Swelling of Irradiated Uranium-Zirconium Alloys on Annealing*

The swelling of irradiated zirconium-3, -6, and -14 wt% uranium alloys on annealing them at temperatures up to 900 C was measured in terms of density change, and the effect of the annealing temperature on the release of fission-product gas was determined. The data show that phase changes influenced the swelling of these alloys and their release of fission-product gas.

Specimens[1] with dimensions of $2.5 \times 1.3 \times 0.23$ cm were cut from fuel plates that had been irradiated in a water channel at 20-60 C. The alloy in each specimen was clad with 0.08 cm of zirconium except on two sides (Fig. 1).

The irradiated specimens were heated to the annealing temperature in an evacuated tantalum tube at a rate of 4 C/minute and were maintained at the annealing temperature for ten minutes. After annealing the specimens were cooled in the furnace to room temperature for a density measurement. The density was determined by weighing the specimen in air and in carbon tetrachloride, and the amount of swelling was expressed as the ratio of the change in density on annealing to the density of the specimen after irradiation.

Fission-product gas released from a specimen during heating was collected on activated charcoal cooled with liquid nitrogen. The activity of the adsorbed gas was determined by an end-window Geiger-Müller counter.

The decrease in density and the activity of the gas released from the specimens during annealing are shown in Figs. 2, 3, and 4. In these figures each point on a curve for a specimen represents a cumulative effect of annealing ten minutes at successively higher temperatures. The appropriate temperature of phase changes[2] in the alloys before irradiation is also shown in Figs. 2, 3, and 4.

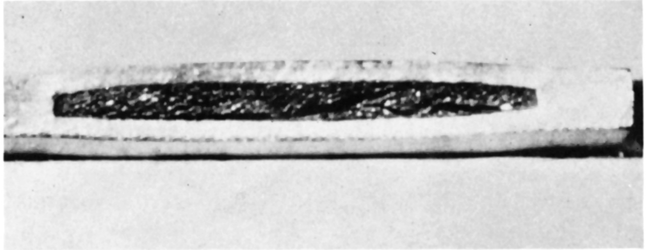

Fig. 1. Macrophotograph of a section of Zr-14 wt% U specimen after irradiation to 2.7 at.% burnup and annealing at 900 C. 3.5 X.

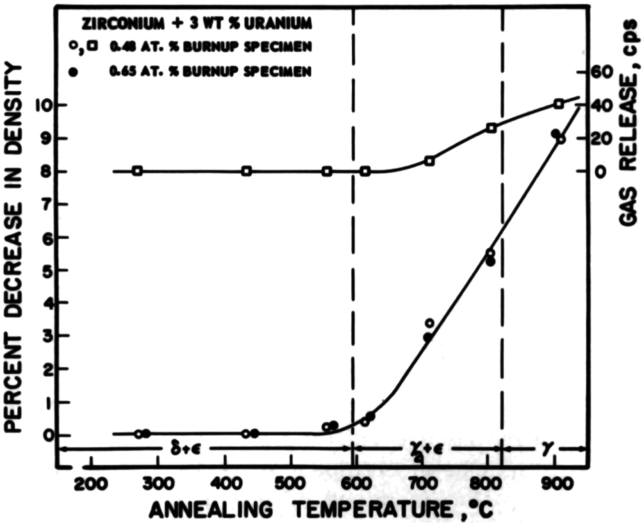

Fig. 2. Decrease in density and increase of fission gas release from irradiated Zr-3 wt% U specimens on annealing.

These data show that a change of phases in an alloy can alter the diameter and number of gas-

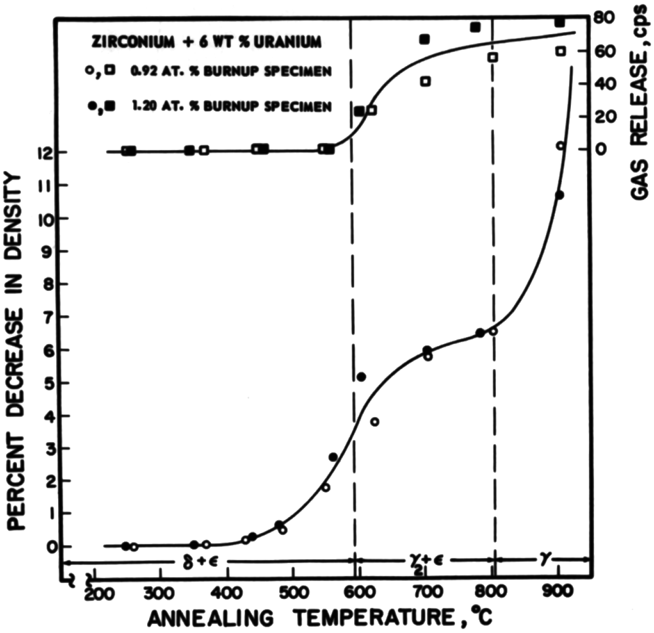

Fig. 3. Decrease in density and increase of fission gas release from irradiated Zr-6 wt% U specimens on annealing.

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*This research was performed under the auspices of the USAEC.

[1] R. E. BAILEY, "Irradiation Effects on Zirconium-Clad Uranium-Zirconium Fuel Plates," ANL-5825 (1958).

[2] F. A. ROUGH and A. A. BAUER, *Constitutional Diagrams of Uranium and Thorium Alloys*, Addison-Wesley Pub. Co., Reading, Mass., p. 85, (1958).

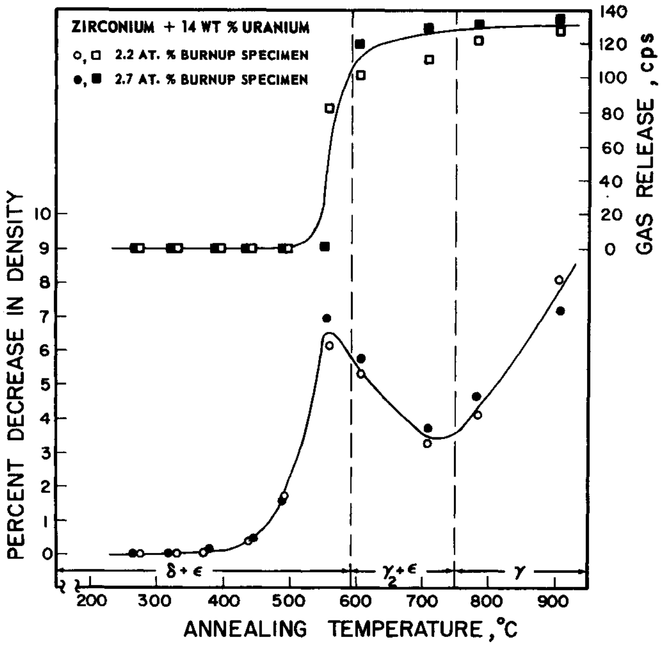

Fig. 4. Decrease in density and increase of fission gas release from irradiated Zr-14 wt% U specimens on annealing.

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 Johnston[3] 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 annealing[4].

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 Webster[5], 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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Received March 30, 1964
Revised May 27, 1964

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

[4] B. A. LOOMIS and D. W. PRACHT, "Swelling of Uranium on Postirradiation Annealing," *J. Nucl. Mat.* **10**, p. 346 (1963).

[5] F. 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).

A Generalization of the Endpoint Method

In asymptotic diffusion theory, the endpoint condition

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

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

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

may be derived[1] 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

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[a] All 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.

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