapolis, Maryland: Naval Postgraduate School

http://hdl.handle.net/10945/31646

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METALLURGICAL EXAMINATION

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by

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Submitted in partial fulfillment
of the requirements
for the degree of
MASTER OF SCIENCE
in
MECHANICAL ENGINEERING

United States Naval Postgraduate School
Annapolis, Maryland
1949

This work is accepted as fulfilling

the thesis requirements for the degree of

MASTER OF SCIENCE
in
MECHANICAL ENGINEERING

from the

United States Naval Postgraduate School

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11324

PREFACE

The work herein described was accomplished in the Solid States Section of the Physics Division at the Oak Ridge National Laboratory during June, July and August of 1948. The purpose of this work was to discover a feasible technique for making metallurgical examinations of highly radioactive samples. The solution of this problem was sought through the use of a replica technique.

Invaluable aid in accomplishing the work herein described was furnished by the following personnel at the Oak Ridge National Laboratory:

Dr. D. S. Billington of Naval Research Laboratory, for his general direction of the work and constant advice and encouragement as well as his indispensible recommendation of the use of Indium metal.

Lieutenant W. Yatchmenoff, U. S. Navy, for much time and effort he spent in helping in the actual laboratory work, and in the ideas he offered which helped in this technique.

Dr. T. A. Read of Columbia University, who did the early work on this method and supplied me with much of the information concerning the methods tried, so as to eliminate many blind alleys of investigation.

Dr. S. Siegel, head of the Solid States Section of the Physics Division, who had the general responsibility for the work done.

Dr. L. T. Newman of Oak Ridge National Laboratory, who supplied much information on microscope and electron microscope techniques, as well as on the replica techniques now in use.

G. M. Carlton, who did most of the work connected with photomicrography.

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I. STATEMENT OF THE PROBLEM

The intense bombardment by neutrons, by other nuclear particles, and by high energy gamma rays to which materials are subjected within an active pile is known to have various effects upon the physical properties of these materials. Accurate measurement of the effects of pile bombardment on the physical properties of irradiated materials are fields for basic study. Such measurements are not, however, simple to perform on materials which have been subjected to pile radiation bombardment because of the induced radioactivity which most materials retain. For this reason, even a simple visual examination cannot be made in an ordinary way on most irradiated samples, but must be done remotely.

The Solid States Section of the Physics Division of the Oak Ridge National Laboratory has among its problems the investigation of the effects of pile radiation upon the properties of metals. Among the effects investigated will be those of radiation exposure upon the visible microstructure of materials such as would be detectable by the usual microscopic metallographic techniques. For this reason, it has become important that some method for metallographic examination of highly radioactive samples be developed.

II. CHOICE OF SOLUTIONS

Consideration has been given to the adaptation of a periscope microscope for use behind lead shielding in a "hot laboratory" to view and photograph radioactive samples. Such a system may be used, but it has two disadvantages: first, the effect of high level radioactivity on lens glasses and lens cements; and second, the difficulty in handling the samples by remote control each time they are viewed. The effect of radiation on glass would be to discolor it, causing obvious difficulties with microscope objectives.

For the above reasons, consideration is here given to the use of a replica technique for the examination of "hot" samples. The basic problem of this report was to find a method whereby an intermediate substance would take the impression, and then to get a general measure of the magnification limits at which the replica would give the desired information. Consideration was given to the development of replica techniques already in use. These were the so-called Faxfilm and Heidenreich methods, details of which are found in references below. These were developed essentially for the electron microscope, and involved the use of plastics to take the impression. These methods were not considered appropriate, since the effect of radiation on plastics is to decompose them, and unstable or unreliable impressions could result. Furthermore, the methods involved manual techniques not easily adaptable for remote-control handling of the plastics and solvents.

It was decided to investigate the possibility of using a soft metal to take the impression. Dr. D. S. Billington, of the Naval Research Laboratory, recommended the use of Indium metal because of its softness and extreme malleability at room temperature. Indium is considerably softer than lead and shows little or no tendency to work harden. At room temperatures, in foil form, unetched, it shows no visible crystal structure (that is, appears homogeneous with no grain structure whatsoever) at above 550X magnification. Furthermore, there are no known nuclear reactions which would make the Indium radioactive to a detectable degree if it were used as planned, for impressions of metal into which radioactivity has been induced by exposure to pile radiation. Obviously, the impressions of the metal specimens would be made after the removal of the specimens from pile radiation bombardment.

III. DETAILS OF TECHNIQUE

It was first necessary to find a proper backing for the Indium sample, since the metal itself is too soft, as well as too expensive, to use in lump form. Steel was tried as a backing. Glue was used as an adhesive, and proved quite unreliable. The flow of the Indium after taking the impression easily broke the adhesion of the glue. An attempt was also made to have the Indium adhere to a polished steel block by applying pressure. The performance of this method was also unreliable. Sometimes the Indium adhered, sometimes it did not, and often it would adhere in part only.

Finally, plastic backing blocks were tried. Several different plastics were used, among them were Lucite, Bakelite, and Polystyrene. The Polystyrene proved by far the most promising. Indium could be made to adhere to a Polystyrene block by using benzol as a solvent with pressure applied by a polished steel block. Details of the technique are as follows:

A. List of Materials

Materials necessary include:

1. A sample of Indium foil clean of oxide, .040 inches thick (in this case it was disc shaped, .3 inches in diameter).
2. A piece of Polystyrene rod about 1 inch in diameter and 1 inch long, with one of the flat ends polished smooth.
3. A cylindrical stainless steel block about the same size as the Polystyrene, with one of the flat surfaces highly polished to

show smooth surface at magnification of 500 or more diameters.

4. Benzol as a solvent.

5. A small, hydraulic, hand-operated press such as is used for plastic mounting of metallography samples. This should have a pressure dial calibrated to about ten per cent accuracy.

B. Preparation of Sample

The following are the steps used in preparing a sample which was eventually used to take the replica: See Illustration I, page 9.

1. The smooth, flat, end surface of the Polystyrene cylinder is wet with benzol and allowed to stand for about one minute. This surface will become very sticky.

2. The disc of clean Indium (.040 inches in thickness and .3 inches in diameter) is then sandwiched between the sticky surface of the Polystyrene and the polished end surface of the steel block. The steel surface should be wet with benzol just before making the sandwich to aid in parting the steel from the Indium later.

3. A force of about 250 pounds is then applied to the assembly, with the press squeezing the Indium flat between the steel and softened Polystyrene surface for about fifteen seconds. This is equivalent to about 3500 pounds per square inch on the basis of the original area, and it presses the Indium to about one-quarter of its original thickness.

4. Careful removal of the steel block leaves the Indium adhering to the Polystyrene with a smooth surface outward. This is the replica blank, and should be allowed to dry thoroughly (of the benzol solvent). A matter of about two days at room temperature seemed sufficient. Of course, several of these blanks could be

made at one time and stored, taking care not to scratch the soft, smooth Indium surface.

C. Taking Replica Impression

The following procedure was followed in taking the replica impressions: See Illustration II, page 10.

(This would normally involve the only work necessary with the radioactive sample in the "hot laboratory" behind any shielding). The specimen, polished and etched, if necessary, as discussed later, is pressed for about ten seconds with a force of from 300 pounds to 500 pounds in the press into the surface of the Indium of the Polystyrene block. This represents a pressure equivalent to about 4250 - 7000 pounds per square inch for the .3 inch diameter specimen used. Care should be taken that the specimen area be smaller than the area of the Indium blank to avoid pulling the Indium away from the Polystyrene when the pressure and sample are removed. The resultant replica in the Indium surface is, of course, a mirror image of the actual metal surface it represents.

IV. RESULTS

Good reproductions up to 500 diameters have actually been observed, and identical areas on original and replica compared on adjacent microscopes. Most specimens show excellent reproduction at 100 diameters, and good to excellent reproduction at 250 diameters. The enclosed photomicrographs of specimen and replica are relatively poor examples of what this technique has been able to do. They were taken early in the development, and later, further pictures were impossible to make since the photomicrograph equipment was disassembled.

With regard to etchant, the questions of whether to etch, or simply to polish a sample, what etchant to use, and how deeply, are actually ones that should be empirically determined on the unirradiated specimen beforehand. Generally, the final polishing and etch, if necessary, will take place after the specimen has been irradiated, and of course, before taking the replica. This can be handled in most "hot" laboratories with the equipment now available. However, if handling techniques to insure against inadvertent scratching can be used, it might give some significant information to try polishing and etching before irradiation.

The answer to the problem of the depth of etch in a particular case is determined, to a large degree, by the type of information desired from the specimen, and the nature of the specimen itself. In general, an etchant is not necessary if the surface is a matrix of components of sharply contrasting hardness, like the Uranium alloy of photomicrograph in Illustrations III and IV, pages 11 and 12.

Polishing alone brought out sufficient contrast using the replica technique. A relatively deep etch is, however, necessary for a particular magnification as compared to that needed for ordinary direct microscopic examination. The actual pressure which gives the best impression is somewhat dependent upon the depth and character of the etch. It may seem quite expensive in the expenditure of Indium metal to try to determine in each particular type of specimen, by trial and error, the pressure and etch best suited. All of the Indium used for this work is, however, recoverable. As soon as the particular sample is not needed, the Indium may be dissolved off the Polystyrene with benzol, washed, and recast into sheets by heating in vacuo. This should be a relatively simple procedure since Indium melts at 157.2 degrees centigrade.

Examination of the 250X magnification of the Uranium alloy of the following photomicrographs will indicate that perhaps there is more detail showing on the replica than is showing on the original. Most of this excess detail is explained by the amount of Indium oxide on the surface of the original Indium foil. This should not be too difficult to remedy. A very short investigation of the problem of removal of the oxide showed that about 0.2 normal solution of hydrochloric acid attacked the oxide in preference to the pure metal. This chemical method of removal of the oxide was investigated after the above photos were taken, and proved helpful for use on the foil before making the replica blank. However, some of the pits left behind by the dissolved-away oxide remained in the Indium surface even after pressing it to the Polystyrene with the polished steel block. It seems reasonable to believe that smooth oxide-free foil can be obtained (perhaps shipped, immersed in kerosene) from the commercial sources of the pure metal. Furthermore, there is probably a more favorable concentration of hydrochloric acid for cleaning the foil surface.

The following materials were examined microscopically, using two adjacent microscopes at up to 550X magnification, to compare identical areas on the replica and original. These areas were delineated by microscopic identifying scratches.

1. Beryllium-copper - Etched. (Photomicrographs included herein on Illustration V, page 13.) Showed excellent reproduction to 250X, and good reproduction to 470X.
2. Uranium alloy - (Photomicrographs are included herein on Illustrations III and IV, pages 11 and 12.) Showed excellent reproduction to 250X.
3. Stainless steel - Etched only to show the large grain size, gave excellent reproduction to 450X.
4. Stentor stainless steel (12% retained Austenite) - Specially

etched. Showed sufficient detail at 550X to give a good estimate of the percentage of retained Austenite for which the etchant used was designed. This operation was considered the most strenuous demand upon the replica technique.

V. CONCLUSIONS AND RECOMMENDATIONS

I believe the major conclusions of the effort described above are:

1. That the use of Indium for metallographic replica work is theoretically and practically possible;
2. That the development of this technique will probably be a very economic method of making metallographic examination of many irradiated metals;
3. That this method is particularly well adapted to the unique problem of making replicas behind shielding.

For security reasons, I have not discussed any actual investigations of radiation effects, or the results thereof; the purpose here being to develop a technique which is useful in such tests, but may, conceivably, also be of significance elsewhere in the field of metallography or microscopy. The following closely related problems are considered important for further investigation:

1. To find methods for more positive adhesion between the backing block and the Indium;
2. To find means of providing for a more oxide-free and smooth initial surface for the Indium;
3. To study the effects of various types and depths of etchants, and various pressures on the quality of the reproduction;
4. To test the effectiveness of the method on many more metals and alloys.

ILLUSTRATION I
PREPARATION OF SAMPLE
FULL SCALE

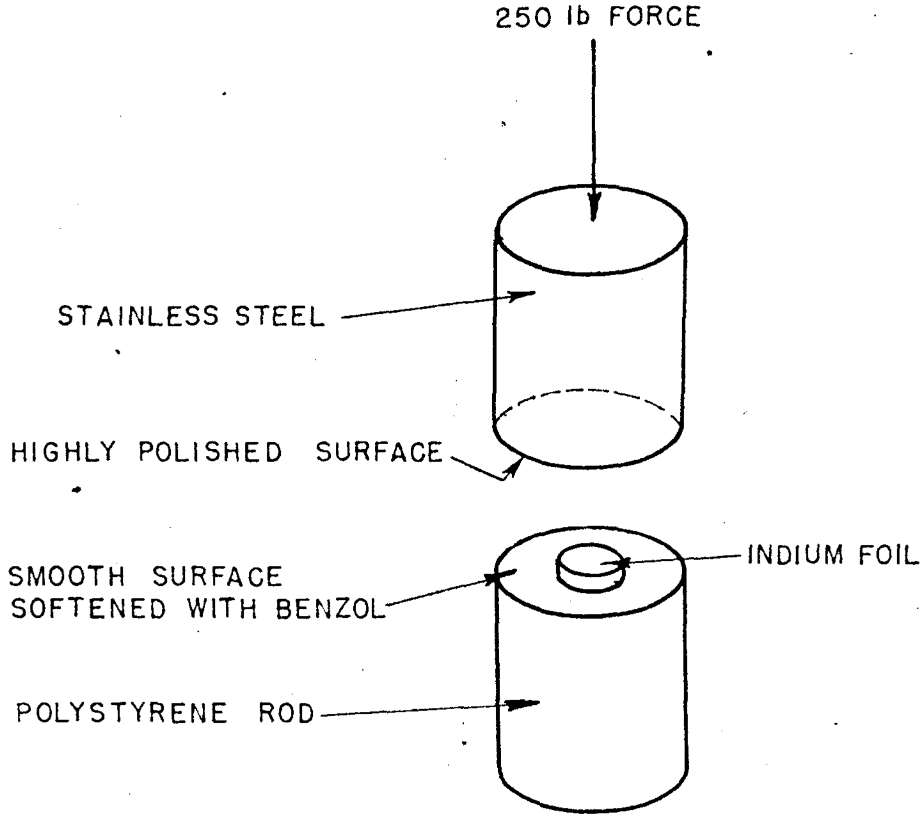


ILLUSTRATION II
TAKING OF REPLICA IMPRESSION
FULL SCALE

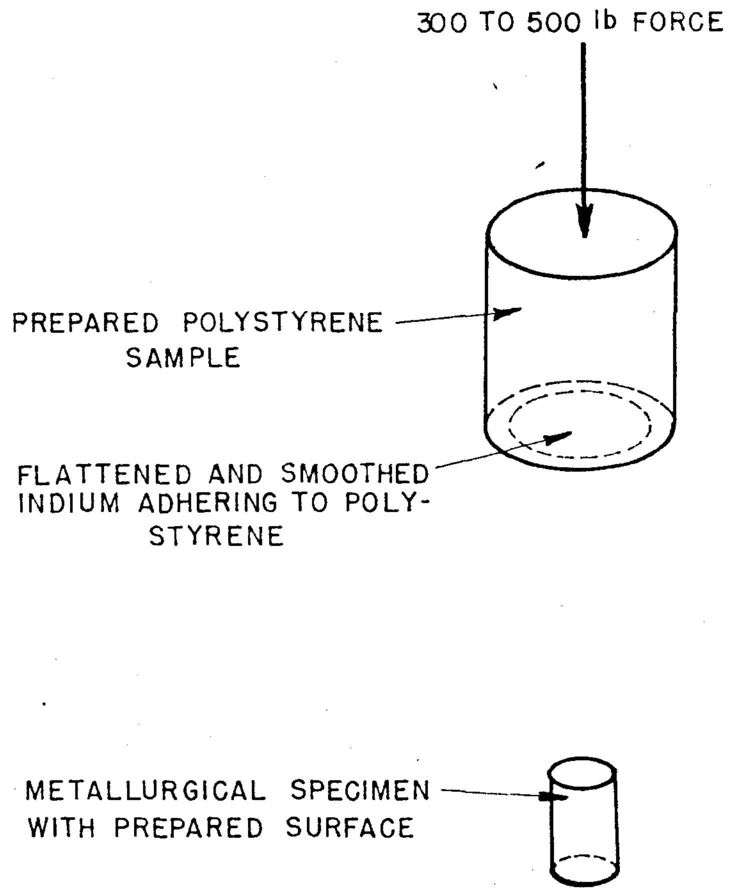
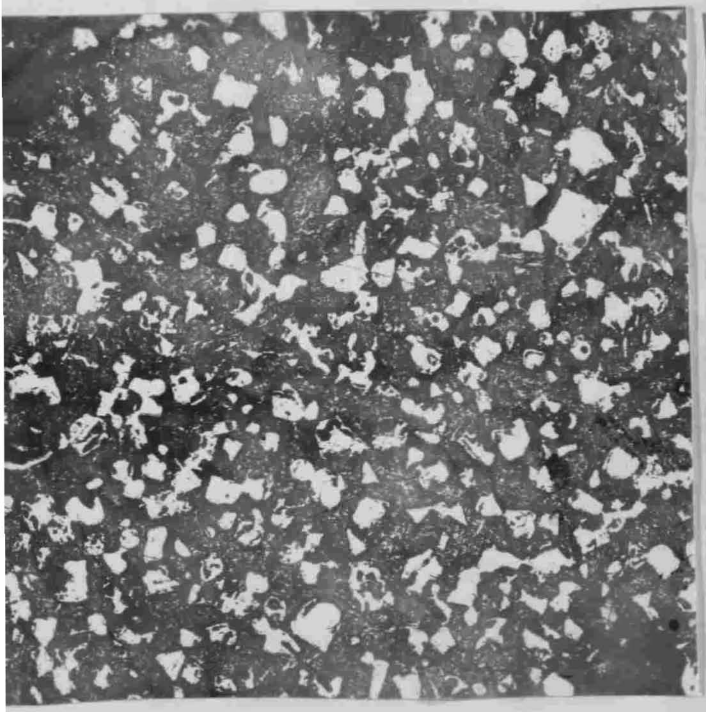
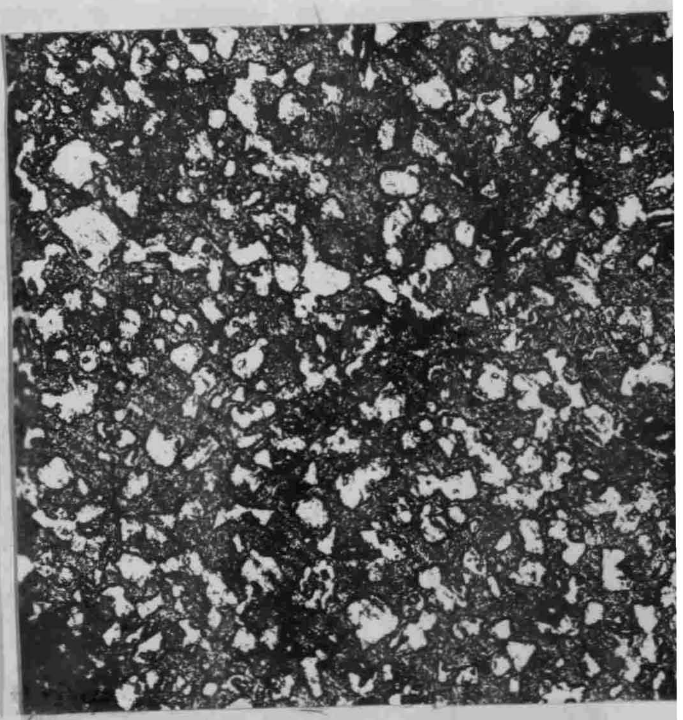


ILLUSTRATION III





X

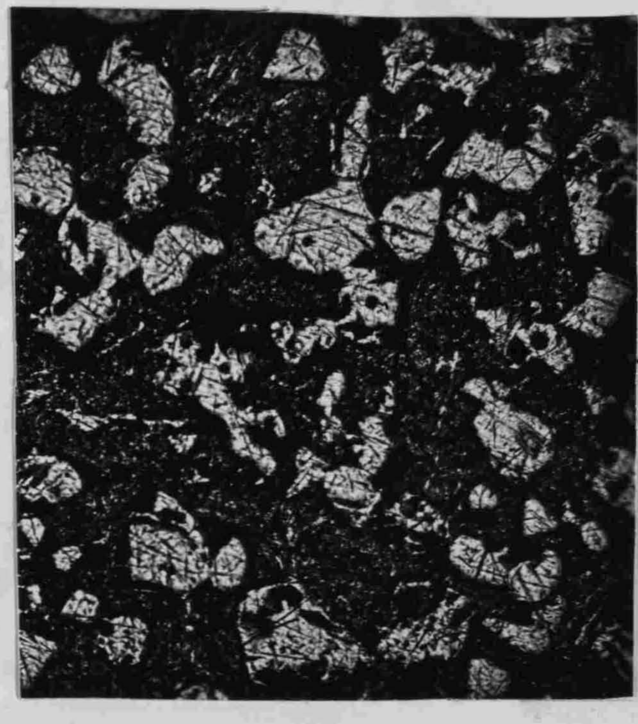
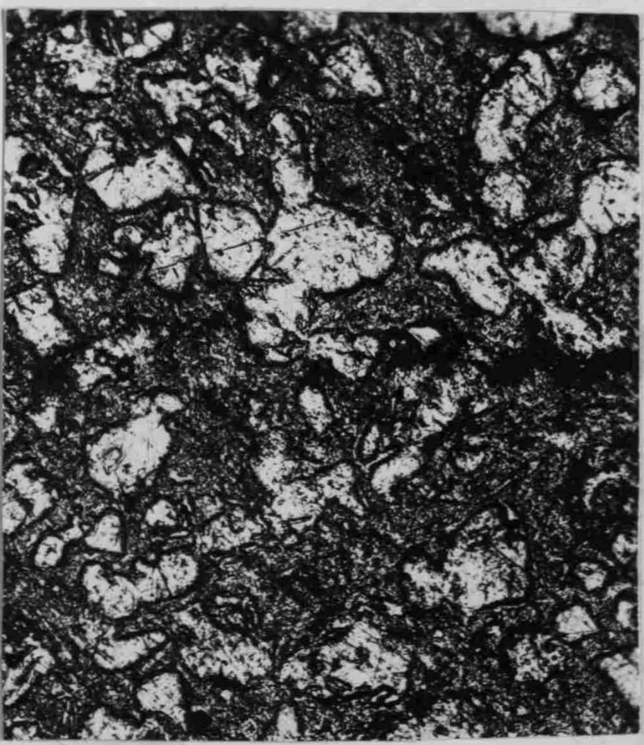
Original - Uranium alloy
Magnified 100X

Replica - Uranium alloy
Almost identical magnification and area

Here the effect of Indium-oxide is evident in the slightly greater detail visible on the replica than is on the original. All grains, however, visible on the original can be identified on the replica. The fact that the replica is a mirror image makes the comparison somewhat tedious.

The peculiarly shaped grain, a little above the center of this field, was an aid to its identification and location; as were the small holes scratched in the original, showing on its upper left and lower right-hand corners.

ILLUSTRATION IV

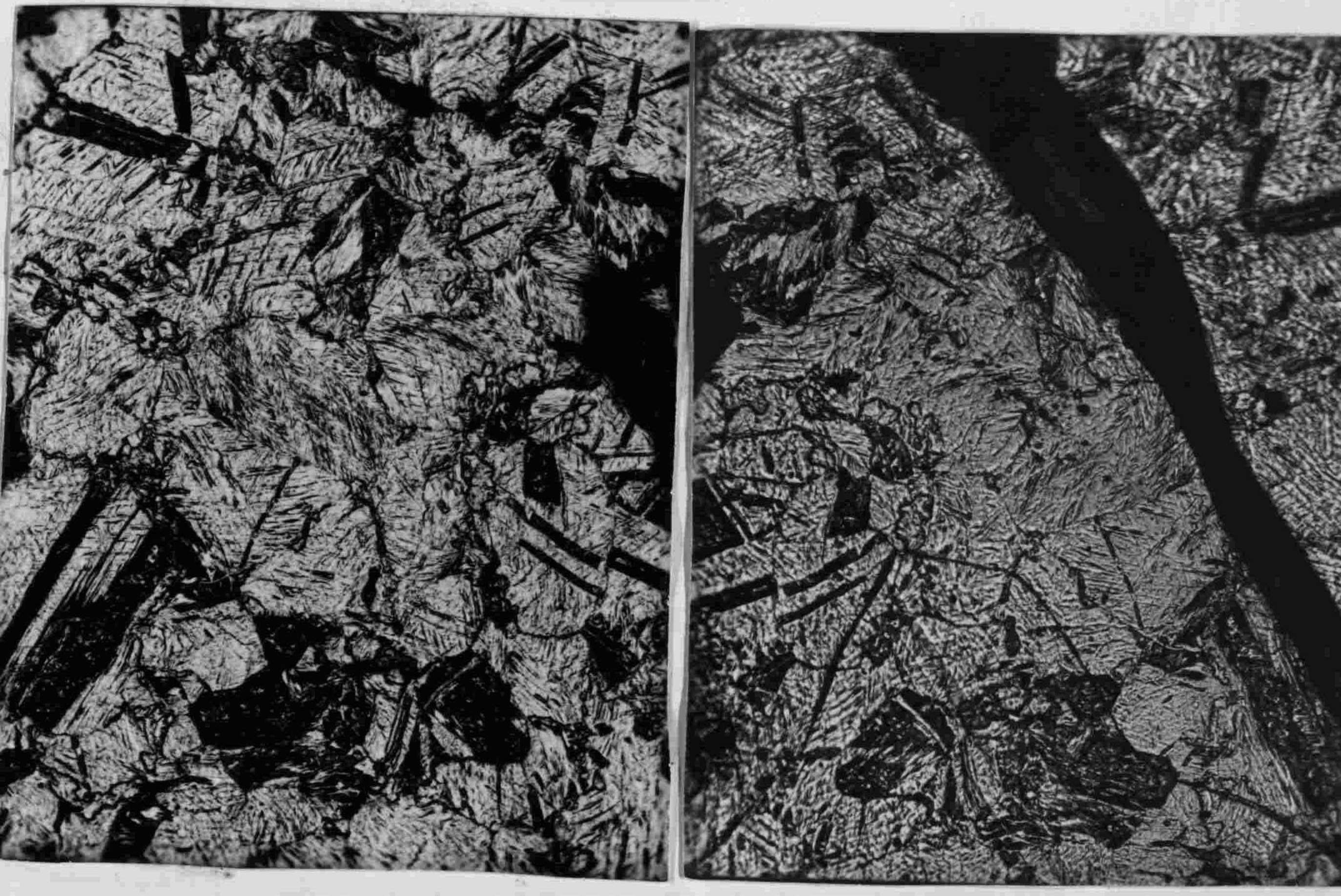
X

Original - Uranium alloy
Magnified 250X.

Replica - Uranium alloy
almost identical magnification
and area.

These are near the center of the same fields as are shown on the 100X magnification of Enclosure I. The Indium-oxide here seems to have the effect of causing a greater difference between the two photomicrographs.

ILLUSTRATION V



X

Original - Beryllium-Copper
Magnified 250X

Replica - Beryllium-Copper
Almost identical magnification and area.

This replica looked better under the microscope than any taken up to that time and looked accurate up to 550 diameter magnification. The scratches, however, illustrate one of the weaknesses of any such replica technique. The scratches were caused from handling before the replica could be photographed. The heavy scratch across the upper right-hand corner has been enough to warp most of the field so that not only was the area under the scratch affected, but the whole upper right-hand corner was pulled badly out of focus. There is another long thin scratch that cuts a sector out of the lower quarter of the field. To evaluate this replica, one must therefore compare a few grains near the center of the field which were relatively undistorted. Some remarkable details were here preserved despite the magnification.

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