

Fig. 4. Decrease in density and increase of fission gas release from irradiated Zr-14 wt% U specimens on annealing.

filled bubbles within the material and, consequently, the swelling. The change in the number and diameter of bubbles may be due to sweeping of bubbles before the moving phase boundaries or to enhanced diffusion of the gas atoms along the changing phase boundaries. Since the microstructures of these specimens were not examined, it is not possible to describe the detailed manner whereby either of these processes can increase or decrease the swelling. The data of Johnston³ from annealed irradiated zirconium-8 wt% uranium alloys suggest that phase changes can cause increased swelling.

Alloys containing 6 wt% uranium (Fig. 3) and 14 wt% uranium (Fig. 4) begin to swell on postirradiation annealing at about 450 C which is well below the temperature of a phase change. The onset of swelling at 450 C is probably due to the formation of gas-filled bubbles within and adjacent to the uranium-rich phase since uranium begins to swell at about 450 C on postirradiation annealing⁴.

The evolution of fission-product gas started at about 600 C, the temperature of a phase change in these alloys. These results are in agreement with the conclusion that a change of phases in an alloy can alter the diameter and number of gas-filled bubbles and, consequently, the swelling since the change of phases could allow gas atoms to have access to a free surface. The results of Stubbs and Webster⁵, who investigated the evolution of fission-product gas from a zirconium-5 wt% uranium alloy, show that gas evolution increases during irradiation at about 600 C.

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A Generalization of the Endpoint Method

In asymptotic diffusion theory, the endpoint condition $% \left({{{\left[{{{\left[{{{\left[{{{c}} \right]}} \right]}_{t}}} \right]}_{t}}}} \right)$

$$\phi^{as}(-z_0) = 0 \tag{1}$$

is used as a boundary condition in the solutions of the diffusion equation^a for a homogeneous medium with isotropic scattering if no particles are crossing the surface z = 0 from the region z < 0. As is well known, (1) is obtained from the general solution $\phi(z) = \phi^{as}(z) + \phi^{tr}(z)$ for z > 0 of the homogeneous half-space problem $\{1 - \Lambda\}\phi(z) = 0$.

We want to emphasize that a more general condition

$$\phi_s^{\rm as}\left(-z_0\right) = A \tag{2}$$

may be derived¹ from the general solution $\phi_s(z) = \phi_s^{as}(z) + \phi_s^{tr}(z)$ of the inhomogeneous half-space problem $\{1 - \Lambda\}\phi_s(z) = \sigma(z)$ with a source term $\sigma(z) = \int_0^1 d\mu \,\mu^{-1} e^{-z/\mu} S(\mu)$. $S(\mu)$ is the angular distribution of particles crossing the surface z = 0 in terms of the cosine of the angle between the direction of particles and the inner normal on the surface z = 0. $S(\mu)$ is normalized to unit intensity of the entering particles.

For two inhomogeneous integral equations $\{1 - \Lambda\}\phi_I(z) = \sigma_I(z)$ and $\{1 - \Lambda\}\phi_{II}(z) = \sigma_{II}(z)$ with the same material constant *c* in both, but source terms $\sigma_I(z)$ and $\sigma_{II}(z)$ determined by different angular distributions of entering particles, the ratio

³W. V. JOHNSTON, "The Effects of Transients and Longer-Time Anneals on Irradiated Uranium-Zirconium Alloys," KAPL-1965 (1958).

⁴B. A. LOOMIS and D. W. PRACHT, "Swelling of Uranium on Postirradiation Annealing," J. Nucl. Mat. 10, p. 346 (1963). ⁵F. J. STUBBS and C. B. WEBSTER, "The Release of

⁵F. J. STUBBS and C. B. WEBSTER, "The Release of Fission Product Rare Gas from a Uranium/Zirconium Alloy During Irradiation in the BEPO Reactor," AERE C/M 372 (1959).

^aAll coordinates are measured in mean-free-path units $1/\Sigma$, where Σ is the total cross section. As usual, *c* is the mean number of particles emenating from one collision, Λ is the integral transport operator.

¹K. O. THIELHEIM, Paper presented at the Physikertagung in Hamburg, Sept. 9, 1963.

$$A_I / A_{II} = \int_0^\infty dz \; \frac{\phi(z)}{\phi(0)} \, \sigma_I(z) / \int_0^\infty dz \; \frac{\phi(z)}{\phi(0)} \, \sigma_{II}(z) \tag{3}$$

of boundary values $A_I = \phi_I^{as}(-z_0)$ and $A_{II} = \phi_{II}^{as}(-z_0)$ is obtained from

$$\int_{0}^{\infty} dz \left[\frac{\phi(z)}{\phi(0)} \left\{ 1 - \Lambda \right\} \left(\frac{\widetilde{\phi}_{I}(z)}{A_{I}} - \frac{\widetilde{\phi}_{II}(z)}{A_{II}} \right) \right]$$
$$= \frac{1}{A_{I}} \int_{0}^{\infty} dz \, \frac{\phi(z)}{\phi(0)} \, \sigma_{I}(z) - \frac{1}{A_{II}} \int_{0}^{\infty} dz \, \frac{\phi(z)}{\phi(0)} \, \sigma_{II}(z)$$

and

$$\int_{0}^{\infty} dz \left[\frac{\phi(z)}{\phi(0)} \left\{ 1 - \Lambda \right\} \left(\frac{\widetilde{\phi}_{I}(z)}{A_{I}} - \frac{\widetilde{\phi}_{II}(z)}{A_{II}} \right) \right]$$
$$= \int_{0}^{\infty} dz \left[\frac{\phi(z)}{\phi(0)} \left\{ 1 - \Lambda \right\} \left(\frac{\widetilde{\phi}_{I}^{tr}(z)}{A_{I}} - \frac{\widetilde{\phi}_{II}^{tr}(z)}{A_{II}} \right) \right]$$
$$= \int_{0}^{\infty} dz \left[\left(\frac{\widetilde{\phi}_{I}^{tr}(z)}{A_{I}} - \frac{\widetilde{\phi}_{II}^{tr}(z)}{A_{II}} \right) \left\{ 1 - \Lambda \right\} \frac{\phi(z)}{\phi(0)} \right] = 0,$$

where $\widetilde{\phi}_{l}^{as}(z) = A_{l}e^{-(z+z_{0})/\Sigma L}$ and $\widetilde{\phi}_{ll}^{as}(z) =$

 $A_{II}e^{(z+z_0)/\Sigma L}$ are asymptotic parts of special solutions of the two inhomogeneous integral equations.

From (3) a general expression

$$A = A_{\pi/2} \int_{0}^{\infty} dz \ \sigma(z) \ \phi(z) / \phi(0)$$
 (4)

is found for the boundary value A, from which it may be calculated for any distribution $S(\mu)$ and any value of c with the help of the known homogeneous solution $\phi(z)/\phi(0)$.

$$A_{\pi/2} = \frac{1}{\Sigma L} \sqrt{\frac{2[(\Sigma L)^2 - 1]}{1 - (\Sigma L)^2 (1 - c)}}$$
(5)

is the boundary value for $S(\mu) = \delta(\mu)$ as a function of diffusion length L.

If, for example, the entering particles have an isotropic distribution, $S(\mu) = 1$,

$$A_{\rm is} = \frac{2}{\Sigma L c} \sqrt{\frac{2[(\Sigma L)^2 - 1]}{1 - (\Sigma L)^2 (1 - c)}} , \qquad (6)$$

or, if they have a cosine distribution, $S(\mu) = 2\mu$,

$$A_{\cos} = \frac{4\sqrt{1-c}}{c} \sqrt{\frac{2[(\Sigma L)^2 - 1]}{1 - (\Sigma L)^2 (1 - c)}} .$$
(7)

It may be verified from (3) that for any given value of $c \le 1$, $A_{\pi/2} \le A \le A_0$, where A_0 is the boundary value for orthogonal incidence of particles on the surface, $S(\mu) = \delta(\mu - 1)$.

The application of (2) as a boundary condition to solutions of diffusion equation is very much facilitated by A approaching its value at c = 1 very rapidly as $c \rightarrow 1$. Thus, for all weak absorbers, $\Sigma L \gg 1$, the boundary value A may be taken as a constant, independent of the chemical nature of

the material and dependent on the angular distribution of the entering particles only. For example,

$$A_{\pi/2} = \sqrt{3},$$

$$A_{is} = 2\sqrt{3},$$

$$A_{cos} = 4,$$

$$A_{cos^2} = 6z_0 = 4.262,$$

$$A_{cos^3} = 4z_0^2 + 12/5 = 4.419,$$

$$A_0 = 5.036 \quad \text{for } \Sigma L \gg 1.$$
(8)

The generalized endpoint condition (2) reduces to (1) for vanishing intensity of particles entering the surface. Although (2) is derived from the infinite half-space problem, it may be applied to finite-body problems in the same way as is commonly done with condition (1).

A comparison of (2) with the conventional condition

$$\left[\frac{c}{4}\phi_s^{\text{diff}}(z) - \frac{c}{6}\frac{d}{dz}\phi_s^{\text{diff}}(z)\right]_{z=0} = 1, \qquad (9)$$

which does not account for different angular distributions, shows that the flux determined by (9) may differ from the exact asymptotic behavior by more than a factor 2.

Finally, it may be of interest to mention a relation between the boundary value A of the generalized endpoint method and the albedo

$$\beta = 1 - \sqrt{1 - c} \left[\int_0^\infty dz \,\sigma(z) \,\phi(z)/\phi(0) - \frac{1}{\Sigma L} \int_0^\infty dz \,\frac{\phi(z)}{\phi(0)} \int_z^\infty dz' \,\sigma(z') \right]$$
(10)

of a half space z > 0. With $S(\mu') = \delta(\mu' - \mu)$ one finds

$$\beta_{\mu} = 1 - \sqrt{1 - c} \frac{A_{\mu}}{A_{\pi/2}} (1 - \mu/\Sigma L),$$
 (11)

for example, $\beta_{\pi/2} = 1 - \sqrt{1 - c}$.

The approximate formula

$$\beta = 1 - A \sqrt{1 - c} / \sqrt{3}$$
, for $\Sigma L \gg 1$, (11')

is found after multiplying (11) by $S(\mu)$ and integrating over μ .

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