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**GAS RELEASE AND COMPRESSION PROPERTIES IN
BERYLLIUM IRRADIATED AT 600 AND 750°C**

J. M. Beeston

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

IDAHO NUCLEAR CORPORATION
NATIONAL REACTOR TESTING STATION
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U. S. ATOMIC ENERGY COMMISSION

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BY

J. M. Beeston*

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IN NUCLEAR SCIENCE ABSTRACTS

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ABSTRACT

Helium released from beryllium irradiated at 600 and 750°C was measured and related to a macro-diffusion coefficient, D . The value of D at 600°C was 6.8×10^{-14} and at 750°C was 1.13×10^{-9} cm²/sec. The magnitude of the macro-diffusion coefficient at 750°C agrees with that predicted for gas diffusion in beryllium, but at 600°C agrees better with the self-diffusion coefficient. The activation energy, G , for release of helium in the equation $D = D_0 \exp\left[-\frac{G}{kT}\right]$ is then 5 eV, which corresponds to the activation energy for re-solution processes. The recoil mechanism for helium release was considered and was shown to be small when appreciable gas was released from the samples. The microstructure of the beryllium showed larger gas-bubble formation in the grain boundaries than within the grains. The compression yield strength of the beryllium irradiated to 1×10^{21} n/cm² $E > 1$ MeV at these temperatures increased only slightly, and although appreciable plastic deformation was sustained in the compression testing, the ultimate compression strength decreased.

SUMMARY

Temperatures of 600 and 750°C are high enough to anneal irradiation hardening effects in beryllium, so that only a slight increase in compression yield strength occurs.

The fraction of helium released from the cylindrical beryllium samples by recoil processes is small above 600°C.

Grain growth is hindered by the presence of irradiation and the formation of gas bubbles in beryllium.

The magnitude of the macro-diffusion coefficient for helium in beryllium at 750°C appears to agree with a grain boundary or surface diffusion mechanism, but at 600°C agrees better with the predicted self-diffusion coefficient in beryllium.

The activation energy obtained from a consideration of diffusion at these temperatures is normally associated with bulk diffusion processes.

The microstructure of the beryllium revealed larger and more numerous gas-bubble formation in the grain boundaries of the irradiated beryllium both with increased time and temperature.

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GAS RELEASE AND COMPRESSION PROPERTIES IN BERYLLIUM IRRADIATED AT 600 AND 750°C^[a]

I. INTRODUCTION

The manner of diffusion and aggregation of helium in beryllium during irradiation at high temperature is a matter of considerable interest both to its use as a structural material and to theories of gas diffusion mechanisms. The behavior of helium in beryllium, irradiated at 70 to 100°C to high exposure and examined or subjected to post-irradiation heating, has been the subject of a number of studies^[1, 2]. Although the inert gases have low solubilities in metals, the retention at 70°C of large amounts of irradiation-produced helium in beryllium is known to occur. Irradiation at elevated temperature has been conducted^[3, 4], and effects on the mechanical properties, swelling, stress rupture, and metallography noted.

A mechanism whereby the helium bubbles impede the motion of dislocations has been noted^[5] and their effect on mechanical property changes outlined. We present evidence that the gas bubbles at the grain boundaries, in conjunction with irradiation, hinder the grain growth that would otherwise occur at 750°C. In this investigation, cylindrical beryllium samples were sealed in capsules and irradiated at high temperature so that the helium given off could be collected and measured. The samples had geometry suitable for compression testing and for analysis for macro-diffusion coefficients. The amount of helium gas likely to be released from the beryllium by recoil or knockout processes from the nuclear reactions was calculated. In some theories^[6, 7] of inert gas release from reactor materials, bodily migration of the gas bubbles is held to occur through a mechanism of surface self-diffusion. This mechanism should result in a large diffusion coefficient. In some cases, a low-activation energy has been noted for surface self-diffusion while, at other times, high activation energy has been obtained^[8, 9].

Although these tests provided only two temperatures and one duplicate to verify precision of the diffusion model, we were able to determine an activation energy for the range of temperature of testing.

[a] Work performed under Contract AT(10-1)-1230 to the Atomic Energy Commission.

II. MATERIALS AND PROCEDURE

The beryllium specimens tested were from hot-extruded, hot-pressed material in the form of solid cylinders, 1/2 inch diameter by 1-1/2 inches long, in accordance with the l/d ratio of 3 for medium length specimens given in ASTM E9-61. The nominal composition of the beryllium is given in Table I.

TABLE I

CHEMICAL COMPOSITION OF BERYLLIUM

Component	Percent
Beryllium	98.50 (min)
BeO	1.20 (max)
C	0.15 (max)
Fe	0.15 (max)
Al	0.14 (max)
Si	0.00 (max)
Mg	0.08 (max)

The specimens were encapsulated in Type 347 stainless steel cans and purged with argon to remove the air. Six thermocouples (four attached to the stainless steel can and two penetrating the can and contacting the beryllium) were used to monitor and regulate the temperature of the beryllium. The temperature was controlled by changing the ratio of helium to nitrogen in a thermal barrier gas annulus around the specimen can. The temperature was estimated to have been controlled to within $\pm 10^\circ\text{C}$ except for Capsule 6; wherein, for a period of 2 out of 7 reactor cycles, it may have reached 50°C higher than the set point of 600°C .

Control specimens were encapsulated, purged with argon, and heated for equivalent lengths of time. They were also thermally cycled an equivalent number of times as the irradiated specimens. No effect of thermal stress was observed on specimens in either capsule.

The density of the irradiated beryllium determined by pycnometer measurements was 1.834 g/cm^3 for the 750°C samples, exposure $0.75 \times 10^{21} \text{ n/cm}^2$, $E > 1\text{MeV}$ [a] and 1.826 g/cm^3 for the 600°C samples, exposure $1.08 \times 10^{21} \text{ n/cm}^2$ (a density decrease from the control sample density of 1.854 g/cm^3 of 1 to 1.5 percent). The growth measurements were hampered by disintegration of the magnesium oxide insulators. The magnesium oxide was evidently reduced by the beryllium vapor (vapor pressure about 10^{-10} atmospheric[10]) and the melted magnesium formed a film on the beryllium. The film was selectively etched away with a solution of ammonium chloride. A final etchant of sulfuric-chromic-phosphoric acid, recommended for preparation of beryllium surfaces preparatory to tensile testing, was used to clean up the samples before compression testing.

[a] All exposure values are for neutron energies $E > 1\text{MeV}$.

III. COMPRESSION PROPERTIES

The specimens were compression tested using a Tinius Olsen tester with a Tinius Olsen sub-press similar to that recommended in ASTM Designation E9-61. The mechanical properties of the as-received beryllium, the control specimens, and the irradiated specimens are given in Table II. The results

TABLE II

MECHANICAL PROPERTIES OF COMPRESSION-TESTED BERYLLIUM

Material	Yield Strength 0.2 Percent Offset (ksi)	Ultimate Strength (ksi)	Plastic Strain (percent) [a]
As-received samples	44.0 ^[b]	191.7	30.1
Capsule 1 -- control samples ^[c] (600°C; 3532 hours; 105 cycles)	46.4	210.1	30.7
Capsule 2 -- control samples (750°C; 1589 hours; 55 cycles)	47.0	212.0	26.5
Capsule 3 -- control samples (600°C; 1992 hours; 52 cycles)	51.2	184.5	27.8
Capsule 6 -- 600°C; 3559 hours (irradiated to 1.08×10^{21} n/cm ²) ^[d]	48.5	147.9	19.6
Capsule 7 -- 750°C; 1586 hours (irradiated to 0.75×10^{21} n/cm ²)	49.3	147.1	19.9
Capsule 8 -- 600°C; 1973 hours (irradiated to 0.88×10^{21} n/cm ²)	64.9	174.7	25.2

[a] Plastic strain was calculated as the strain at the fracture point minus the strain at the yield point.

[b] All values are averages of three measurements.

[c] Control samples were heated at temperatures and times indicated and were thermally cycled the number of cycles equivalent to reactor scrams and shutdowns.

[d] All exposure values are for neutron energies > 1 MeV.

indicate a slight increase in yield strength so that the high temperature anneals out the irradiation hardening and pinning effect of the dislocation trapping defects. The presence of the gas bubbles in the grain boundaries reduces the ultimate strength after plastic flow. The amount of plastic deformation is reduced to approximately 60 percent for the higher temperature and higher exposure. However, appreciable plastic strain still occurs (≈ 20 percent) in the room-temperature compression tests.

IV. MEASURED GAS RELEASED

The helium in the capsule annulus was measured in a Hot Cell gas-sampling apparatus after irradiation. The total helium was calculated on the basis of 22 cm^3 produced He per cm^3 Be per $1 \times 10^{22} \text{ n/cm}^2 > 1 \text{ MeV}$ ^[11]. The irradiation exposure was determined from flux-wire monitors (Fe, Ti, and Ni) placed around the outside of the specimen cans. The data are given in Table III.

TABLE III
HELIUM GAS RELEASE MEASUREMENTS

Capsule Number	Exposure ($\text{n/cm}^2 > 1 \text{ MeV}$)	Volume Beryllium (cm^3)	Total He Produced (cm^3)	Annulus He (cm^3 STP)	Total He in Annulus (percent)
6	1.08×10^{21}	14.40	34.2	0.093	0.27
7	0.75×10^{21}	14.38	23.9	3.213	13.43
8	0.88×10^{21}	14.34	27.7	0.0097	0.035

V. RECOIL OR KNOCKOUT HELIUM

Before describing the calculation of the macro-diffusion coefficient, the question of the fraction of helium released from the beryllium by recoil processes will be considered. The recoil release of helium from the slowing down of alpha particles was calculated from an average energy for the alpha particles from the nuclear reactions. The alpha particles produced within an effective range distance of the surface were assumed to escape.

1. CALCULATION OF AVERAGE ALPHA PARTICLE ENERGY

The ratio of the helium production rates (N_p) from the two reactions,

is given as

$$\frac{N_{p,1}}{N_{p,2}} = \frac{\overline{\sigma_{a,1}}}{\overline{\sigma_{a,2}}} \frac{\phi_{n,1}}{\phi_{n,2}} \approx 4$$

where

1 and 2 denote Reactions (1) and (2), respectively

$\overline{\sigma_a}$ = average reaction cross section

ϕ_n = total flux above the threshold energy.

Since the net result of each reaction, therefore, is two helium atoms, 80 percent of the helium produced would be expected to have energies characteristic of Reaction (1), and 20 percent would be expected to have energies characteristic of Reaction (2).

The reaction probability, ψ , of each reaction is proportional to the product of the reaction cross section, σ , and the number of incident neutrons, n , at that

energy. The value of n was taken from Cranberg et al[12]; σ_2 for Reaction (2) is from Stelson and Campbell[13]; and σ_1 for Reaction (1) is the difference between Marion's[14] total σ_x and σ_2 . The average neutron reaction energy was approximately 4 MeV for Reaction (1) (agreeing with Marion's $2.43 + 1.57 = 4.0$ MeV) and 2.7 MeV for Reaction (2).

Nonrelativistic ($\frac{v^2}{c^2} \approx 10^{-4}$) conservation of energy and momentum, starting from rest with energy Q , gave an average alpha particle energy, E_{He} , for Reaction (1b) ≈ 0.10 MeV; E_{He} for Reaction (2) ≈ 1.05 MeV; and E_{He} for Reaction (1a) ≈ 0.14 MeV.

The values, E_{He} , were weighted according to production rates, $N_{p,1}/N_{p,2}$, and Marion's branching ratio, $1a/1b \approx 90$ percent, to give a total average value of $E_{He} = 0.32$ MeV for both reactions.

2. GAS RELEASED BY RECOIL

The effective range of α -particles with energy of 0.32 MeV was determined, and the helium released within this distance of the surface of the beryllium was compared with the total gas produced and measured.

The stopping power of a sample of electron density, N , to a bullet of charge, Ze , and mass, M , is given by Jackson[15] and Segré[16] for nonrelativistic conditions as

$$\frac{-d\epsilon}{dx} = \frac{2 \pi M N Z^2 e^4}{m \epsilon} \ln \frac{Z m \epsilon}{M h \langle v \rangle} \quad (3)$$

where

m = electron mass

$\langle v \rangle$ = average of electron orbital frequencies of target atom

ϵ = energy of particle

which can be integrated to obtain the range. The range, R , is given by

$$R = K \left[\ln | \ln Y_0 | + 2 \ln Y_0 + \frac{(2 \ln Y_0)^2}{2.2!} + \frac{(2 \ln Y_0)^3}{3.3!} + \dots \right] \quad (4)$$

where

$$K = \frac{M h^2 \langle v \rangle^2}{8 \pi m N Z^2 e^4}$$

and

$$Y_0 = \frac{2m \epsilon_0}{M h \langle v \rangle}$$

The range of the 0.32-MeV α -particles in Be^9 is then $\approx 2.56 \times 10^{-5}$ cm which agrees reasonably well with Wilson's^[17] nomograph.

Since only those α -particles at a distance, R , with direction normal to the surface can escape, an effective range from which all α -particles escape can be determined. It was calculated that the ratio of helium escaping to that produced within the depth, R , is

$$f = \frac{\int_0^R 2\pi R^2 (1 - \cos \theta) dt}{\int_0^R 4\pi R^2 dt} = \frac{1}{4} \quad (5)$$

Therefore, only 1/4 of the alpha particles which are produced within one range of the surface escape in the slowing down process. An effective range, R_e , from which all would escape, thus, can be defined as

$$R_e = \frac{R}{4} = 6.4 \times 10^{-6} \text{ cm}.$$

The fraction (f_p) of helium produced and, hence, released from within this effective range is just the ratio of the annulus volume of thickness, R_e , to the total specimen volume and is

$$f_p = 2.36 \times 10^{-5}.$$

The helium released from recoil processes is, thus, the product of the fraction (f_p) and the total helium produced and is given in Table IV. The percent of the measured helium released from recoil processes is also indicated. These are

TABLE IV

HELIUM SOURCE AND FRACTION RELEASED BY RECOIL

Capsule	Helium Concentration in Annulus (c_a)	Source ($\text{cm}^3 \text{ He}/$ $\text{cm}^3 \text{ Be-sec}$)	Helium Released by Recoil (cm^3)	Helium Released by Recoil (Percent)
6	0.0369	1.64×10^{-7}	8.07×10^{-4}	0.867
7	0.634	2.59×10^{-7}	5.64×10^{-4}	0.0176
8	0.00383	2.39×10^{-7}	6.54×10^{-4}	6.74

insignificant amounts (except for Capsule 8) compared to that measured; hence, permeation or diffusion processes must be the predominant process for release of helium from the specimens. The helium concentration in the annulus and the average production rate of helium are also given.

VI. METALLOGRAPHY AND FRACTOGRAPHY

At the temperature of 600 and 750°C, recrystallization and grain growth occurs in the hot-pressed and hot-extruded beryllium. Photomicrographs of transverse sections of both control and irradiated compression-tested samples, Parts B through G, made with polarized light (Figure 1), show a general enlargement of the grain size as compared with the as-received microstructure, Part A. Irradiation produced defects, and the formation of gas bubbles hinders the grain growth at 750°C, as is clearly seen by comparing Parts F and G. In the irradiated material, the grain size is approximately the same, Parts C, E, and G.

An effect of irradiation on the stringers formed during extrusion can be seen in longitudinal sections, shown in Figure 2. The stringers are broken up more in the irradiated samples held at similar temperatures for the same time. The irradiation, thus, aids in the polygonization of the microstructure.

From the electron fractography (Figure 3), we see that fracture by cleavage principally occurs in the control specimens and intergranular failure occurs in the irradiated. The gas bubbles are larger at the higher exposure of gas concentration and at the higher temperature. The gas appears to diffuse to the grain boundaries and migrate to the specimen surface. The gas bubbles are more numerous and larger in the grain boundaries (Figure 3, Part D) than in the cleavage planes.

- A. AS RECEIVED, HOT-PRESSED, HOT-EXTRUDED.
 B. HEATED AT 600°C FOR 1992 HOURS.
 C. IRRADIATED TO 0.88×10^{21} N/CM² AT 600°C FOR 1973 HOURS

- D. HEATED AT 600°C FOR 3532 HOURS.
 E. IRRADIATED TO 1.08×10^{21} N/CM² AT 600°C FOR 3559 HOURS.

- F. HEATED AT 750°C FOR 1589 HOURS.
 G. IRRADIATED TO 0.75×10^{21} N/CM² AT 750°C FOR 1586 HOURS.

MAGNIFICATION: 200X, POLARIZED LIGHT, TRANSVERSE SECTIONS.

FIG. 1 EFFECT OF TEMPERATURE AND IRRADIATION ON GRAIN GROWTH OF HOT-EXTRUDED BERYLLIUM.

A. HEATED 600°C FOR 1992 HOURS.

B. IRRADIATED TO 0.88×10^{21} N/CM² AT 600°C FOR 1973 HOURS.

C. HEATED 600°C FOR 3532 HOURS.

D. IRRADIATED TO 1.08×10^{21} N/CM² AT 600°C FOR 3559 HOURS.

E. HEATED 750°C FOR 1589 HOURS.

F. IRRADIATED TO 0.75×10^{21} N/CM² AT 750°C FOR 1586 HOURS.

MAGNIFICATION: 200X, LONGITUDINAL SECTIONS.

FIG. 2 EFFECT OF TEMPERATURE AND IRRADIATION ON THE BREAKUP OF THE STRINGER FORMATION IN HOT-EXTRUDED BERYLLIUM.

A, HEATED 600°C FOR 1992 HOURS.

B, IRRADIATED TO $0,88 \times 10^{21}$ N/CM² AT 600°C FOR 1973 HOURS

C, HEATED 600°C FOR 3532 HOURS.

D, IRRADIATED TO $1,08 \times 10^{21}$ N/CM² AT 600°C FOR 3559 HOURS

E, HEATED 750°C FOR 1589 HOURS.

F, IRRADIATED TO $0,75 \times 10^{21}$ N/CM² AT 750°C FOR 1586 HOURS

MAGNIFICATION: 16,000X.

FIG. 3 ELECTRON FRACTOGRAPHS OF CONTROL AND IRRADIATED SAMPLES FROM COMPRESSION-TESTED BERYLLIUM.

VII. MACRO-DIFFUSION COEFFICIENT

Considering the model for calculation of a macro-diffusion coefficient, it is seen that we have a source for the production of helium such that the concentration of helium in the sample increases as a function of time. Concurrently, a concentration gradient is formed as the helium diffuses into the annular space around the specimen. To determine the diffusion of He in Be, two procedures were used:

- (1) Fick's diffusion equation was solved for a finite cylinder, and the solution was generalized to include the production of He in the specimens. This procedure leads to a difficult analysis and inaccuracies unless fundamental modes alone can be considered.
- (2) A computer code for one-dimensional diffusion[18] was used to determine the helium concentration, C_0 , in the annulus as a function of the diffusion coefficient, D . The value of D corresponding to the measured concentration, C_0 , could then be determined.

These procedures are now described.

1. FICK'S DIFFUSION EQUATION

The solution to Fick's diffusion equation,

$$\nabla^2 C - \frac{1}{D} \frac{\partial C}{\partial t} = 0 \quad (6)$$

for the concentration, $C(r, Z, t)$, in a finite cylinder of radius, ρ , length, ℓ , is given[19] as

$$C \propto \left[\sum_{n=0}^{\infty} \frac{1}{2n+1} \sin \frac{(2n+1)\pi Z}{\ell} \exp \frac{-(2n+1)^2 \pi^2 Dt}{\ell^2} \right] \times \left[\sum_{m=1}^{\infty} \frac{J_0\left(\frac{\beta_m r}{\rho}\right)}{\beta_m \cdot J_1(\beta_m)} \exp \frac{-\beta_m^2 Dt}{\rho^2} \right] \quad (7)$$

where β_m is a root of the Bessel function, $J_0(\beta)$. After a long time, t_0 , only the fundamental modes in these expansions will be significant, and the concentration, C , is proportional to

$$f(r, Z) \exp - FDt$$

where

$$F = \frac{\pi^2}{l^2} + \frac{\beta_1^2}{\rho^2}.$$

However, for $10^{-10} < D < 10^{-8}$, only $t_0 > 10^8$ sec is sufficient to consider fundamental modes alone, and a direct solution based on fundamental modes would be erroneous for $t < t_0$.

2. COMPUTER CODE

The computer code for one-dimensional diffusion[18] solves by difference methods the equation

$$D \nabla^2 C + S - \frac{\partial C}{\partial t} = 0 \quad (8)$$

where

$$\nabla^2 C = \frac{\partial^2 C}{\partial r^2} + \frac{1}{r} \frac{\partial C}{\partial r}$$

and

S = the constant source of helium production.

The output of this code is $C(r, t)$ for fixed D . The code requires that $\frac{\partial C}{\partial r}$ and C be continuous at every point, and boundary conditions are applied so that $\frac{\partial C}{\partial r} = 0$ at $r = 0$ (specimen center) and $r = \rho_0$ (the outside radius of the annular gas volume). D in the annular gas volume was set at an arbitrary large number ($\approx 10^{-6}$ cm²/sec) to ensure a uniform concentration in that region.

Since the code does not consider diffusion from the cylinder ends, two cases were considered to compensate for this: (a) Bucklings Equal and (b) Surfaces Equal.

In Bucklings Equal, we require the conditions:

- (1) Sample volumes to remain the same, $V_{sb} = V_{sa}$; b's refer to before and a's refer to after adjusting.
- (2) Annulus volumes to remain the same, $V_{ob} = V_{oa}$.

$$(3) \frac{\pi^2}{l_b^2} + \frac{\beta_1^2}{\rho_b^2} = \frac{\beta_1^2}{\rho_a^2}, \text{ which adjusts the cylinder specimen}$$

surface so that the total diffusion in the radial direction alone is the same as the radial plus end diffusion in the specimen before adjustment.

In Surfaces Equal, we require the conditions:

$$(1) V_{sb} = V_{sa}$$

$$(2) V_{ob} = V_{oa}$$

Surface areas to remain the same, $2 \pi \rho_b^2 + 2 \pi \rho_b \ell_b$
 $= 2 \pi \rho_a \ell_a$, which ensures that the cylindrical surface
area of the adjusted specimen is equal to the total cylindrical
surface plus end areas of the original specimens.

With these values determined, problems were solved with the code for different D's to give curves of D versus C_a (annular gas concentration) from which D for the measured concentration, C_0 , could be determined for samples in each capsule. Values of D and C_a are given in Table V, from which Figures 4 and 5

TABLE V
COMPUTER CODE SOLUTIONS OF C_a FOR D

Capsule	Temperature (°C)	Diffusion Time T (sec)	Bucklings Equal		Surfaces Equal	
			(D)	(C_a)	(D)	(C_a)
6	600	1.45×10^7	1×10^{-14}	0.0125	1×10^{-14}	0.01416
			1×10^{-13}	0.0393	1×10^{-13}	0.04428
7	750	6.38×10^6	1.1×10^{-9}	0.605	1.1×10^{-9}	0.655
			2.0×10^{-9}	0.720	2.0×10^{-9}	0.775
8	600	8.10×10^6	1×10^{-14}	0.00768	1×10^{-14}	0.00867
			1×10^{-15}	0.00253	1×10^{-15}	0.00286

were constructed. The helium source and concentration in the annulus (also indicated in the figures) are given in Table III. A possibility of a leaky gas sampling valve in the measurement of the annulus volume and a discrepancy between the calculated and measured annulus volume by a factor of two was investigated by making solutions for both values. The values of the D's were the same as indicated in Figure 4.

This result may have been expected since the question did not concern the accuracy of the He measured in the annulus (Table III) so that only the value of C_0 and the corresponding annulus volume mesh points changed. These compensated each other. The total time the specimens were in the reactor at power was taken as the diffusion time since the temperature, hence diffusion, was greatly reduced the remainder of the time.

The data were plotted as C_a versus logarithm of D to interpolate for the value of D corresponding to C_0 given in Table IV. It was expected that the true D would be close to the Bucklings Equal case since the diffusion should approach that due to the fundamental modes as t approaches $t_0 = 10^8$ sec. It was discovered, however, that the concentration gradient in the specimen is steeper near

FIG. 4 D VERSUS C_0 FOR He IN Be AT 600°C.

FIG. 5 D VERSUS C_0 FOR He IN Be AT 750°C.

the outer surface (most diffusion occurs for $0.80\rho < r < \rho$), and at short times the diffusion should be proportional to the total surface area of the specimen; accordingly, an average of the two cases is presented as the most probable macro-diffusion coefficient and is summarized in Table VI.

TABLE VI

SUMMARY OF MOST PROBABLE D

Capsule	Surfaces Equal	Bucklings Equal	Most Probable D
6	5.6×10^{-14}	8.0×10^{-14} cm ² /sec	6.8×10^{-14} cm ² /sec
7	0.98×10^{-14}	1.29×10^{-9} cm ² /sec	1.13×10^{-9} cm ² /sec
8	1.5×10^{-15}	1.8×10^{-15} cm ² /sec	1.65×10^{-15} cm ² /sec

The macro-diffusion coefficient has been obtained from a model that does not consider microscopic phenomena. Thus, although there is an order of magnitude difference in the value of D for Capsules 6 and 8 at the temperature of 600°C for diffusion times that differ by a factor of about two, some difference can be predicted from consideration of microscopic phenomena. The contribution from bulk diffusion, D_g , and grain boundary diffusion, D_b , to the macroscopic diffusion can vary with the diffusion time as shown by an equation derived by LeClaire[20]:

$$\frac{D_b}{D_g} = \frac{2 (\pi D_g t)^{1/2}}{\delta} \cot^2 \alpha$$

where

δ = the effective grain boundary width

α = angle between line of constant concentration and grain boundary

t = total time of diffusion down the boundary.

These experimental results can be compared with those predicted from Zeners'[21] theoretical expression for the self-diffusion coefficient and with Rich and Walters'[3] results. Thus, $D_s = 0.7 \exp \frac{-2.16 \text{ eV}}{k T}$ so that at 600°C, $D_s = 2.5 \times 10^{-13}$ and, at 750°C, $D_s = 1.7 \times 10^{-11}$. From the results of Rich and Walters, wherein they conclude that the gas diffusion rates are 100 to 1000 times greater than D_s , we would obtain at 600°C, $D \approx 10^{-11}$ to 10^{-10} and, at 750°C, $D \approx 10^{-9}$ to 10^{-8} . We see that our results at 750°C agree reasonably well with that predicted for gas diffusion in beryllium; however, at 600°C our macro-diffusion coefficient is about the same as predicted for self-diffusion in beryllium and, hence, is much smaller than for gas diffusion. Since the knockout helium fraction is larger for Capsule 8, we shall use the value of D for Capsule 6 in computing the activation energy in the next section. The calculated helium released by recoil was less than 1% for capsule 6 and the correction is therefore smaller.

VIII. ACTIVATION ENERGY

The temperature dependence of D can be obtained from the relationship^[22] that

$$D = D_0 \exp - \frac{G}{R T}$$

where

G = the activation energy.

Using the most probable value of D for Capsules 6 and 7 from Table VI and the corresponding temperatures, a solution of the equation then gives a value of the activation energy, G, of 115,000 cal/g atom or 5 eV per atom and a value of D_0 as 7.1×10^{19} cm²/sec. This represents a large activation energy and would normally be associated with bulk diffusion and re-solution processes. However, the metallography and electron fractography definitely indicate a principal grain boundary effect. The procedure appears to be feasible for obtaining macro-diffusion coefficients, and more experiments need to be done.

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