cy. F.D.Shearin/5-16-66/va

ALMD66051581200

UNITED STAT
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IN REPLY REFER TO:
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May 12, 1966

Dr. John F. Eichelberger
Monsanto Research Corporation
Mound Laboratory
P. O. Box 32
Miamisburg, Ohio 45342

Subject: PATENT REVIEW - PAPER

Dear Dr. Eichelberger:

Please be advised there is no objection from a patent viewpoint to declassification and publication of the following paper which was submitted for review by copy of your letter of May 6, 1966 to Mr. H. F. Carroll:

"Autoradiographic Studies of the Penetration of Tritium in Stainless Steel" by J. D. Braun and G. L. Downs

which you desire to present at the Twentieth Meeting of the Metallography Group at Denver, Colorado on May 18, 1966.

Very truly yours,

William B Hollton

for Roland A. Anderson
Assistant General Counsel
for Patents

cc: H. F. Carroll
T. W. Laughlin, DTIE
Attn: Philip Rosser
D. W. King

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MOUND LABORATORY MONSANTO
66-5-158 A

May 6, 1966

Dr. L. M. Redman
University of California
Los Alamos Scientific Laboratory
P. O. Box 1663
Los Alamos, New Mexico 87544

Dear Doctor Redman:

Enclosed is a copy of a letter to Mr. H. F. Carroll requesting declassification of a paper which Mound Laboratory wishes to present at "The Twentieth Meeting of the Metallography Group" which is to be held in Denver on May 18, 1966.

I would like for you and Dr. D. W. Sherwood, ALO, to review this paper for reasons which are made clear in the letter to Mr. Carroll. A copy of the paper is enclosed for your use. I have asked Dr. Sherwood to inform Mr. Carroll of your opinions.

Thank you for your assistance in this matter.

Very truly yours,

Original Signed
By J. F. Eichelberger

J. F. Eichelberger
Coordinating Organization Director

JFE:va
Enclosures (2)

bc: D. L. Scott

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LABORATORY MONSANTO
66-5-158 (F)

May 4, 1966

AIRMAIL

Mr. D. W. King, Patent Attorney
U. S. Atomic Energy Commission
Albuquerque Operations Office
Albuquerque, New Mexico 87115

Dear Mr. King:

Enclosed is a rough draft copy of a paper entitled "Autoradiographic Studies of the Penetration of Tritium in Stainless Steel" by J. D. Braun and G. L. Downs which we wish to present at "The Twentieth Meeting of the Metallography Group" which is to be held at Denver, Colorado, May 18, 1966.

Would you please advise by telephone by May 16, 1966, if we may present this paper. The abstract has already been cleared so we do not feel that any patentable information will be divulged in this final paper. This meeting will be attended by AEC contractor employes.

To expedite clearance of this information, copies are simultaneously being sent to R. A. Anderson and H. W. Carroll for declassification review. If I can be of further help, please contact me.

Very truly yours,

Original Signed
By J. F. Eichelberger

J. F. Eichelberger
Coordinating Organization Director

Document transmitted [redacted] with contains [redacted] RICTED DATA

JFE/hk
Enclosure

cc: Mr. H. F. Carroll
Mr. R. A. Anderson
Mr. W. B. Creamer

bc; Mr. G. A. Essig
Mr. F. D. Shearin
File

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MOUND LABORATORY-MONSANTO
LETTER FILE NO. 66-5-158 (A)

May 6, 1966

Dr. D. W. Sherwood
Albuquerque Operations Office
U. S. Atomic Energy Commission
P. O. Box 5400
Albuquerque, New Mexico 87115

Dear Doctor Sherwood:

Enclosed is a copy of a letter to Mr. H. F. Carroll requesting declassification of a paper which Mound Laboratory wishes to present at "The Twentieth Meeting of the Metallography Group" which is to be held in Denver on May 18, 1966. Your copy of the paper is also enclosed. Would you please give this paper a thorough review for reasons which are made apparent in the letter to Mr. Carroll.

I am sending a copy of the paper to Dr. L. M. Redman, LASL. Perhaps you can discuss the paper with him or Dr. Krohn. I would appreciate your telephoning your comments to Mr. Carroll.

Very truly yours,

Original Signed
By J. F. Eichelberger

J. F. Eichelberger
Coordinating Organization Director

JFE:va
Enclosures (2)

bc: D. L. Scott

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Autoradiographic Studies of the Penetration of Tritium in Stainless Steel

Japnell D. Braun and Gene L. Downs

Introduction:

The use of autoradiography as a tool for measurement of the penetration of gases into metals is relatively recent. Although tritium has been used widely for autoradiography in biological systems, it has not been used extensively in metallurgical systems. Huber et al.[1] studied the distribution of hydrogen in titanium using tritium as the tracer for autoradiographs, and Cupp and Flubacher[2] reported their work of autoradiographic measurements of the diffusion of tritium in zirconium. Other related work reported in the literature is by Gomberg[3] on copper alloys and by Bruk and Nikolaev[4] on titanium and zirconium.

The penetration of tritium in stainless steel has been investigated by counting methods for measurement of the tritium concentration. Salmon and Randall,[5,6] who studied the diffusion of hydrogen and tritium in stainless steel and in palladium at elevated temperatures by radiocounting, reported activation energies and diffusion coefficients for tritium and hydrogen.

In this study we are attempting to measure tritium penetration by autoradiography and correlate this data with concentration-penetration data obtained by radiocounting methods. Measurement of the amount and location of tritium in a metal sample by autoradiography is a valuable addition to usual analytical methods and in some cases affords measurement of tritium concentration unobtainable by the

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usual radiocounting techniques which required the removal of samples by precision machining in increments of 0.002 - 0.005 inches. An autoradiograph, however, can show the variation in tritium concentration over incremental sample depths of 10 microns (0.0004 inches) or less and may be the only convenient method for obtaining ~~penetration~~ information (when penetration is less than 0.002 inches) ~~where very thin metal samples are involved~~.

Experimental:

All of the autoradiographic measurements were made on type 304 stainless steel. The samples were sectioned from the test ~~container~~ sample. Prior to mounting in plastic they were electroplated with nickel for edge preservation during polishing. They were then mounted in Bakelite, wet-ground through 600 grit SiC papers, rough polished with 1 micron Al_2O_3 on silk, and finally polished with Cr_2O_3 on microcloth. The polished samples were lightly electro-etched in a solution of 12% HCl in ethyl alcohol at 12 volts for approximately 5 seconds. For best results in autoradiography a light etch is preferred; furthermore, some etching is necessary as the photographic emulsion does not adhere well to a highly polished surface.

Eastman Kodak Company liquid nuclear tracking emulsions were used. These emulsions are solid at room temperature and below; they must be warmed to be applied to a sample and should be stored under refrigeration. Both the NTB-2 and NTB-3 emulsions were used successfully. The NTB-3 is a more sensitive emulsion; however, the background is usually higher than for type NTB-2. Lengths of

exposure varied from a few minutes to as long as 24 hours, depending on the concentration levels of tritium in the samples.

From the data furnished by Kodak on the range of various particles in the emulsion it was determined that a coating three microns thick would trap all of the beta particles from tritium decay. Coatings one micron thick would still contain all but a small percentage of the beta particles and would give improved resolution for photomicrographic measurements.

In discussing the techniques used a few remarks on emulsion coating techniques should be made. It is important to get an even coating, without passing over any part of the sample more than once. We finally adopted a method of dipping a small glass rod in emulsion and rolling it smoothly across the sample surface perpendicular to the direction of tritium penetration to avoid dragging any surface contamination from the higher concentration level to the lower one. The emulsion thickness, measured micrographically, was between 1 and 3 microns.

After the samples were coated with emulsion, they were placed in a light-tight box and exposed for varying periods of time. The samples were refrigerated while exposures were being made.

The emulsion was developed in place. Both coating and developing were carried out under limited, dark red lighting using a series 2 Wratten filter. Developing procedures were those recommended by Kodak: develop 2 minutes in Dektol(1:1), stop in distilled water,

fix 5 minutes in Kodak Fixer F-5, wash in distilled water 5 minutes, and dry slowly in a dust-free atmosphere.

The samples were then photographed at 100 and 1000X to show the location and intensity of exposed emulsion grains as related to the underlying grain structure of the sample.

Results:

Figure 1 shows photomicrographs at 100X of typical samples after 22 hours exposure using NTB-2 emulsion. Penetration in these samples varied between 15 and 20 mils, which agreed well with radiocounting data for these samples. Delete

To obtain quantitative measurements from the autoradiographs for the concentration of tritium, a method for determining the amount of exposure of the emulsion must be applied. The general procedure used by Cupp and Flubacher[2] in their work on tritium penetration in zirconium was modified for this study. Photomicrographs were taken at 1000X (using Polaroid P/N film) at various points across the sample. These points were chosen to correspond with given concentration levels in the sample determined from radiocounting measurements. The exposed emulsion grains were then counted on each photographic negative using a light box with a grid marked in 10 millimeter squares. For a 1000X negative this resulted in a count of exposed emulsion grains per 100 square microns. For the high intensity photographs exposed grains in at least 20 squares were counted. The remainder were counted over the entire area of the photomicrograph.

For each exposure time a standard of 304 stainless steel was run simultaneously and a background count obtained for that particular run. This was subtracted from the counts obtained for the samples. Background counts averaged about 2 counts per 100 square microns on the samples studied. Figures 2 and 3 show representative samples at the various concentration points. Table 1 shows the results of counts over the entire concentration range for two samples and two exposure times. Figure 4 shows the concentration of tritium expressed as exposed grains of emulsion per 100 square microns vs. distance on the samples. In these curves no correction has been applied for overlap of exposed emulsion spots; a much larger number of data points must be obtained before an accurate concentration-penetration curve can be determined by this method. ~~However,~~ From these preliminary quantitative measurements the method appears promising for the study of tritium penetration in metals at all concentrations of tritium.

References:

(1) O. J. Huber, J. E. Gates, A. P. Young, M. Pobereskin, and P. D. Frost, Trans. AIME, 209, 918 (1957).

(2) C. R. Cupp and P. Flubacher, J. Nucl. Mater., 6: 2, 213 (1962).

(3) H. J. Gomberg et al., "High Resolution Autoradiography for Study of Grain Boundaries in Metals" Final Report, Engineering Research Institute, University of Michigan (1954).

(4) B. J. Bruk and G. I. Nikolaev, Doklady Akad. Nauk. USSR, 116, 78 (1957).

(5) D. Randall and O. N. Salmon, Knolls Atomic Power Laboratory Report, KAPL-904 (1953).

(6) O. N. Salmon and D. Randall, Knolls Atomic Power Laboratory Report, KAPL-984 (1954).

TABLE 1. EXPOSED EMULSION GRAINS PER 100 SQUARE MICRONS FOR SAMPLES OF 304 STAINLESS STEEL CONTAINING TRITIUM

Tritium Concentration (ppm)	Exposed Grains/100 Square Microns			
	1 hour exposure		1/2 hour exposure	
	#196	#861	#196	#861
0.01	11.8	8.7	1.8	2.2
0.10	40.0	31.8	13.4	12.9
1.0	79.1	71.2	35.6	37.4
10.0	108.3	113.3	80.1	79.7
25.0	too dense	too dense	too dense	too dense

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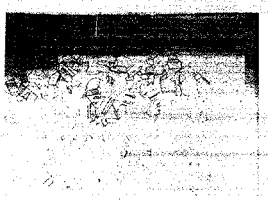
cy. 6 - R. A. Anderson, AECW
cy. 7 - D. W. Sherwood, ALO
cy. 8 - L. M. Redman, LASL
cy. 9 - J. F. Eichelberger

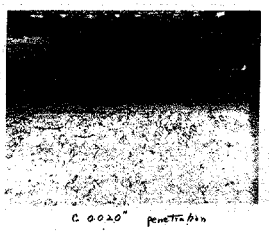
Figure 1.

Autoradiographs Showing Tritium Penetration in Type 304 Stainless Steel. Magnification 100X. Exposure 22 hrs.

a. 0.015" penetration



b. 0.015-0.018" penetration



c. 0.020" penetration

Figure 2.

Variations in Tritium Concentration 304 Stainless Steel. Autoradiographs 100X. Exposure, ½ hr

50 ppm

10 ppm

1 ppm

0.1 ppm

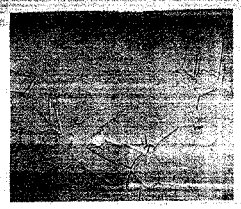
0.01 ppm

0 ppm

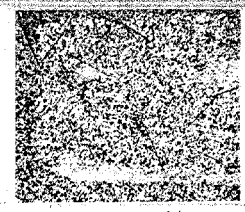
Standard, 0 ppm.

Figure 3.

AUTORADIOGRAPHS SHOWING TRITION CONCENTRATION IN TYPE 304 STAINLESS STEEL 1000 X

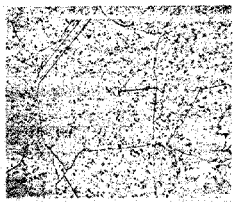


Standard, as polished and etched

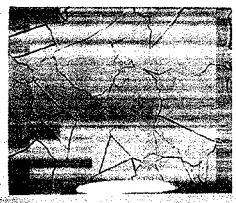


Sample 196, 10 ppm ½ hr exposure

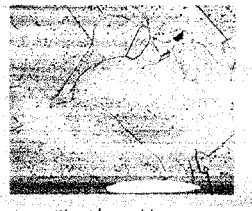
Standard, 0 ppm, ½ hr exposure



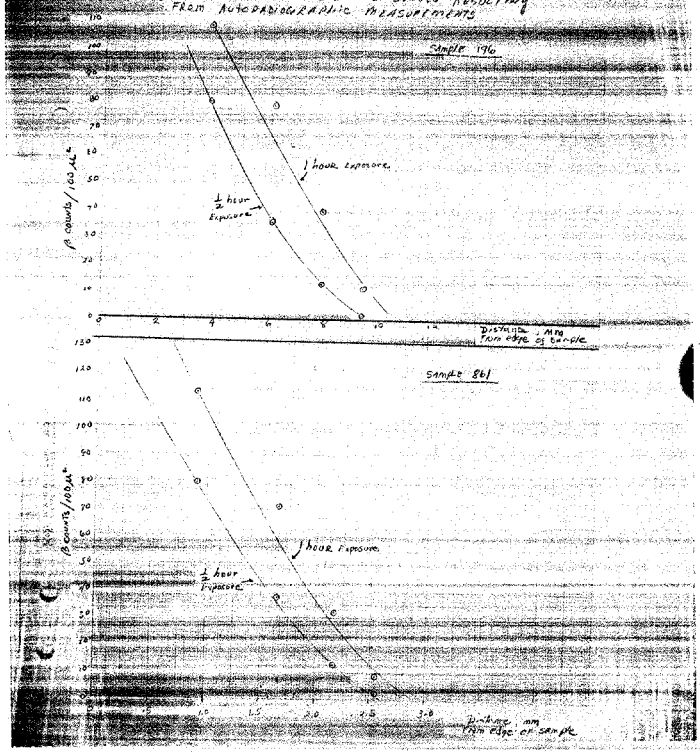
Sample 196, 1 ppm ½ hr exposure



Sample 196, as polished and etched



Sample 196, 0.1 ppm ½ hr exposure

Figure 4.

CONCENTRATION VS DISTANCE CURVES RESULTING FROM AUTORADIOGRAPHIC MEASUREMENTS