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METALLURGY AND CERAMICS

DIMENSIONAL INSTABILITY OF URANIUM–IV

Final Progress Report [for] July 1, 1958–March 31, 1959

By
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L. Seigle

February 1960

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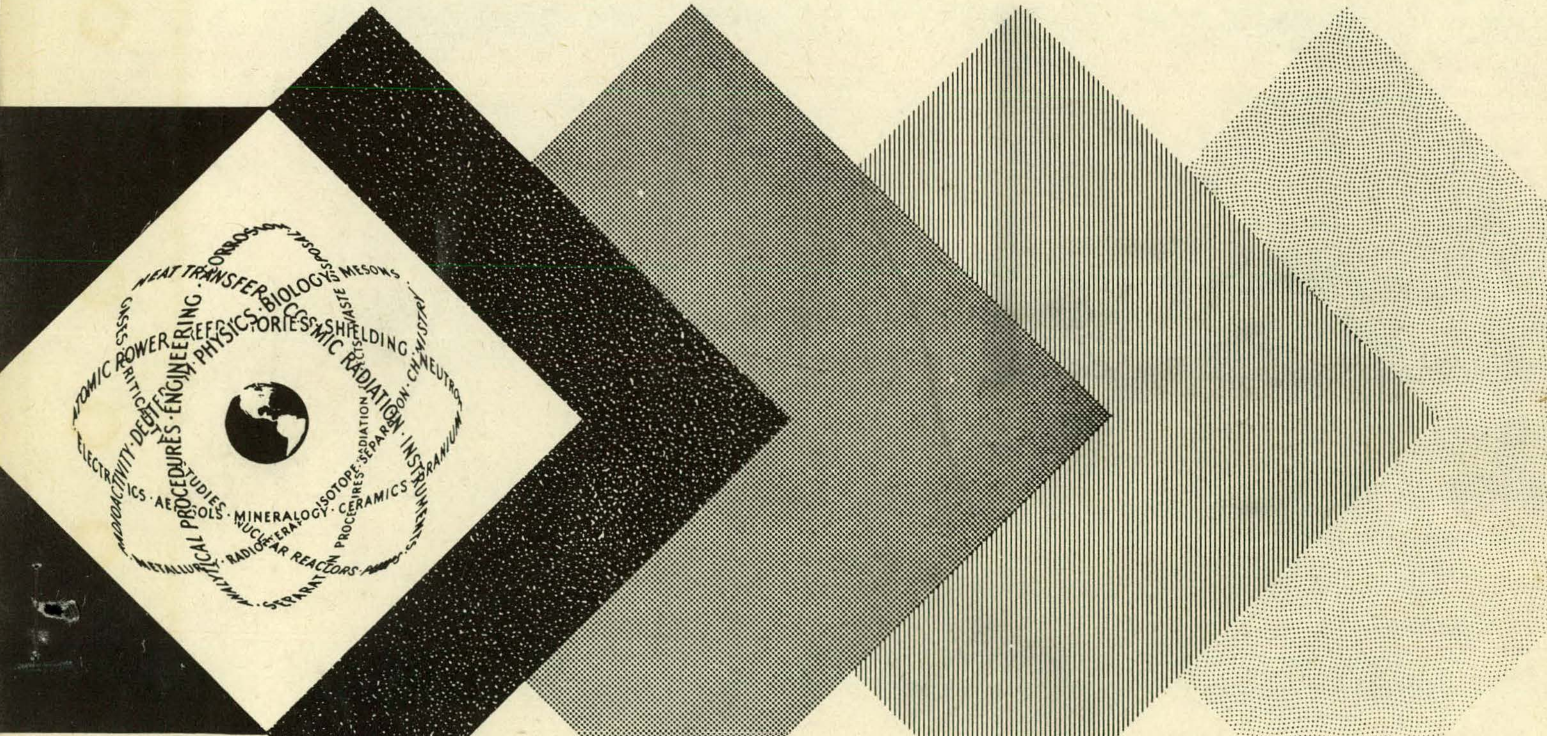

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TABLE OF CONTENTS

ABSTRACT

Several experiments were performed in order to provide experimental basis for the diffusional theory of the dimensional instability of uranium. Alpha-uranium rods possessing a constant preferred orientation but varying grain size were irradiated in the MTR. Contrary to a prediction based on the theory, no significant effect of grain size on irradiation-induced growth could be detected. Also, the self-diffusion coefficient of alpha-uranium was measured as a function of crystallographic direction, since the theory is based on the anisotropic diffusion of vacancies. The average of two measurements made in each direction at 640°C are:

$$D_{[100]} = 1.8 \times 10^{-14} \text{ cm}^2/\text{sec.}$$

$$D_{[010]} = 0.72 \times 10^{-14} \text{ cm}^2/\text{sec.}$$

$$D_{[001]} = 0.66 \times 10^{-14} \text{ cm}^2/\text{sec.}$$

The anisotropy is not nearly so large as had been anticipated, and the above differences are within the experimental error.

I. INTRODUCTION

It is generally agreed that the irreversible growth of alpha-uranium under neutron bombardment is somehow connected with its anisotropic properties and several theories have been advanced to explain the mechanism.

Anisotropy in thermal expansion and mechanical properties has served as the basis for theories advanced by Pugh, Bettman and others.(1) Seigle and Opinsky,(2) on the other hand, have proposed that anisotropic diffusion of displaced atoms and vacancies, produced in the lattice by fission fragments and fast neutrons, could be responsible for the observed effects. Their theory advanced the idea that the net diffusional flux of displaced atoms occurs preferentially in the [010] crystallographic direction, and emphasized the role that grain boundaries play as vacancy and interstitial sinks, thereby allowing growth in that direction. The present program was undertaken to provide an experimental test of the diffusional theory, and had as its objectives:

(a) To determine the effects of grain size upon the growth characteristics of irradiated alpha-uranium; and

(b) To determine the anisotropy in the self-diffusion coefficient of alpha-uranium.

II. EFFECT OF GRAIN SIZE ON THE GROWTH OF ALPHA-URANIUM DURING IRRADIATION

According to the diffusional theory, grain boundaries and free surfaces act as sinks for the vacancies and interstitial atoms. Since an excess of these defects is created during irradiation, it was predicted that the rate of uranium growth should be inversely related to the grain size. An experiment was designed to test the theory wherein a set of alpha-uranium rods were irradiated which were of constant preferred orientation and different grain size. The details and results of the first attempt of this experiment were reported in the First Annual Progress Report.(3) It was noted there that an effect of grain size on growth was indicated, but the magnitude of the effect was far less than had been anticipated on the basis of theoretical calculations which assumed that only grain boundaries may act as sinks.(2)

The specimens used in this initial experiment had been given only a cursory X-ray examination in order to be sure that the preferred orientation did not vary. Visual inspection of forward reflection Laue patterns indicated that the textures of the polycrystalline samples were similar, as expected, but in view of the small differences in growth encountered, it was felt that the experiment should be repeated using specimens in which a more exact determination of preferred orientation would be made. Complete pole figure studies are exceedingly difficult and time consuming. On the other hand, the coefficient of thermal expansion is relatively easy to measure and furnishes a reasonable criterion of similarity of preferred orientation in polycrystalline alpha-uranium.

A second set of specimens was prepared and the coefficients of thermal expansion measured after heat treatment. As reported in the Third Annual Progress Report,(4) the coefficients were quite uniform for specimens of differing grain size, indicating that the textures were alike. These specimens were shipped to the Knolls Atomic Power Laboratory where they were encapsulated. The specimens were later irradiated in position A-9 NE of the Materials Testing Reactor during MTR cycles 104 and 106. They received an integrated flux of 1.2×10^{20} n v t thermal neutrons as determined with the aid of a cobalt-aluminum flux monitor. The temperature was calculated to be 180°C at the specimen surface.

Irradiation-induced changes in specimen dimensions as measured at KAPL are given in Table I, and data from the first experiment are included for comparison. Photographs of the specimens before and after irradiation are shown in Figs. 1(a) to 1(e). Fig. 2 is a graphical representation of the results of these experiments and also includes data reported by Chiswik.(5) The results of the first experiment performed at this laboratory, and also Chiswik's results, had been interpreted as indicating a small but possibly significant grain size effect. The second experiment, however, shows a trend in the opposite direction, and suggests that it is not safe to conclude that any significant grain size effect exists at all. If any such effect exists, it is clearly quite small under the given experimental conditions.

Fig. 2 also presents the growth rates in the $[010]$ direction of "true" and "lineage" alpha-uranium single crystals. The value for the true single crystal of $G_{[010]} = 420$ is taken from the work of Paine and Kittel(6) and appears to be a reasonably well substantiated value. The absolute magnitude of the growth rate under irradiation of lineage single crystals is not known with certainty. Turkalo(7) concluded that lineage single crystals grow about 1.5 times as fast as true single crystals. Although a precise calculation was not made, Paine and Kittel state their results are in qualitative agreement with Turkalo's. Irradiations of the single crystals referred to above were conducted in the MTR under conditions similar to those for the polycrystals tested, and therefore the data are considered to be reasonably comparable.

It is significant that the lineage or imperfect single crystals grew much more rapidly under irradiation than the true or more perfect single crystals. Although the percent difference in growth rates could not be computed with accuracy, Paine and Kittel state that a large difference certainly exists. Hence, it appears that the presence of sub-boundaries influences growth rates although grain boundaries seemingly do not.

As already discussed by two of the authors,(8) one of these facts may explain the other. Individual crystallites of polycrystalline alpha-uranium become highly distorted during irradiation due to the mechanical interference of one growing grain with another. It is conceivable that the influence of sub-grain boundaries and possibly other crystalline imperfections produced during irradiation obscures that of true grain boundaries. Therefore the growth rate of a polycrystal is not a function of the nominal grain size but rather some effective sub-grain size which may vary during test. Further information regarding the influence of crystal imperfection on growth rates would be helpful and this might be best obtained with the aid of lineage single crystals rather than polycrystals of alpha-uranium, since the degree of imperfection could probably be more easily controlled and maintained in the single crystals. Also, lineage single crystals of alpha-uranium are produced without excessive difficulty. Of some significance too may be the fact that the polycrystalline specimens used in the second irradiation experiment grew considerably more than those used in the first experiment for the same total irradiation. Since the former specimens were fabricated from higher purity material, it is conceivable that purity may also be an important factor in irradiation growth.

III. MEASUREMENT OF THE ANISOTROPY OF SELF-DIFFUSION OF ALPHA-URANIUM

The measurement of the self-diffusion coefficients of alpha-uranium in each of its major crystallographic directions developed into an unusually difficult task. The experimental problems have already been discussed in detail in previous reports(3,4,9) and will be merely listed here:

(a) The preparation of large true single crystals of alpha-uranium.

(b) The development of a method for depositing adherent layers of the tracer isotope U^{233} onto the surface of the single crystals.

(c) The development of a method for measuring the very shallow penetration which must be expected from the limiting diffusion temperature of 640°C.

(d) The high toxicity of U^{233}, which required that all phases of the experiment involving the isotope be performed with unusual safety precautions.

A detailed summary of the experimental procedures finally adopted was given in the Third Annual Report(4) and is briefly reviewed below.

Single crystals of alpha-uranium prepared by the grain-coarsening method of Fisher(10) had flats ground on the surfaces perpendicular to one of the major crystallographic directions and were coated with a thin layer of a uranium alloy containing 5% U^{233} in natural uranium. The deposit was made by thermal evaporation in vacuum immediately after the ground surface had been cleaned by cathodic sputtering. The resulting diffusion couples were sealed in evacuated quartz capsules and annealed for times varying between six to ten weeks at 640°C.

Subsequent to the diffusion anneal, the ground faces of the diffusion couples were exposed to Eastman Kodak type NTA nuclear track emulsions which are sensitive only to highly ionizing radiations such as alpha emission. Two exposures were made on each diffusion couple; one with the specimen in direct contact with the emulsion, and the other with a thin (0.00035 inch) nickel foil interposed between the two. An identical pair of exposures were made using a bulk sample of the 5% U^{233} alloy originally deposited on the surfaces. After development of the photographic plates, the density of alpha particle tracks in the emulsion was measured using an optical grid set in the eyepiece of a Zeiss petrographic microscope fitted with a dark field condenser. All operations were carried out in a filtered drybox maintained under a negative pressure.

The outlined method is essentially a scheme for following the variation of surface concentration of the radiotracer with time and the corresponding solution to the diffusion equation gives

$$D = Q^2 / \pi C^2 t \qquad (1)$$

in which Q is a constant equal to the concentration of isotope in the initially deposited layer, times the layer thickness; C is the concentration of isotope at the surface at time t; and t is the diffusion time. The details of the procedure for computing the diffusion coefficients from nuclear track counts are given in the appendix.

Measurements of the diffusion coefficients in the three crystallographic directions were repeated twice using different annealing times. The results are presented in Table II. Contrary to expectations, no marked anisotropy of diffusion appears. These results are in agreement with the value $D = 2.3 \times 10^{-14}$ cm²/sec. for the diffusion of gold in dilute solution into polycrystalline alpha-uranium at 640°C obtained earlier with a standard sectioning technique.(3) Kidson,(11) at Chalk River, Canada, has made a number of measurements on very coarse-grained polycrystals at 647°C and reports D between 1.5 and 3.2×10^{-13} cm²/sec. Adda,(12) of the French Atomic Energy Commission, has also made measurements on polycrystalline material and reports a value of approximately 1×10^{-14} cm²/sec. at a temperature near the α-β transformation temperature.

This agreement suggests that the measured values are correct to at least an order of magnitude. Using the procedure described by Yagoda(13) the total counting error was computed, the results of which indicate that the reported D values are reliable to no better than ± 100%.

Other sources of error may be present in the experiment which are much more difficult to appraise. These are (a) the possible existence of substructure in the single crystals, and (b) the presence of a diffusion-inhibiting layer at the interface between the crystals and the deposited layer of isotope.

If the single crystals which served as the diffusion medium contained subboundaries, or if these had formed during the diffusion anneal, it is conceivable that diffusion could have taken place along these boundaries.

Secondly, if a very thin layer of oxide were formed at the interface between the base crystal and the evaporated layer of isotope, in spite of the extraordinary care taken during the preparation of the diffusion couples, then the diffusion coefficients measured might be that of uranium through a uranium oxide, instead of self-diffusion in metallic uranium.

In order to check the first possibility, a back reflection Laue X-ray photogram was taken on the [010] crystal after diffusion. This was compared with a photogram taken on the same crystal before fabrication of the diffusion couple for evidence of polygonization. The examination revealed that the spots were single and remained so after diffusion. Fig. 3(a) is the X-ray pattern taken before diffusion and 3(b) after diffusion. However, Jaumot and Smith(14) have found evidence of abnormal diffusion in zinc at low temperatures in single crystals which exhibited sharp Laue spots. Hence, the absence of gross substructure is no guarantee that imperfections have not influenced the diffusion rates. This is a very difficult issue to resolve.

Two approaches were taken to check the second possible experimental defect, i.e., the formation of a diffusion-inhibiting oxide layer. One consisted of fabricating diffusion couples from polycrystalline uranium with a natural uranium deposit using the identical technique to that for the U-U^{233} couples. These natural uranium couples could eventually be taken outside the dryboxes for metallographic examination. Unfortunately, the process of metallographic mounting and polishing marred the original surface to such an extent that the interface could not be observed. The second approach involved measuring the self-diffusion of gamma-uranium by the described technique and comparing the measured diffusion coefficients with those obtained by other investigators using standard sectioning techniques. Several measurements in the gamma phase agreed reasonably well with those of other investigations(15,16,17) as shown in Fig. 4. It may therefore be concluded that a serious diffusion-inhibiting barrier does not exist, at least not one which is effective at higher temperatures.

Although the precision of the measurements is probably not high, a marked diffusional anisotropy does not appear to exist. Variations larger than the 5% - 10% found by other investigators(14,18,19) in cadmium and zinc could easily be present, but order of magnitude differences are not indicated. It appears that the diffusion rate in the [100] direction is somewhat higher than the other two, but this difference is barely large enough to be outside of the possible limits of error of the method.

IV. DISCUSSION

The experiments carried out in this project do not furnish any experimental support for the diffusional theory of growth of alpha-uranium under irradiation. Within the accuracy of measurement, there exists no effect of grain size on growth rate and no marked anisotropy of diffusion. Growth under irradiation and perhaps also diffusion in alpha-uranium must be regarded as relatively unexplained processes, although several hypotheses exist or could be put forward.

The data on hand clearly establish that processes occurring within the crystals are more important than those occurring at crystal boundaries. Whether these are diffusional or mechanical processes cannot be stated with certainty at the moment. The various possibilities which have been put forward are discussed by the authors in another publication(8) and it is concluded that the meager experimental data available do not yet permit any clear distinction to be made.

The demonstrated effect of substructure on the growth of alpha-uranium single crystals is of outstanding interest, and further study of this subject might throw light on the underlying growth mechanism. The authors have suggested(8) that sub-grain boundaries might be playing the role of vacancy and interstitialcy sinks postulated for grain boundaries in the original diffusional theory.(2) If this were true, however, one might expect to see a greater difference between the growth rates of "true" single crystals, with few or no sub-boundaries, and "lineage" single crystals with very many sub-boundaries. True single crystals actually grow more rapidly than they theoretically should if only the surfaces acted as effective vacancy and interstitialcy sinks. This might be interpreted as an indication that point defect absorption at isolated dislocations plays a role in the growth process. Rough calculations(8) have shown that for normal dislocation densities and grain sizes, as many defects would be eliminated at dislocation lines as at grain boundaries. Exactly how the process of elimination at dislocations might lead to anisotropic growth (numbers of possibilities have been discussed) is, however, presently a matter of pure speculation.

Unfortunately the single crystal diffusion data, while of interest in themselves, do not help to clarify the growth problem. Although a high degree of diffusional anisotropy does not appear to exist, it is still quite possible from the data obtained that D for the [010] direction is, for example, one-half that for the other directions. This is already sufficient anisotropy of diffusion from the theoretical viewpoint. More accurate D values are necessary, therefore, to test the theory, but unfortunately these are not so easily obtained.

V. SUMMARY AND CONCLUSIONS

In the work carried out over the past several years, a theory of the growth of alpha-uranium under irradiation has been put forward, based upon the idea that growth occurs by the anisotropic diffusion of generated vacancies and interstitial atoms to grain boundaries and free surfaces. In order to test this theory, an experimental study has been carried out of the effect of grain size on the growth rate of polycrystalline alpha-uranium under irradiation, and of the rate of self-diffusion as a function of crystallographic direction. It has been demonstrated that grain size has no appreciable effect on the irradiation growth of polycrystals. Furthermore, a high degree of diffusional anisotropy apparently does not exist in alpha-uranium. It must be concluded, therefore, that the diffusional hypothesis is not valid as it was originally proposed.

The available data do not permit a clarification of the growth mechanism of alpha-uranium. This may still involve either diffusional or mechanical processes. If a diffusion mechanism operates, however, it must differ in detail from that originally visualized. Various modifications have been discussed but more experimental data are needed for further understanding of this problem.

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TABLE I

RESULTS OF IRRADIATION EXPERIMENTS

	Specimen	Average Grain Size	% Increase in Length	% Decrease in Diameter
First Experiment	A	40 microns	12.5	6.5
	B	30 "	13.9	6.8
	C	20 "	13.6	7.3
	D	20 "	14.3	7.2
Second Experiment	A	20 "	24.4	10.8
	B	60-150 "	28.3	11.3
	C	20 "	26.2	11.1
	D*	60-150 "	22.6	1.7

*Measurements on this specimen are approximate, due to distortion during irradiation.

TABLE II

SELF-DIFFUSION COEFFICIENTS

Crystallographic Direction	Annealing Time at 640°C	D, cm^2/sec.
[100]	46 days	1.9×10^{-14}
	48 days	1.7×10^{-14}
[010]	74 days	0.80×10^{-14}
	41 days	0.64×10^{-14}
[001]	44 days	0.96×10^{-14}
	55 days	0.35×10^{-14}

TABLE III

CORRECTED SELF-DIFFUSION COEFFICIENTS

Specimen	D, 1st Approx. cm^2/sec.	D, 2nd Approx. cm^2/sec.	D, 3rd Approx. cm^2/sec.
[100] , 46 days	2.9×10^{-14}	2.2×10^{-14}	1.9×10^{-14}
[100] , 48 days	2.7×10^{-14}	2.0×10^{-14}	1.7×10^{-14}
[010] , 74 days	1.5×10^{-14}	0.97×10^{-14}	0.80×10^{-14}
[010] , 41 days	1.5×10^{-14}	0.80×10^{-14}	0.64×10^{-14}
[001] , 44 days	2.0×10^{-14}	1.2×10^{-14}	0.96×10^{-14}
|001| , 55 days	0.91×10^{-14}	0.43×10^{-14}	0.35×10^{-14}

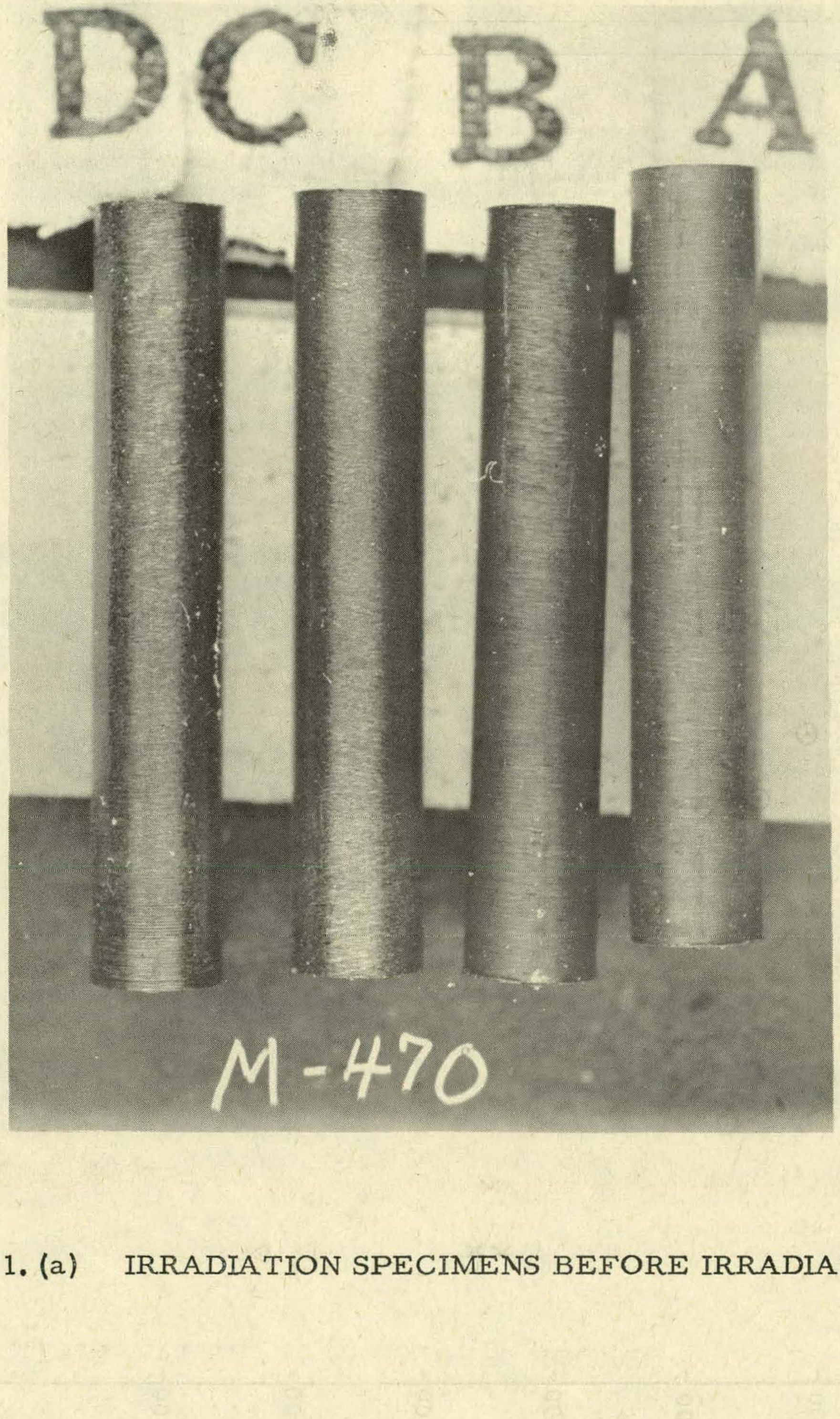
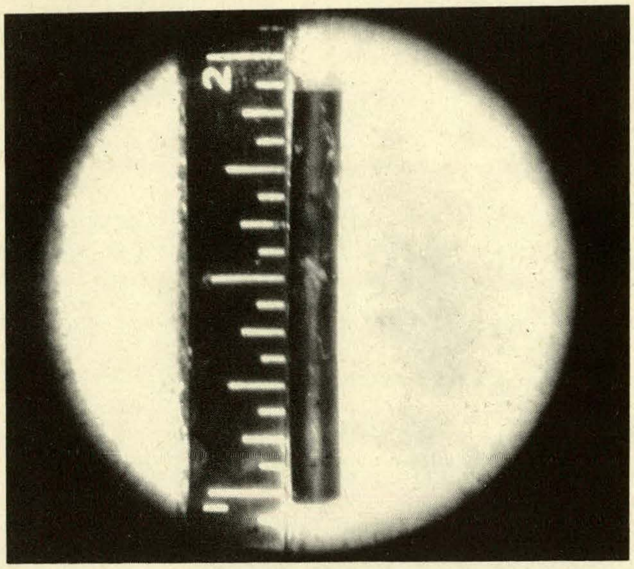
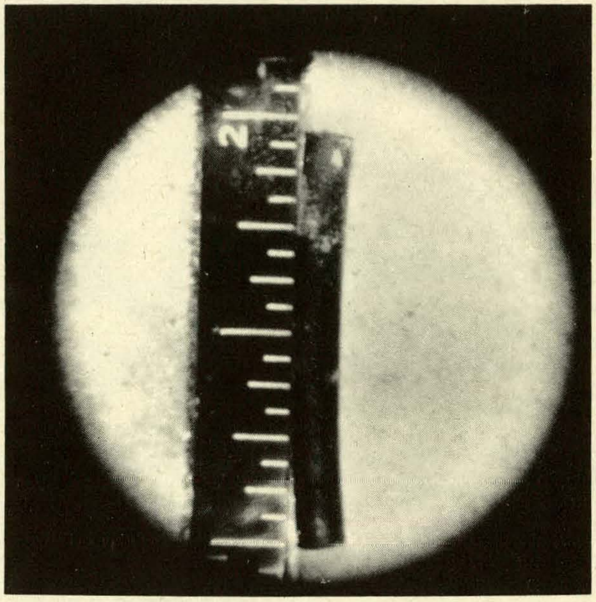


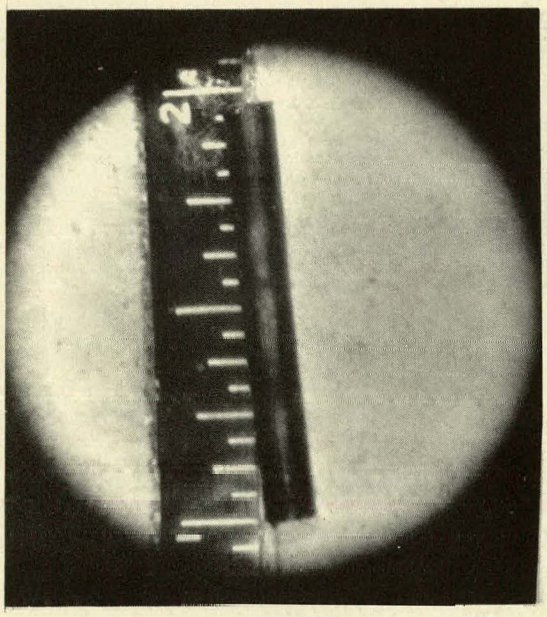
FIG. 1. (a) IRRADIATION SPECIMENS BEFORE IRRADIATION



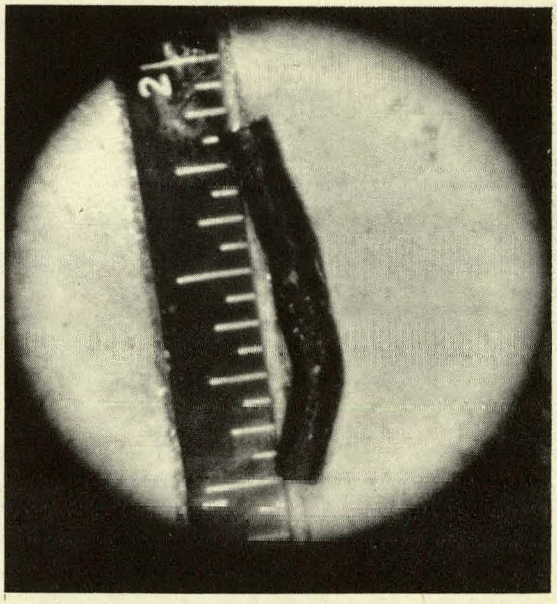
1 (b) Specimen A



1 (c) Specimen B



1 (d) Specimen C



1 (e) Specimen D

IRRADIATION SPECIMENS AFTER IRRADIATION

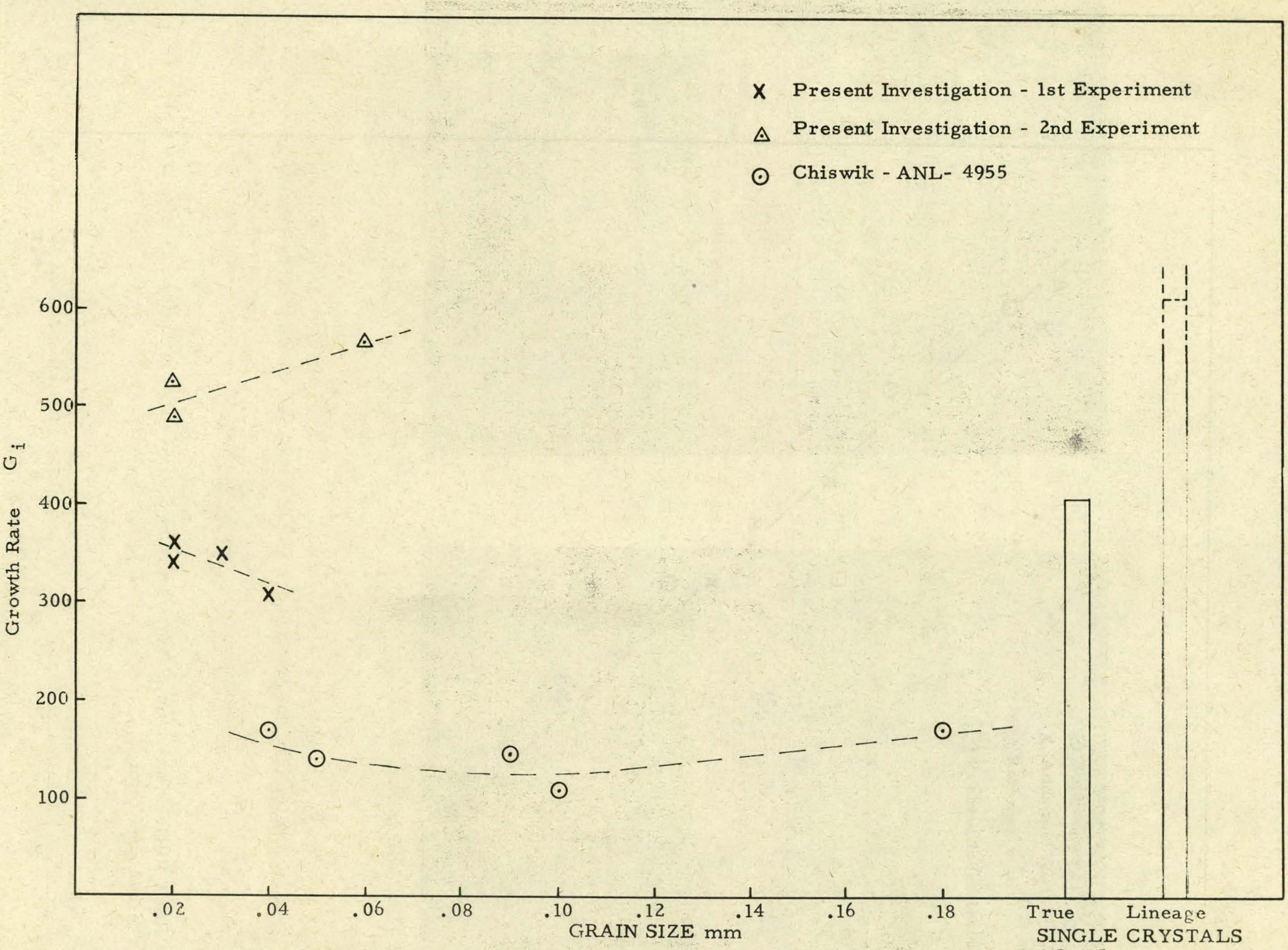


FIG. 2 - EFFECT OF GRAIN SIZE ON GROWTH RATE OF IRRADIATED ALPHA URANIUM

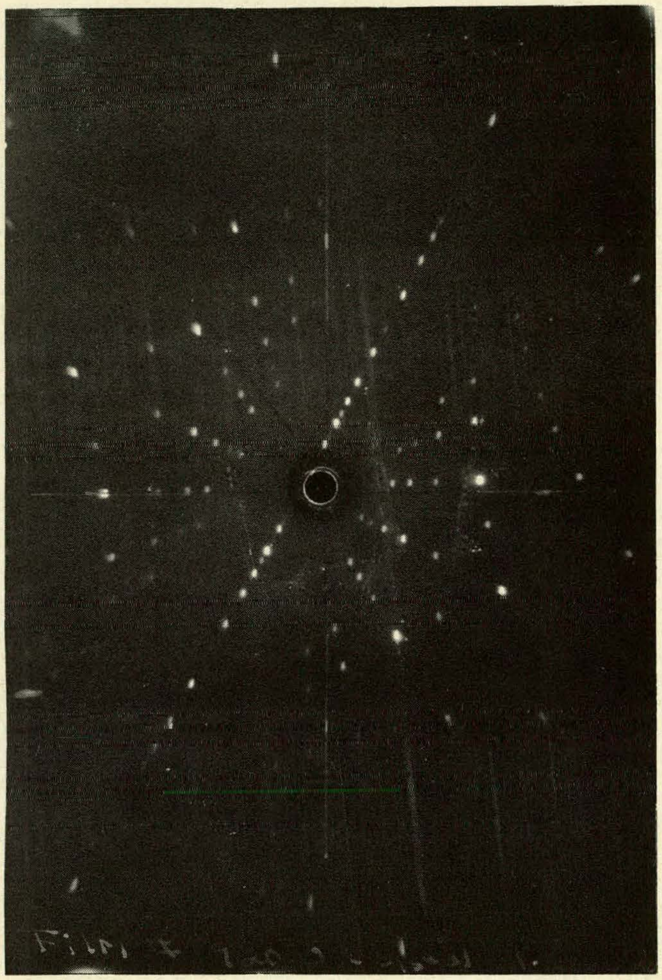


Fig. 3. (a)
Before Diffusion

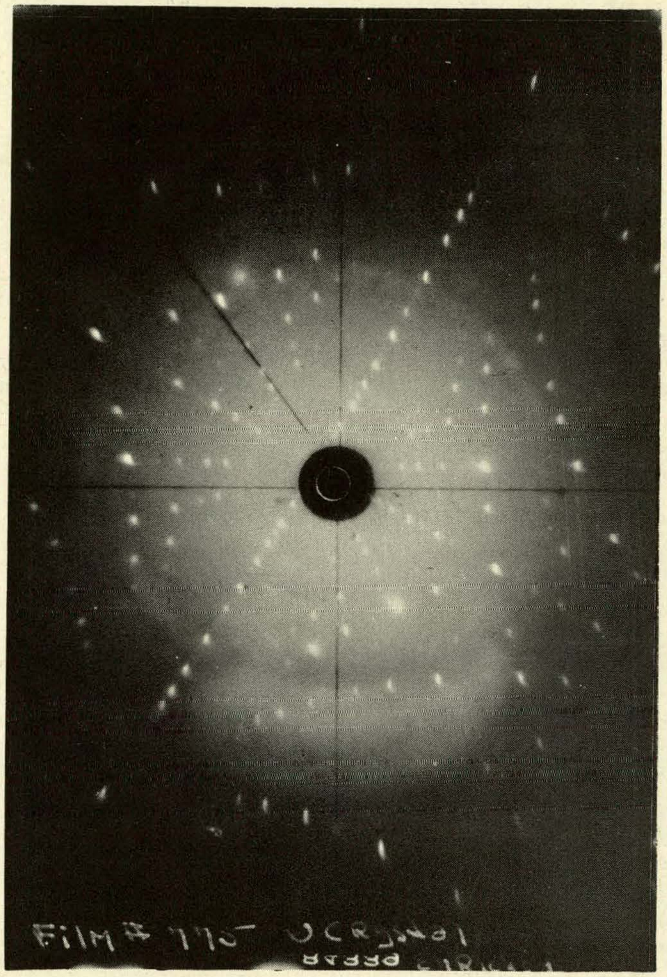


Fig. 3. (b)
After Diffusion

Back Reflection Laue Patterns taken on the [010] oriented single crystal Diffusion Couple.

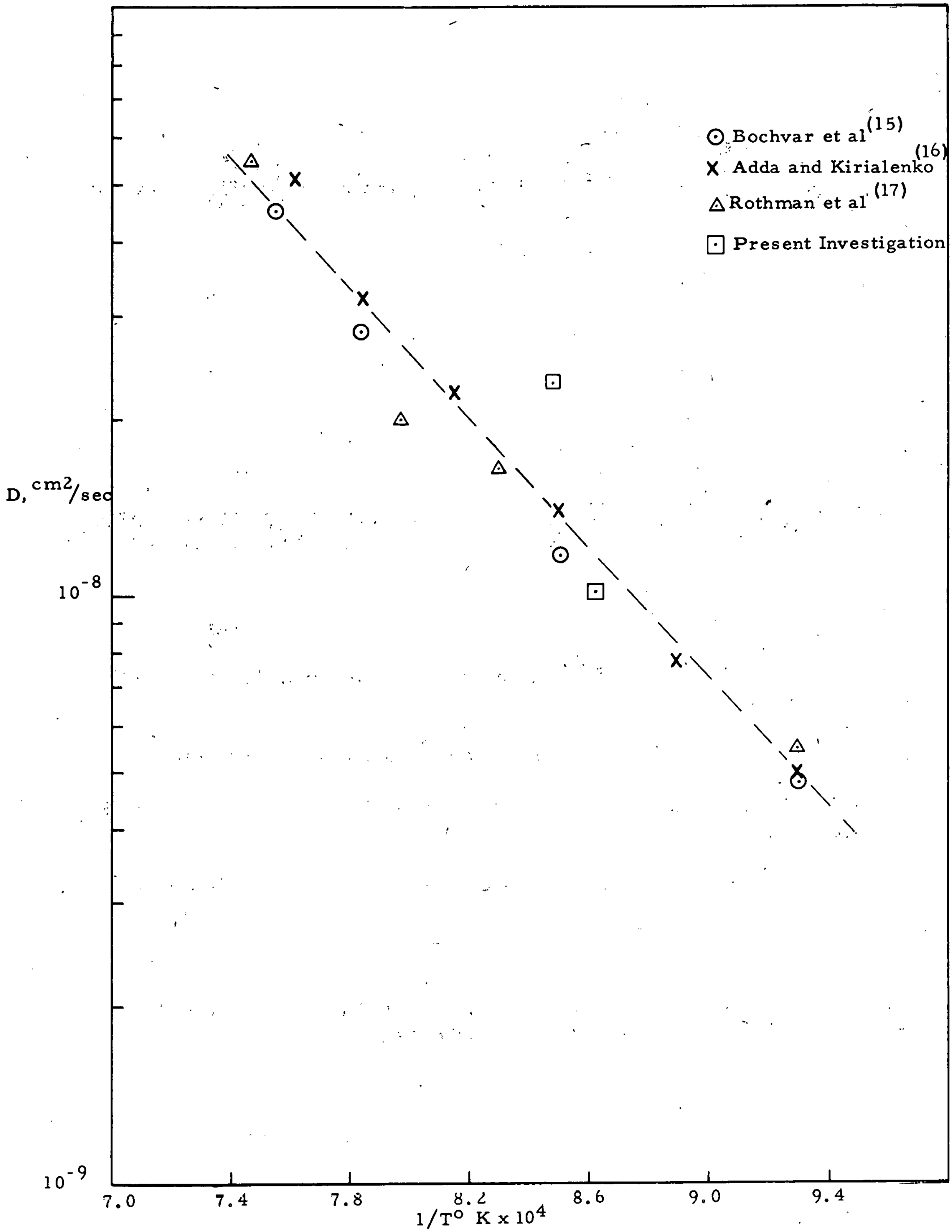
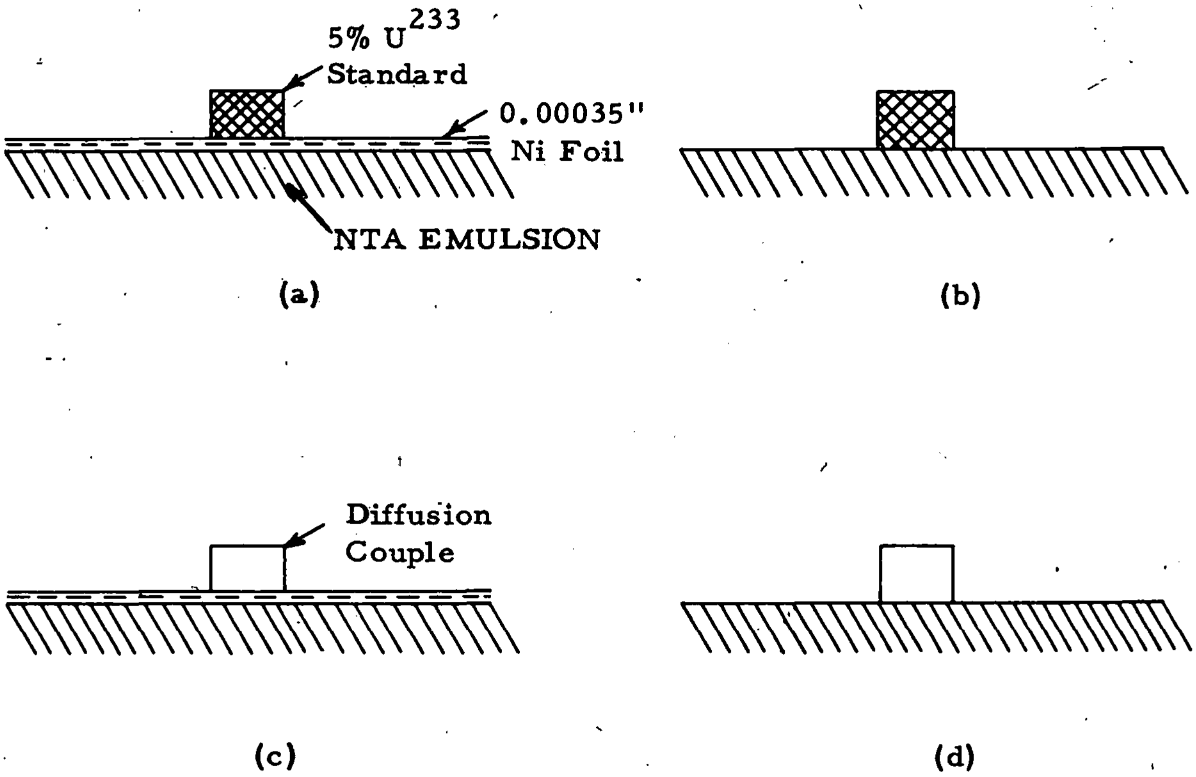


FIG. 4. SUMMARY OF SELF-DIFFUSION MEASUREMENTS ON GAMMA URANIUM.



(a) A standard alloy of 5% U^{233} is exposed to the emulsion with the foil interposed.

(b) The same standard is exposed to the emulsion without the foil.

(c) The diffusion couple after diffusion anneal is exposed with the foil interposed.

(d) The diffusion couple after diffusion anneal is exposed without the foil.

FIG. 5. SCHEMATIC REPRESENTATION OF NUCLEAR TRACK EMULSION EXPOSURES

APPENDIX

Computation of the Self-Diffusion Coefficients

The solution to the diffusion equation for conditions set up in this experiment is

$$D = \frac{Q^2}{\pi t C^2} \qquad (1)$$

where D is the self-diffusion coefficient, Q is the concentration times the thickness of the initial deposit, and C is the surface concentration at time t .

In order to calculate Q and C, four nuclear track exposures were made as illustrated in Fig. 5. Exposures 5(b) and 5(d) were used to evaluate Q as follows: It is assumed to begin with that the average percentage of U^{233} in the diffusion zone is equal to

$$5 \times \frac{C_{sb}}{C_{ub}}$$

where C_{sb} is the track density measured on exposure 5(d), and C_{ub} is the track density measured on exposure 5(b). This implies that the U^{233} in the diffusion couple is uniformly distributed over the diffusion zone. Since this is obviously not the case, the value of Q obtained is clearly an approximation. The approximation, however, does permit the calculation of a diffusion coefficient which can then be used to obtain a more accurate Q and in turn a more accurate D as explained below.

The range, R_s, of alpha particles in uranium is calculated from the Bragg-Kleeman relation,(13)

$$R_s = 3.19 \times 10^{-4} \; R_a \; \frac{\psi_s}{d_s} = 8.56 \times 10^{-4} \text{ cm} \qquad (2)$$

R_a in this equation is the range of U^{233} alpha radiation in air, ψ_s is the square root of the atomic weight and d_s the density of alpha uranium. The estimated percentage of U^{233} in the diffusion zone is used to determine the thickness of the initial deposit,

$$\tau = R_s \frac{C_{sb}}{C_{ub}}$$

assuming all of the U^{233} lies within R_s from the surface. Q is then equal to:

$$\frac{C_{sb} \times R_s \times 0.95}{C_{ub}} \quad \text{grams/cm}^2$$

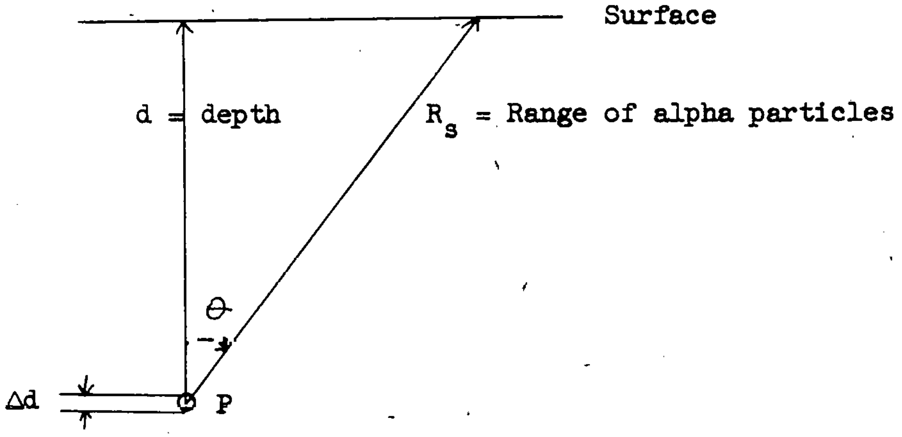
The surface concentration after diffusion is evaluated from exposures 5(a) and 5(c). The presence of a nickel foil between the photographic emulsion and the specimen serves as a barrier to the alpha radiation. Only those particles emitted from a narrow zone at the specimen surface possess sufficient energy after leaving the uranium to penetrate the foil and enter the emulsion. The depth of this surface zone was calculated to be 1.32 x 10^{-4} cm by the use of exposures 5(a) and 5(b), by a method similar to that which follows. It is required that this depth be shallow relative to the depth of diffusion so that the concentration measured is representative of the surface concentration.

The surface concentration $C = \dfrac{0.95\ C_{sf}}{C_{uf}}$ grams/cm^3

where C is the surface concentration, C_{sf} is the track density measured on exposure 5(c), and C_{uf} is that measured on exposure 5(a). Substituting the quantities for Q and C in equation (1), we get:

$$D = \frac{[R_s]^2\ [C_{sb}]^2\ [C_{uf}]^2}{\pi t\ [C_{sf}]^2 [C_{ub}]^2} \qquad (3)$$

The value of D computed from equation (3) is inaccurate due to the error in Q caused by the assumption of a uniform distribution of isotope in the diffusion zone as described above. This may be used to obtain a more accurate D, as follows:



The fraction of the total number of alpha particles emitted by the particle of uranium, P, at depth, d, which reach the surface is

$$F = \frac{1 - \cos\theta}{2} = \frac{1}{2}\left(1 - \frac{d}{R_s}\right)$$

The number of alphas reaching the surface per unit time from the layer Δd in thickness at depth d equals $\Delta C = \alpha F \gamma \Delta d$ per unit area. γ = number of emitting centers per cm^3, and α = rate of alpha emission of U^{233}.

$$\Delta C = \tfrac{1}{2} \alpha \gamma \left(1 - \frac{d}{R_s}\right) \Delta d$$

$$C = \int_0^{R_s} \tfrac{1}{2} \alpha \gamma \left(1 - \frac{d}{R_s}\right) \delta d$$

$$\frac{C_{ub}}{C_{sb}} = \frac{\tfrac{1}{2}\, \alpha/R_s\, \gamma_u \int_0^{R_s} (R_s - d)\, \delta d}{\tfrac{1}{2} \cdot \alpha/R_s \int_0^{R_s} \gamma_s (R_s - d)\, \delta d}$$

where $$\gamma_s = \frac{Q}{\sqrt{\pi D t}}\, e^{-d^2/4Dt}$$

is the number of emitting centers/cm^3 in the diffusion couple, and

where γ_u = constant, is the number of emitting centers/cm^3 in the standard alloy.

Thus,

$$\frac{C_{ub}}{C_{sb}} = \frac{\gamma_u \int_0^{R_s} (R_s - d)\, \delta d}{\frac{Q}{\sqrt{\pi D t}} \int_0^{R_s} e^{-d^2/4Dt} (R_s - d)\, \delta d}$$

$$\frac{C_{ub}}{C_{sb}} = \frac{\tfrac{1}{2} R_s^2 \gamma_u}{\frac{Q}{\sqrt{\pi D t}} \left[2Dt\left(e^{-R_s^2/4Dt} - 1\right) + R_s \int_0^{R_s} e^{-d^2/4Dt}\, \delta d\right]}$$

$$Q = \frac{C_{SB} R_s^2 \gamma_u}{C_{uB} R_s \left[\frac{2}{R_s} \sqrt{Dt/\pi} \left(e^{-R_s^2/4Dt} - 1\right) + \operatorname{erf} R_s/\sqrt{4Dt}\right]} \qquad 4)$$

Substituting the value of D obtained from equation (3) into equation (4), and solving for Q, we can make a series of successive approximations which reduce the error in Q and consequently the error in D. Table III lists the values of D obtained from each of three successive approximations.

The counting errors in C_{ub} and C_{sb} were calculated using the method described by Yagoda(13) to be 10%, and the error in C_{uf} and C_{sf} 5%. Since R_s is obtained from the empirical Bragg-Kleeman relation, equation (2), the error in this value is not known. Assuming it is no more than 10%, the error in the relative value of the self-diffusion coefficients is 80% from these sources alone.