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Report to
Savannah River Operations Office
United States Atomic Energy Commission

A Metallographic Study of Gas Behavior in Uranium Irradiated with Alpha Particles

H. M. Green

September 28, 1962

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TABLE OF CONTENTS

	<u>Page No.</u>
ABSTRACT	10
I. INTRODUCTION	11
II. CONCLUSIONS	12
III. EXPERIMENTAL METHODS	13
A. Materials Used	13
B. Cyclotron Irradiation	13
C. Post-Irradiation Annealing	14
D. Metallographic Examination	15
IV. OBSERVATIONS	15
A. Ingot Uranium	15
B. Dingo Uranium	17
C. High Purity As-Cast Uranium	18
D. Uranium-Molybdenum Alloys	18
E. U - 0.3 ^W /o Cr - 0.3 ^W /o Mo	18
F. U - 2 ^W /o Zr	19
G. U - 3.9 ^W /o Si	20
H. Thorium	20
V. DISCUSSION	20
A. Unalloyed Uranium	20
B. Alloys	22
C. Comparison with Swelling in Neutron-Irradiated Uranium	23

TABLE OF CONTENTS (Continued)

	<u>Page No.</u>
VI. TABLES AND FIGURES	25
VII. REFERENCES	56
APPENDIX A	A-1
APPENDIX B	B-1

LIST OF TABLES AND FIGURES

	<u>Page No.</u>
TABLE I - Materials Used in Cyclotron Irradiation Experiments	25
TABLE II - Summary of Post-Irradiation Annealing Treatments	26
TABLE III - Summary of Interrupted Irradiation and Annealing Experiments	27
TABLE IV - Summary of Electron Micrograph Data on Class I Bubbles as a Function of Alloy Content	28
TABLE V - Occurrence of Class II Bubbles as a Function of Alloy Content and History	29
FIGURE 1 - Photomicrograph of cyclotron-irradiated ingot uranium as irradiated.	30
FIGURE 2 - Photomicrographs of bubble formation in cyclotron-irradiated ingot uranium after post-irradiation anneals for 90 hours at various temperatures.	31
FIGURE 3 - Electron micrographs of bubbles in cyclotron-irradiated ingot uranium after post-irradiation anneals for 90 hours at various temperatures.	32
FIGURE 4 - Photomicrographs of bubble formation in cyclotron-irradiated cold-worked ingot uranium after post-irradiation anneals for 90 hours at various temperatures.	33
FIGURE 5 - Photomicrographs of bubble formation in cyclotron-irradiated ingot uranium after various combinations of isothermal and thermal cycling post-irradiation anneals.	34
FIGURE 6 - Photomicrographs of bubble formation in ingot uranium after continuous cyclotron irradiation and post-irradiation annealing, compared with interrupted cyclotron irradiations and anneals.	35
FIGURE 7 - Photomicrographs of bubble formation in ingot uranium after various cyclotron irradiations, each exposure followed by 16-hour heat treatment at 900°C.	37
FIGURE 8 - Photomicrographs of bubble formation in ingot uranium after various cyclotron irradiations, each exposure followed by 10 cycles from 730°C to 600°C.	38

LIST OF TABLES AND FIGURES (Continued)

	<u>Page No.</u>
FIGURE 9 - Photomicrographs of bubble formation in cyclotron-irradiated arc melted ingot uranium after various post-irradiation anneals.	39
FIGURE 10 - Photomicrographs of bubble formation in cyclotron-irradiated dingot uranium after post-irradiation anneals at 900°C for various times.	40
FIGURE 11 - Photomicrographs of bubble formation in cyclotron-irradiated dingot uranium after post-irradiation anneals at 975°C for various times.	41
FIGURE 12 - Photomicrographs of bubble formation in cyclotron-irradiated dingot uranium after post-irradiation anneals at 1075°C for various times.	42
FIGURE 13 - Photomicrographs of bubble formation in cyclotron-irradiated dingot uranium after post-irradiation anneals for 90 hours at various temperatures.	43
FIGURE 14 - Photomicrographs of bubble formation in cyclotron-irradiated cold-worked dingot uranium after various post-irradiation anneals.	44
FIGURE 15 - Photomicrographs of bubble formation in cyclotron-irradiated dingot uranium after various combinations of isothermal and thermal cycling post-irradiation anneals.	45
FIGURE 16 - Photomicrographs of bubble formation in cyclotron-irradiated high-purity uranium after various combinations of isothermal and thermal cycling post-irradiation anneals.	46
FIGURE 17 - Photomicrographs of bubble formation in cyclotron-irradiated U - 1.6 W/o Mo alloy after various post-irradiation anneals.	47
FIGURE 18 - Photomicrographs showing phase transformation in cyclotron-irradiated U - 1.6 W/o Mo after a post-irradiation thermal cycling anneal (25 cycles, 715°C for 10 hours, 600°C for 1 hour).	48
FIGURE 19 - Photomicrographs of cyclotron-irradiated U - 2.75 W/o Mo after post-irradiation anneals for 90 hours at various temperatures.	49

LIST OF TABLES AND FIGURES (Continued)

	<u>Page No.</u>
FIGURE 20 - Photomicrographs of bubble formation in cyclotron-irradiated beta phase U - 0.3 W/o Cr - 0.3 W/o Mo after various post-irradiation anneals.	50
FIGURE 21 - Photomicrograph of bubble formation in cyclotron-irradiated beta phase U - 0.3 W/o Cr - 0.3 W/o Mo after a post-irradiation thermal cycling anneal (25 cycles, 715°C for 10 hours, 600°C for 1 hour, 4 hours at 900°C).	51
FIGURE 22 - Photomicrographs of bubble formation in cyclotron-irradiated U - 2 W/o Zr after various post-irradiation anneals.	52
FIGURE 23 - Electron micrographs of cyclotron-irradiated U - 2 W/o Zr after post-irradiation anneals for 90 hours at various temperatures.	53
FIGURE 24 - Photomicrographs of bubble formation in cyclotron-irradiated U - 3.9 W/o Si after various post-irradiation anneals.	54
FIGURE 25 - Photomicrographs of bubble formation in cyclotron-irradiated "reactor grade" thorium after various isothermal post-irradiation anneals.	55
FIGURE B-1 - Bubble diameter versus post-irradiation annealing temperature.	B-2

ABSTRACT

The behavior of helium in uranium and in several uranium alloys was studied metallographically after room temperature irradiation with alpha particles in a cyclotron, followed by various heat treatments. The observed gas bubbles were classified in two classes, differing in size, concentration and mechanism of formation. These bubbles appear to resemble those resulting from fission product gases in neutron-irradiated uranium; the findings of this study seem to be applicable to the understanding of the factors affecting bubble formation and thus to the suppression of swelling in neutron-irradiated materials.

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I. INTRODUCTION

The volume increase of uranium-base metal fuels irradiated at elevated temperatures (above 350°C) is an important factor limiting the operating temperature of nuclear reactors. A major cause of this swelling is believed to be bubbles, which result from the gaseous fission products, krypton and xenon. Little is known about the factors affecting the nucleation and growth of such bubbles. With proper appreciation of the factors governing the formation of bubbles the design of metallic fuels would be on a sounder basis.

Experiments with neutron-irradiated fuels have yet to provide a consistent theory of gas bubble formation. Such experiments are inherently expensive and do not readily lend themselves to control of the irradiation variables. Information regarding the behavior of inert gas atoms in metals can be obtained by introducing inert gas atoms into metallic specimens by bombardment in a cyclotron. Alpha particle irradiation has been successfully used to introduce helium into copper,^(1,2) beryllium⁽³⁾ and aluminum;^(4,5) the behavior of the helium was studied as a function of the conditions of post-irradiation annealing. The work reported here was undertaken to apply this technique to uranium-base material. It was expected that relatively simple experiments could be performed under controlled conditions, leading to identification of the factors affecting bubble formation and growth.

Specimens of various uranium compositions were subjected to cyclotron irradiations that provided a band of metal containing a gas atom concentration equivalent to that obtained in neutron-irradiated uranium at about 1.9 %^a burn-up of total atoms. In general, the samples received the complete cyclotron irradiation before being subjected to a variety of heat treatments. For some samples, the cyclotron irradiation was interrupted several times for heat treatment. These interrupted irradiation-anneals were intended to provide additional information regarding nucleation and growth of bubbles.

II. CONCLUSIONS

- (1) Post-irradiation anneals of uranium-base materials that had been cyclotron-irradiated with alpha particles resulted in gas bubble formations similar to that observed in uranium-base materials that were neutron-irradiated. The relationship between the behaviors of the cyclotron-irradiated material and the neutron-irradiated material has not been clearly defined.
- (2) Gas bubbles of two classes were found in uranium-base materials that were irradiated with alpha particles in a cyclotron and then annealed at temperature of 600 to 1075°C. Class I bubbles have diameters less than 1 micron and densities of the order of 10^{13} to 10^{15} per cc. Class II bubbles have diameters of 10 to 150 microns and densities of the order of 10^3 to 10^5 per cc. Bubbles of intermediate size were not seen.
- (3) The number and distribution of Class I bubbles appear to be related primarily to the availability of nucleation sites.
- (4) Activation energy for nucleation and growth of Class I bubbles in gamma phase uranium was determined to be 20 Kcal/mole at temperatures between 900 and 1075°C; this value is in good agreement with that reported for the self-diffusion of gamma uranium.
- (5) In the dilute alloys of uranium tested, Class I bubbles were in general smaller in size and greater in number than in unalloyed uranium subjected to the same annealing conditions.
- (6) In uranium metal richer in additions, Class II bubbles form less easily during gamma phase anneals.
- (7) Post-irradiation thermal cycling treatments did not, in general, alter the formation of bubbles in materials included in this study.

III. EXPERIMENTAL METHODS

A. Materials Used

The materials used in this investigation include unalloyed uranium representing different production methods and metallurgical conditions, uranium alloys and thorium. The materials are described in Table I. The specimens (1 inch by 0.25 inch) used in the cyclotron-irradiation experiments were machined or rolled to a thickness of about 40 mils. Prior to the cyclotron irradiation, some of the uranium and uranium alloy specimens were annealed for two hours at 850°C while other specimens were irradiated in either the as-cast or as-alpha worked conditions. The thorium specimens were prepared from two grades of material, identified as "reactor grade" and "high purity".

B. Cyclotron Irradiation

The specimens were irradiated with alpha particles in the MIT cyclotron. The specimen holder, a water-cooled aluminum block, has been described by Ellis and Evans.⁽⁶⁾ The specimens, clamped to the specimen holder, were probably below 50°C throughout the irradiation. The initial 31.5 Mev energy of the alpha particles was reduced to less than 30 Mev by causing them to pass through 2-1/2 mils of aluminum foil before striking the specimen.

The approximate helium concentrations in the irradiated samples are calculated from the integrated alpha beam current, the area of the alpha beam, and the thickness of the helium-rich layer. On the assumption of uniform alpha particle distribution, the gas concentration in terms of the gas volume ratio (GVR, the volume of gas measured at STP per unit volume of metal) is given by the equation:

$$\text{GVR} = \frac{I_t \text{ KV}}{V N_0} \quad \text{Eq (1)}$$

where I_t is the integrated alpha beam current in microampere-hours,

K is the number of doubly ionized alpha particles per microampere-hour (1.16×10^{16}),

V_0 is the volume of one mole of gas at STP (22.4 liters),

V is the volume of irradiated material in which the alpha particles are stopped, and

N_0 is Avogadro's number, 6.02×10^{23} .

The specimens were irradiated in the cyclotron for exposures of about 50 microampere-hours. The uncertainty in exposure is due to uncertainties involved in measuring the alpha beam current and to small fluctuations in the beam current during irradiation. The volume of material in which the alpha particles are stopped is also subject to error because (1) the beam is not uniform along any coordinate and (2) the thickness of the band containing the alpha particles cannot be observed before annealing. In metallographic studies after post-irradiation anneals, a band of helium gas bubbles about 0.3 mils thick is observed about 4 to 5 mils below the surface. The appearance of the band indicates that the alpha particle beam was uniform for about 0.1 inch of the 0.25-inch wide specimen. Assuming that the beam current is a measure of the alpha particles impinging along the full length of the specimen holder, 4.5 inches, the helium concentration for a 50 microampere-hour exposure is calculated from Equation 1 to be about 9 GVR. The concentration of krypton and xenon resulting from neutron irradiation of natural uranium is about 4.7 GVR per a/o burnup.⁽⁷⁾ Therefore, the helium concentration in the small volume of the sample in which the alpha particles are stopped is equivalent to the gas concentration obtained in neutron-irradiated uranium at about 1.9 a/o burnup of total atoms.

C. Post-Irradiation Annealing

Following the 50 microampere-hour irradiations the 1-inch long samples were sectioned transversely into specimens about 0.050-inch long which were then given various post-irradiation annealing treatments. The details of the heat treatments and their purposes are summarized in Tables II and III. The irradiated specimens were heat treated in Vycor or quartz capsules containing about one atmosphere of helium at the annealing temperature. Samples were wrapped in tantalum to prevent reaction with the container. After the heat treatment the capsules were cooled in air.

D. Metallographic Examination

Samples after each heat treatment were prepared for optical microscopy by mechanical polishing and appropriate etching. Unalloyed uranium was etched chemically in a solution of 1 part glacial acetic acid and 1 part nitric acid. The U - 2 ^W/o Zr samples were etched in a solution of 10 parts nitric acid, 10 parts sulfuric acid, and 1 hydrofluoric acid. Thorium samples were examined as polished.

Electron microscopy studies were performed on replicas of samples that were repolished mechanically following the optical microscopy examination. The specimens were either cathodic vacuum etched⁽⁸⁾ or etched in the glacial acetic-nitric acid solution after a light electropolish.⁽⁹⁾ Electron micrographs were taken from replicas of what seemed to be the regions of maximum uniform bubble density. Bubble diameters and densities were determined from the electron micrographs by point counting techniques.

IV. OBSERVATIONS

The presence of the gas band was not detected with optical microscopy in the samples in the as-irradiated condition; subsequent heat treatments as listed in Table II were necessary to develop an observable bubble formation. The resulting bubbles were divided into two size classes; Class I, diameters ranging from 0.03 to 1.0 microns, and Class II, diameters ranging from 10 to 150 microns. Summaries of the effects of the type of unalloyed uranium, alloy content, pre-irradiation history and annealing conditions on the formation of Class I and Class II bubbles are given in Tables IV and V respectively. The method used for the determination of average bubble diameter is given in Appendix A.

A. Ingot Uranium

- (1) No gas band was observed either as individual bubbles or as an etching effect in the as-irradiated specimens. (Fig. 1).
- (2) Class I bubbles were developed in the gas band after anneals at 600°C and above (Fig. 2).

- (3) Heat treatments for 90 hours at 900, 975, and 1075°C give increasingly larger Class I bubbles (Table IV); bubble diameters have increased by a factor of about three from 900 to 1075°C (Fig. 3).
- (4) Growth of Class I bubbles at 900°C is slow (Table IV).
- (5) Class II bubbles were observed only after annealing at 1075°C. These bubbles can cause bumping of the surface (Fig. 2d).
- (6) A portion of the bubble band appears free of bubbles in some of the samples; a boundary is found in the middle of the bubble-free region (Fig. 2e).
- (7) Little if any difference in bubble structure, (i.e. size and distribution), is seen in the cold-worked material given similar annealing treatments (Fig. 4).
- (8) The appearance of the bubble structure resulting from annealing cold worked ingot in the beta phase (730°C) is characteristically different than that resulting from either alpha or gamma phase anneals (Fig. 4).
- (9) Thermal cycling in the alpha region and between the alpha-beta phases did not change the characteristic structure of the bubble band (Fig. 5).
- (10) A change of the characteristic bubble formation was observed when the irradiation was interrupted for various annealing cycles (Figs. 6, 7, and 8).
- (11) Limited studies of arc melted ingot uranium indicated bubble formation similar to that observed in both the annealed and cold worked ingot uranium (Fig. 9).

B. Dingot Uranium

- (1) Class II bubbles were observed after annealing treatments at temperatures as low as 900°C (Fig. 10). The number and size of the Class II bubbles increased with both temperature and time at temperature (Fig. 10, 11, 12, and 13).
- (2) The bubble band between the Class II bubbles contains a bubble-free region with a boundary running through the center of the band (Fig. 11).
- (3) Post-irradiation annealing within the beta phase (Fig. 14) appears to have resulted in a significantly lower Class I bubble density than that developed at either 600 or 900°C of above.
- (4) After the 90-hour post-irradiation anneal at 600°C the sample initially in the cold-worked condition shows a closely spaced network of sub-boundaries within the helium gas band (Fig. 14a).
- (5) Thermal cycling in the alpha phase caused no change in the bubble formation; however, 100 alpha-beta thermal cycles apparently caused the formation of relatively large Class I bubbles (about 0.4 micron diameter) (Fig. 15b). Twenty-five alpha-beta cycles did not result in the larger bubbles (Fig. 15c). (It should be noted that the cycling patterns of the two samples were not identical in temperature or times.)
- (6) The combination of thermal cycling and isothermal heat treatment does not cause unusual swelling (Fig. 15); however, some bubble coalescence is observed after the 4-hour isothermal gamma treatment following the 100 alpha-beta thermal cycles (Fig. 15d and e).

C. High Purity As-Cast Uranium

- (1) Class I bubbles appear smaller in the high purity uranium than in ingot uranium when the materials are given the same heat treatment (Fig. 16).
- (2) Class II bubbles were observed in material given 25 thermal cycles between the alpha-beta phases followed by 90 hours at 900°C (Fig. 16c). Annealing for 24 hours at 900°C (Fig. 16a) did not develop Class II bubbles.

D. Uranium-Molybdenum Alloys

- (1) Class I bubbles were not resolved at 1500X in either U - 1.6 ^W/o Mo or U - 2.75 ^W/o Mo after the various post-irradiation annealing treatments up to 1075°C; however, an "etching effect" associated with the gas band was observed in both materials (Figs. 17 and 19).
- (2) Thermal cycling of the U - 1.6 ^W/o Mo alloy through the alpha-beta transformation did not result in Class I bubbles that were resolved at 1500X (Fig. 18). However, at the expected location of the gas band, a single phase is found in contrast to the two-phase structure of the normal alloy.
- (3) Although Class I bubbles were not resolved, Class II bubbles were formed in the 1.6 ^W/o Mo alloy during anneals of 90 hours at 975°C (Fig. 17d) and in the 2.75 ^W/o Mo alloy during anneals of 2 hours at 1075°C (Table V).

E. U - 0.3 ^W/o Cr - 0.3 ^W/o Mo

- (1) For post-irradiation treatments between 600 and 975°C, bubble formation in the U - 0.3 ^W/o Cr - 0.3 ^W/o Mo alloy is similar to that observed in ingot uranium. No Class II bubbles were formed after 90 hours at 975°C (Fig. 20d).
-

- (2) Thermal cycling in the alpha-beta phases does not appear to change the characteristics of the Class I bubbles (Fig. 21). (The cracks observed in material are believed to be the result of pre-irradiation heat treatment).

F. U - 2^w/o Zr

- (1) Class I bubbles were not resolved at 1500X after isothermal anneals up to 975°C; however, anneals at 1075°C revealed their presence (Fig. 22c).
- (2) Thermal cycling treatments had no noticeable effect on the Class I bubble formation (Fig. 22d).
- (3) Electron microscopic examination of specimens annealed at 600 and 900°C (Fig. 23) revealed the following:
- (a) Bubbles approximately one half the diameter of those found in unalloyed uranium after comparable heat treatments.
 - (b) An oriented pattern of Class I bubbles formed during the 600°C anneal (Fig. 23a). These bubbles had little tendency to form at the surfaces of the second phase that had spheroidized during this anneal.
 - (c) Smaller and more numerous Class I bubbles formed at 900°C than at 600°C. The 900°C bubbles are randomly distributed in some areas of the bubble band (Fig. 23c) and aligned in parallel arrays in other areas (Fig. 23b).

G. U - 3.9^w/o Si

- (1) Gross Class II bubble formation was observed during anneals at 900 and 975°C (Fig. 24). (No data were obtained at lower annealing temperature).

H. Thorium

- (1) Voids at grain boundaries were formed in the gas band during anneals at 900°C and above. The porosity is apparently the result of eutectic melting of impurities and gas agglomeration (Fig. 25).

- (2) No Class I bubbles were observed within the grains (Fig. 25).

V. DISCUSSION

A. Unalloyed Uranium

In post-irradiation annealing of unalloyed uranium from 600 to 1075°C gas bubbles of two classes are formed: Class I bubbles with diameters smaller than 1 micron and in concentrations of 10^{13} to 10^{15} bubbles per cc and Class II bubbles with diameters in the range of 10-150 microns and in concentrations of about 10^3 to 10^5 bubbles per cc. Bubbles of intermediate size were not seen. Class I bubbles are estimated to account for volume increases up to about 10 percent. Class II bubbles resulted in apparent volume increases of the order of several hundred percent. The initial annealing experiments at 600°C showed little if any difference between bubble formation behavior in the various grades of unalloyed uranium. As a result, annealing was performed at higher temperatures within the beta and gamma phase region to enhance bubble formation and growth.

Regardless of uranium purity, Class I bubbles appear to be nucleated homogeneously. During isothermal anneals in the gamma phase region, Class I bubbles both increased in size and decreased in number with either longer annealing time or with higher temperature, temperature having a more pronounced effect than time. Measurements of bubble diameters in specimens annealed for a constant time of 90 hours showed a three-fold increase in size

from 900 to 1075°C. These data permitted calculation of the activation energy of the bubble nucleation and growth (see Appendix B). This activation energy, 20 Kcal/mole, is essentially the same as that for self-diffusion of uranium in the gamma phase,⁽¹⁰⁾ indicating that the growth of the bubble is influenced by a diffusion process.

On the other hand, little difference was seen in the size of Class I bubbles resulting from anneals in the alpha phase temperature region, 600°C, and in the gamma phase temperature region, 900°C. This absence of a difference may be the result of a very slow rate of growth throughout this temperature range or to the limited accuracy of measurement of bubble diameters.

For uranium in the cold-worked condition immediately prior to cyclotron irradiation, there appeared to be little effect of the cold work on the bubble formation characteristics during the various anneals. The most significant effect occurred during the anneals in the beta phase region where the Class I bubble density is significantly less than for both the annealed and cold worked specimens subsequently annealed in either the alpha or gamma region. It is not known whether this lower bubble density is a result of the higher strength of the beta phase, or is associated with some anomalous behavior, since data were not obtained on uranium specimens initially in the annealed condition.

As noted above, Class II bubbles formed more easily in uranium when the uranium was of greater purity. This is particularly evident in samples of dingot uranium where Class II bubbles occurred during 900°C anneals, whereas a temperature of 1075°C was required for formation of similar bubbles in ingot uranium which has a significantly higher impurity content. As noted in the preceding section (IV. A. Item (6) and B. Item (2)) when Class II bubbles are formed the appearance of the bubble band adjacent to the Class II bubbles is often significantly altered in that the central part of the bubble band appears free of any Class I bubbles. Also, a line of discontinuity appears in the center of the band.

Thermal cycling of cyclotron-irradiated samples did not, in general, appear to alter the behavior of either Class I or Class II bubbles. However,

in dingot uranium thermal cycling between the alpha and beta phase regions did result in a significant growth of Class I bubbles; no such effect was noted in ingot uranium. The only effect of thermal cycling on Class II bubble behavior was seen in high purity uranium, which developed Class II bubbles during a combined heat treatment of alpha-beta thermal cycling followed by a 90-hour anneal at 900°C.

B. Alloys

The most significant observation on bubble formation behavior in dilute alloys of uranium, specifically U - 1.6 ^W/o Mo, U - 2.75 ^W/o Mo and U - 2 ^W/o Zr, is that the Class I bubbles that were formed are smaller than those formed in the unalloyed uranium. Class I bubbles were not resolved in these alloys by optical microscopy after isothermal anneals in either the alpha, beta or gamma phases. Electron microscopy performed on the U - 2 ^W/o Zr alloy did reveal Class I bubbles and confirmed that these were in fact smaller than those formed in unalloyed uranium.

In the U - 2 ^W/o Zr alloy it is interesting to note that the Class I bubbles formed during a 600°C alpha anneal are both larger and fewer in number than those resulting from a 900°C gamma anneal. The reason for this difference is not known but it is probably related to a difference in availability of nucleation sites for bubble formation.

Although Class I bubbles cannot be resolved by optical microscopy in the alloys containing either molybdenum or zirconium, the gas band is readily apparent in the micrographs as a result of the significant difference in etching characteristics of the matrix in this region. For some unexplained reason the metal within the gas band appears to be a single phase in contrast to the normal two-phase structure of these alloys. No identification of this phase was made but its occurrence may be related to an effect of the very fine bubble dispersion on the transformation characteristics of the alloys during the post-irradiation heat treatment.

The very limited work done on the U - 3.9 ^W/o Si alloy and on thorium was insufficient to permit any significant conclusions on these alloys. In

neither case were Class I bubbles observed; only Class II bubbles or gross grain boundary porosity formed during the high temperature anneals. Some of this porosity may be the result of liquid phase formation during the anneals. Thorium is known to contain impurities that will result in eutectic melting at grain boundaries at a temperature of no more than 850°C.

C. Comparison with Swelling in Neutron-Irradiated Uranium

Results of post-irradiation annealing experiments with uranium irradiated in-pile at low temperatures are similar to those described in this report for uranium irradiated with alpha particles in the cyclotron. For example, Eldred⁽¹¹⁾ reports that when a natural uranium sample that had been irradiated to 0.17 percent burn-up at about 300°C was annealed at 880°C, the density initially decreased at a rapid rate and then at a slow rate until a 10 percent decrease accumulated after 50 hours. At this time, the rate of change in density increased rapidly until the over-all density decrease reached 30 percent; after this point no further decreases in density were reported for times up to 100 hours. Eldred reports that, prior to neutron irradiation, the specimen contained about 0.1 percent total porosity. Presumably the small but relatively rapid density decrease observed in the first few hours was caused by the formation of Class I bubbles. The subsequent gradual increase of volume to 10 percent swelling after about 50 hours may have been related to growth of the pores that were present prior to irradiation; the rapid swelling rate after 50 hours occurred by loss of coherency, causing the observed growth of Class II bubbles. Eldred reports that between 20 and 50 hours the diameters (0.4 micron) and densities (about 10^{12} per cc) of small bubbles remained constant. The larger bubble diameters and smaller densities reported by Eldred compared with those in Table IV may be due to the fact that Eldred's data are based on (a) examination at 1200X, where the smaller and more numerous bubbles were not resolved, or (b) burn-up giving gas concentrations approximately one tenth of that present in the alpha bombarded material.

A similar pattern of change of density with time was found by Churchman and Barnes⁽⁷⁾ for material irradiated to higher burn-up, 0.4 percent. Following

irradiation at temperatures less than 300°C, samples were annealed at temperatures below 500, at 575, at 800 and at 1000°C, for up to 100 hours; these samples swelled less than 0.5 percent, 2 percent, 4 percent and 20 percent respectively. The data indicate that swelling occurred mostly in the first few hours and did not increase significantly with increasing time at temperatures of 575 or 800°C.

Greenwood's data⁽¹²⁾ suggested that the large volume increases observed for uranium irradiated at 600°C are associated with thermal cycling; i.e., in the absence of thermal cycling, swelling no greater than about 10 percent per atomic percent burn-up was observed at irradiation temperatures as high as 600°C for unalloyed uranium. If coherency is lost at boundaries between grains, at twins or at second-phase particles, the formation of Class II bubbles can result.⁽¹³⁾ Stresses leading to loss of coherency may result from thermal cycling or phase transformations.

VI. TABLES AND FIGURESTABLE IMaterials Used in Cyclotron Irradiation Experiments

Material	Pre-Irradiation History
Uranium, ingot	Cast, extruded, rolled and annealed 2 hrs at 850°C
Uranium, ingot	Cast, extruded and rolled
Uranium, ingot	Arc melted, machined
Uranium, dingot	Extruded, rolled and annealed 2 hrs at 850°C
Uranium, dingot	Extruded and rolled
High-purity uranium	Cast, machined
U-1.6 ^{w/o} Mo	Cast, extruded, rolled and annealed 2 hrs at 850°C
U-2.75 ^{w/o} Mo	
U-0.3 ^{w/o} Cr-0.3 ^{w/o} Mo	Cast, extruded, oil quenched from 800°C
U-2 ^{w/o} Zr	Cast, extruded, rolled and annealed 2 hrs at 850°C
U-3.9 ^{w/o} Si	Cast, machined
U-3.9 ^{w/o} Si	Cast, epsilonized 7 days at 800°C, machined
Thorium (reactor grade)	Cast, extruded, rolled and annealed 2 hrs at 850°C
Thorium ("high-purity")	

TABLE II

Summary of Post-Irradiation Annealing Treatments

(For uranium and uranium alloys after alpha-particle irradiation)

Heat Treatment			Purpose
	Temp. (°C)	Time (hrs)	
Isothermal	600	90	To study the kinetics of helium gas bubble nucleation, growth, and coalescence.
	730	90	
	900	2 to 1000	
	975	2 to 90	
	1075	2 to 90	
Thermal Cycling	550 20	1*) 0.3) 50 cycles	Alpha thermal cycle - to study the effect of internal stresses due to anisotropic thermal expansion on bubble nucleation, growth, and coalescence.
	A. 730 600 B. 715 600	1) 1) 100 cycles 10.5**) 1.5) 25 cycles	Alpha-beta thermal cycle - to study the effects of phase transformations on bubble nucleation, growth, and coalescence.
	800 475	1*) 47) 10 cycles	Alpha-gamma thermal cycle - same as above. (Long time at 475°C adopted to permit decomposition of the gamma phase in U-10 ^{w/o} Mo alloys.***)
Combined Isothermal and Thermal Cycling	900 followed by 550 20	90 1) 0.3) 50 cycles	To study the effect of thermal cycling on the growth of bubbles that were formed by annealing at high temperatures.
	900 followed by 730 600	90 1) 1) 100 cycles	To study the effect of phase transformation on the growth of bubbles that were formed by annealing at high temperatures.
	Alpha-beta thermal cycle ("B." above) followed by 900 1000 900	4 4 90	To study the effect of a high temperature anneal on the growth of bubbles that were formed by thermal cycling treatments at lower temperatures.

- * Specimens inadvertently held for 20 hours at 550°C prior to initiation of cycling.
 ** This alpha-beta cycle was included for the U-Mo alloys but was also given to some unalloyed specimens for comparison.
 *** Results for U-10 ^{w/o} Mo alloy reported in NMI-1239 (Ref. 13).

TABLE IIISummary of Interrupted Irradiation and Annealing Experiments*

Run No.	Irradiation Schedule (microampere-hours)	
	Group 1	Group 2
1	7	1
2	7 S**	2
3	7	8 S
4	7 S	4
5	10 S	10 S
6	12 S	25 S

* Each group consists of four ingot uranium samples, A, B, C, and D. The heat treatments for the samples after each irradiation are given below:

A - 16 hours at 900°C

B - 16 hours at 550°C

C - 10 thermal cycles 550°C (1 hr) - 20°C (20 min.)

D - 10 thermal cycles 730°C (1 hr) - 600°C (1 hr)

** S = Section of sample prepared for metallographic analysis.

TABLE IV

Summary of Electron Micrograph Data on Class I Bubbles as a Function of Alloy Content

Material ⁽¹⁾	Post-Irradiation Annealing Treatment		Observed Data			Calculated Data		
	Time (hr)	Temp. (°C)	No. of Areas Counted ⁽²⁾	Total Area Counted (mm ²)	No. of Bubbles	$\bar{d}$ (microns)	Bubble Density $N_v \times 10^{-14}$ (cm ⁻³)	$\frac{\Delta V}{V}$ ⁽³⁾ (%)
Dingot uranium	90	900 ⁽⁴⁾	3	4.53×10^{-6}	175	0.0509	7.6	5.53
Ingot uranium	90	900 ⁽⁴⁾	4	4.13×10^{-6}	85	0.0651	3.2	4.77
	1000	900 ⁽⁵⁾	7	6.45×10^{-6}	34	0.0876	1.26	4.64
	90	975 ⁽⁵⁾	3	84.0×10^{-6}	1125	0.0974	1.37	7.14
	90	1075 ⁽⁵⁾	3	66.4×10^{-6}	259	0.170	0.23	6.35
U - 2 W/o Zr	90	600 ⁽⁴⁾	2	21.2×10^{-6}	544	0.040	6.43	2.20
	90	900 ⁽⁴⁾	2	8.69×10^{-6}	1091	0.030	41.5	6.29

(1) All samples annealed prior to irradiation.

(2) In some cases larger areas than those reported above were evaluated, but the above data are considered to be the most reliable of those available.

(3) Swelling due to Class I bubbles only.

(4) Electron micrographs prepared at SRL.⁽⁸⁾(5) Electron micrographs prepared at Manufacturing Laboratories.⁽⁹⁾

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TABLE V

Occurrence of Class II Bubbles as a Function of Alloy Content and History

		Number of Class II Bubbles ⁽¹⁾ (after post-irradiation anneal)											
Material	Condition in which Irradiated	600°C	720°C	900°C					975°C			1075°C	
		90 hr.	90 hr.	2	24	90	300	1000	2	24	90	2	90
Dingot uranium	Annealed	n	n	n	3	2	2	12	30	5	v	v	v
	Cold worked	n	n			2	5	> 1			v		v
Ingot uranium	Annealed	n		n	n	n		n	n		n	> 2	15
	Cold worked	n	n			n	n	?			n		
	Arc melted		n			n	n	n			n		v
High purity uranium	As cast			n	n							10	> 3
U-1.6 ^{w/o} Mo	Annealed	n	n			n	n	n			v		12
U-2.75 ^{w/o} Mo	Annealed	n	n			n		n			n	> 2	n?
U-0.3 ^{w/o} Cr-0.3 ^{w/o} Mo	800°C, 1 hr, oil quench	n	n			n	n	n			n		
U-2 ^{w/o} Zr	Annealed	n				n	n	n			n		v
U-3.9 ^{w/o} Si	As cast										7		
	Epsilonized						17	v			v		

(1) Number of Class II bubbles observed in one transverse section are reported. Symbols have following meaning: n = no Class II bubbles observed, v = fairly continuous void about 50 microns wide.

65X Bt. Lt.

U-361

Fig. 1 - Cyclotron-irradiated ingot
uranium as irradiated.

1500X

U-379-3

(a) 90 hr at 600°C

1500X

U-362-2b

(b) 90 hr at 900°C
See Fig. 3a for same
specimen at 20,000X.

1500X

U-372-1a

(c) 90 hr at 975°C
See Fig. 3b for same
specimen at 5000X.

25X

U-371-1a

(d) 90 hr at 1075°C

1500X

U-371-1c

(e) 90 hr at 1075°C
Center of bubble band.
See Fig. 3c for same
specimen at 7500X.

Fig. 2 - Bubble formation in cyclotron-irradiated ingot uranium after indicated post-irradiation anneals.

20,000X

631-C

(a) 90 hr at 900°C. See Fig. 2b
for same specimen at 1500X.

5000X

1618-E

(b) 90 hr at 975°C.
See Fig. 2c for same
specimen at 1500X.

7500X

1503-A

(c) 90 hr at 1075°C.
See Fig. 2e for same
specimen at 1500X.

Fig. 3 - Bubbles in cyclotron-irradiated ingot uranium
after indicated post-irradiation anneals.

1500X U-379-5a
(a) 90 hr at 600°C

1500X U-380-4a
(b) 90 hr at 730°C

1500X U-374-1b
(c) 90 hr at 900°C

1500X U-375-2b
(d) 90 hr at 975°C

Fig. 4 - Bubble formation in cyclotron-irradiated cold-worked ingot uranium after indicated post-irradiation anneals.

1500X

U-367-2b

(a) 20 hr at 550°C; 50 cycles,
550°C for 1 hr, 20°C for
20 min.

1500X

U-373-1a

(b) 100 cycles, 730°C for 1 hr,
600°C for 1 hr. Compare
Fig. 15b for dingot.

1500X

U-386-9a

(c) 100 cycles, 730°C for 1 hr,
600°C for 1 hr; 4 hr at
900°C.

1500X

U-386-10a

(d) 100 cycles, 730°C for 1 hr,
600°C for 1 hr; 4 hr at
1000°C.

1500X

U-386-11a

(e) 90 hr at 900°C; 100 cycles,
730°C for 1 hr, 600°C for
1 hr.

Fig. 5 - Bubble formation in cyclotron-irradiated ingot uranium after indicated combinations of isothermal and thermal cycling post-irradiation anneals.

One 50 μ amp-hr cyclotron irradiation followed by one post-irradiation heat treatment	Total of six interrupted cyclotron irradiations (50 μ amp-hr cumulative exposure) and post-irradiation heat treatments
<p style="text-align: center;">U-362-2b</p> <p>(a) Post-irradiation anneal 90 hr, 900°C</p> <p style="text-align: center;">U-379-3</p> <p>(c) Post-irradiation anneal: 90 hr, 600°C</p>	<p style="text-align: center;"><u>Group 1*</u></p> <p style="text-align: center;">U-418-4c</p> <p>(b) Heat treatment A: 16 hr, 900°C after each of six exposures</p> <p style="text-align: center;">U-418-3b</p> <p>(d) Heat treatment B: 16 hr, 550°C after each of six exposures</p>

Fig. 6 - Bubble formation in ingot uranium after continuous cyclotron irradiation and post-irradiation annealing, compared with interrupted cyclotron irradiations and anneals (1500X, etched).

* See Group 1 in Table III for irradiation schedule.

(Continued on next page)

One 50 μ amp-hr cyclotron irradiation followed by one post-irradiation heat treatment	Total of six interrupted cyclotron irradiations (50 μ amp-hr cumulative exposure) and post-irradiation heat treatments
<div data-bbox="181 457 718 798"></div> <p data-bbox="586 808 718 840">U-367-2b</p> <p data-bbox="223 851 685 989">(e) Post-irradiation anneal: 20 hr at 550°C then 50 cycles: 1 hr at 550°C; 20 min. at 25°C</p> <div data-bbox="181 1138 718 1457"></div> <p data-bbox="586 1468 718 1500">U-373-1a</p> <p data-bbox="223 1521 735 1627">(g) Post-irradiation anneal: 100 cycles: 1 hr at 720°C; 1 hr at 600°C</p>	<p data-bbox="1090 372 1230 414"><u>Group 1*</u></p> <div data-bbox="875 457 1412 798"></div> <p data-bbox="1288 808 1412 840">U-418-7</p> <p data-bbox="900 851 1445 989">(f) Heat treatment C; 10 cycles: 1 hr at 550°C; 20 min. at 25°C after each of six irradiations</p> <div data-bbox="875 1138 1412 1457"></div> <p data-bbox="1272 1468 1412 1500">U-418-6a</p> <p data-bbox="900 1521 1445 1649">(h) Heat treatment D; 10 cycles: 1 hr at 730°C; 1 hr at 600°C after each of six irradiations</p>

Fig. 6 (Continued)

* See Group 1 in Table III for irradiation schedule.

Fig. 7 - Bubble formation in ingot uranium after various cyclotron irradiations, each exposure followed by 16-hour heat treatment at 900°C (1500X, etched).

* See Table III for irradiation schedule.

Group 1*

Group 2*

U-376-4a

(a) 2 irradiations,
14 microampere-hours

U-384-4a

(b) 4 irradiations,
28 microampere-hours

U-407-1b

(c) 5 irradiations, 28 microampere-hours
gas content equal to that in Fig. 3b.
above.

U-418-6a

(d) 6 irradiations, 40 microampere-hours

U-418-5c

(e) 6 irradiations,
50 microampere-hours

Fig. 8 - Bubble formation in ingot uranium after various cyclotron irradiations, each exposure followed by 10 cycles from 730°C to 600°C, 1 hour at each temperature (1500X, etched).

100X

U-380-3a

(a) 90 hr at 730°C

1500X

U-380-3b

(b) 90 hr at 730°C

1500X

U-374-3b

(c) 90 hr at 900°C

1500X

U-385-6a

(d) 1000 hr at 900°C

Fig. 9 - Bubble formation in cyclotron-irradiated arc-melted ingot uranium after indicated post-irradiation anneals.

1500X

U-402-6b

(a) 2 hours

100X

U-402-7a

(b) 24 hours

1500X

U-446-1d

(c) 90 hours

Fig. 10 - Bubble formation in cyclotron-irradiated dingot uranium after post-irradiation anneals at 900°C for the indicated times.

1500X

U-402-8b

(a) 2 hours

100X

U-372-2a

(b) 90 hours

1500X

U-372-2b

(c) 90 hours

Fig. 11 - Bubble formation in cyclotron-irradiated dingot uranium after post-irradiation anneals at 975°C for the indicated times.

1500X

U-398-3

(a) 2 hours

50X

U-371-2b

(b) 90 hours

1500X

U-371-2e

(c) 90 hours

Fig. 12 - Bubble formation in cyclotron-irradiated dingot uranium after post-irradiation anneals at 1075°C for the indicated times.

1500X

U-379-1a

(a) 600°C

1500X

U-380-1a

(b) 730°C

1500X

U-362-1c

(c) 900°C

1500X

U-372-2b

(d) 975°C

1500X

U-371-2e

(e) 1075°C

Fig. 13 - Bubble formation in cyclotron-irradiated dingot uranium after post-irradiation anneals for 90 hours at indicated temperatures.

1500X

U-379-2a

(a) 90 hr at 600°C
Compare Fig. 13a.

1500X

U-380-2

(b) 90 hr at 730°C
Compare Fig. 13b.

1500X

U-374-2b

(c) 90 hr at 900°C
Compare Fig. 13c.

1500X

U-378-1b

(d) 300 hr at 900°C

1500X

U-375-3e

(e) 90 hr at 975°C

1500X

U-383-1a

(f) 90 hr at 1075°C
Compare Figs. 12c
or 13e.

Fig. 14 - Bubble formation in cyclotron-irradiated cold-worked dingot uranium after indicated post-irradiation anneals.

25X

U-373-2a

(a) 100 cycles, 600°C for 1 hr,
730°C for 1 hr.

1500X

U-373-2b

(b) 100 cycles, 600°C for 1 hr,
730°C for 1 hr. Compare
Fig. 5b for ingot.

1500X

U-392-2a

(c) 25 cycles, 600°C for 15 hr,
715°C for 10.5 hr.

1500X

U-386-1a

(d) 100 cycles, 730°C for 1 hr,
600°C for 1 hr; 4 hr at
900°C.

1500X

U-386-2d

(e) 100 cycles, 730°C for 1 hr,
600°C for 1 hr; 4 hr at
1000°C.

U-386-3a

(f) 90 hr at 900°C; 100 cycles,
730°C for 1 hr, 600°C for
1 hr.

Fig. 15 - Bubble formation in cyclotron-irradiated dingot uranium after indicated combinations of isothermal and thermal cycling post-irradiation anneals.

1500X

U-402-2a

(a) 24 hr at 900°C

1500X

U-402-1c

(b) 2 hr at 1075°C

1500X

U-392-5b

(c) 25 cycles, 1.5 hr at 600°C to 10.5 hr at 715°C;
90 hr at 900°C

Fig. 16 - Bubble formation in cyclotron-irradiated high-purity uranium after indicated combinations of isothermal and thermal cycling post-irradiation anneals.

1500X

A-2836-2a

(a) 90 hr at 600°C

1500X

A-2837-10a

(b) 90 hr at 730°C

1500X

A-2813-1a

(c) 90 hr at 900°C

25X

A-2823-1c

(d) 90 hr at 975°C
(not etched)

100X

A-2860-4a

(e) 90 hr at 1075°C (not etched)

Fig. 17 - Bubble formation in cyclotron-irradiated U - 1.6 ^W/o Mo alloy after indicated post-irradiation anneals.

100X

A-2874-8a

(a)

1500X

A-2874-8b

(b)

Fig. 18 - Phase transformation in cyclotron-irradiated U - 1.6 W/o Mo after a post-irradiation thermal cycling anneal (25 cycles, 715°C for 10 hours, 600°C for 1 hour).

1500X

A-2836-10a

(a) 600°C

1500X

A-2837-11d

(b) 730°C

1500X

A-2813-2c

(c) 900°C

1500X

A-2823-2b

(d) 975°C

Fig. 19 - Cyclotron-irradiated U - 2.75 ^W/o Mo after post-irradiation anneals for 90 hours at indicated temperatures.

1500X A-2836-1a

(a) 90 hr at 600°C

1500X A-2837-1a

(b) 90 hr at 750°C

1500X A-2814-a

(c) 90 hr at 900°C

1500X A-2879-a

(d) 90 hr at 975°C

Fig. 20 - Bubble formation in cyclotron-irradiated beta phase
U - 0.3 w/o Cr - 0.3 w/o Mo after indicated post-irradiation
anneals.

1500X

A-2879-5

Fig. 21 - Bubble formation in cyclotron-irradiated beta phase
U - 0.3 W/o Cr - 0.3 W/o Mo after a post-irradiation
thermal cycling anneal (25 cycles, 715°C for 10 hours,
600°C for 1 hour; 4 hours at 900°C).

1500X A-2830-3a
(a) 300 hr at 900°C

25X A-2791-3a
(b) 90 hr at 1075°C

1500X A-2791-3
(c) 90 hr at 1075°C

1500X A-2874-4b
(d) 25 cycles, 10 hr at 715°C
to 1 hr at 600°C; 90 hr
at 900°C

Fig. 22 - Bubble formation in cyclotron-irradiated U - 2^{W/o} Zr after indicated post-irradiation anneals.

Fig. 23 - Cyclotron-irradiated U - 2^W/o Zr after post-irradiation anneals for 90 hours at indicated temperatures.

100X

A-2830-8a

(a) 300 hr at 900°C
(as mech. pol.)

100X

A-2861-3a

(b) 1000 hr at 900°C
(etched)

25X

A-2821-1a

(c) 90 hr at 975°C
(as mech. pol.)

Fig. 24 - Bubble formation in cyclotron-irradiated U - 3.9 ^{W/o} Si after indicated post-irradiation anneals; irradiated in the as-cast condition (a), and after "epsilonionization" (b,c).

500X

A-2743-3c

(a) 90 hr at 900°C

500X

A-2792-1

(b) 90 hr at 975°C

50X

A-2791-1a

(c) 90 hr at 1075°C

Fig. 25 - Bubble formation in "reactor grade" thorium cyclotron-irradiated in the annealed condition and given the indicated isothermal post-irradiation anneals (as mechanically polished).

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APPENDIX A

Determination of Bubble Diameters

A summary of Class I bubble diameters, densities and swelling as determined from available electron micrographs is given in Table IV. These quantities were derived as follows.

The average diameter, $\bar{d}$, is a weighted mean for the bubbles having diameters in different ranges:

$$\bar{d} = \left(\frac{\sum N_i d_i^3}{N_t} \right)^{1/3} \quad \text{Eq (A-1)}$$

where N_i and d_i are the number and diameter of bubbles falling within certain diameter ranges

and N_t is the total number of bubbles.

The sample volume counted, V_c , was determined as follows:

$$V_c = A_c \bar{d} \quad \text{Eq (A-2)}$$

where A_c is the area counted.

The bubble density, N_v , was calculated from $\frac{N_t}{V_c}$ and is expressed in units of bubbles/cc.

Swelling is defined as the percent change in volume and was determined from the equation:

$$\% \frac{\Delta V}{V} = \frac{V_c - V_o}{V_o} \times 100 = \frac{V_B}{V_o} \times 100 = 100 \frac{\pi}{6} \frac{\bar{d}^2 N_t}{A_c - \frac{100\pi}{6} \bar{d}^2 N_t} =$$

$$\frac{52.4 \bar{d}^3 N_v}{1 - 0.524 \bar{d}^3 N_v} \quad \text{Eq (A-3)}$$

where V_c , the volume counted, and V_o , the initial volume of metal, are related by:

$$V_o = V_c - V_B = A_c \bar{d} - 0.524 \bar{d}^3 Nt \quad \text{Eq (A-4)}$$

where V_B is the total bubble volume.

APPENDIX BDetermination of Activation Energy
for Bubble Nucleation and Growth

The average Class I bubble diameter, $\bar{d}$, reported in Table IV, was used to determine the activation energy required for their formation, i.e., nucleation and growth. For such small bubbles, less than one micron, where surface tension, γ , is dominant in determining the size, the relationship between bubble diameter and temperature has been shown to be:⁽⁵⁾

$$\bar{d} = \left[\frac{4\gamma B}{mR} \right] \frac{1}{T} e^{-Q/RT} = \frac{c}{T} e^{-Q/RT}$$

where the term in brackets is a constant, c , as long as bubble size is controlled by surface tension forces,
 T is the absolute temperature,
and Q is the combined activation energy for bubble nucleation and growth.

According to this equation a plot of the natural logarithm of the product $\bar{d}T$ against $\frac{1}{T}$ should yield a straight line, whose slope is $\frac{Q}{R}$. When the data for ingot uranium given 90-hour post-irradiation anneals are plotted in this manner (Fig. B-1), activation energy of 20 Kcal/mole* is determined from the slope of the resultant straight line. This value is in good agreement with that reported for the self-diffusion of uranium in the gamma phase⁽¹⁰⁾ and suggests that the process responsible for the growth of Class I bubbles is associated with the self-diffusion of uranium. Such agreement would be expected if the helium atoms become associated with the vacancies responsible for the diffusion of uranium.

* A value of 40,000 cal/mole for the activation energy was previously reported in NMI-1239⁽¹³⁾ on the basis of preliminary data.

1952

Fig. B-1 - Bubble diameter versus post-irradiation annealing temperature.
Drawing No. RA-2344.

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