Radiation Effects in Plutonium

What is known? Where should we go from here?

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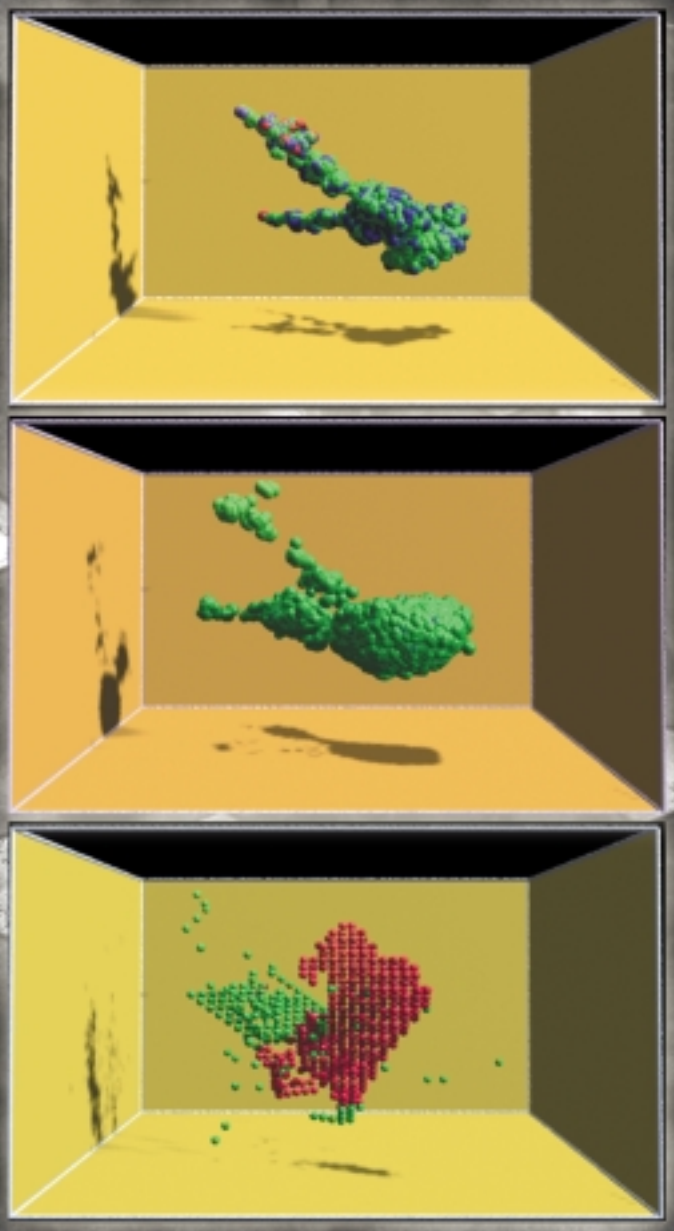

Maintaining confidence in the U.S. nuclear stockpile in the absence of testing presents formidable technical challenges. Chief among them is the assessment of aging effects in the plutonium pit, the central component in the primary of a nuclear weapon. Plutonium is vulnerable to aging because it is a radioactive element, decaying to uranium by emitting an α particle (an energetic helium nucleus He^{++}). Although the main isotope in weapon-grade plutonium, plutonium-239, has a relatively long half-life of 24,000 years, its decay rate is still sufficiently high to lead to a significant buildup of helium and radiation damage within the metal after several decades. The radiation damage is caused mainly by the uranium nuclei, which receive sufficient recoil energy from the decay to knock plutonium atoms from their sites in the crystal lattice of the metal. Empty sites, or vacancies, are left in the wake.

Studies of radiation damage in many metals have shown that these two types of defects (helium in combination with lattice vacancies) can produce a macroscopic effect known as void swelling. On the microscopic level, the vacancies tend to diffuse through the metal and cluster to form voids, or empty spaces. Macroscopically, the net effect is that the metal swells in size. Figure 1 shows all the metallic elements for which radiation-induced void swelling has been observed. Void swelling has also been observed and studied in many alloys.

The record so far indicates that every metal or alloy studied has been found to be susceptible to void swelling at high radiation doses and in the appropriate temperature range. This temperature range is between approximately one-third and three-fifths of the melting point of the metal or alloy. Based on this body of knowledge, the author and others have developed predictive models for many aspects of the swelling process including the generation of lattice vacancies, the creation of helium, the mobility of these defects, and the evolution of clusters of defects.

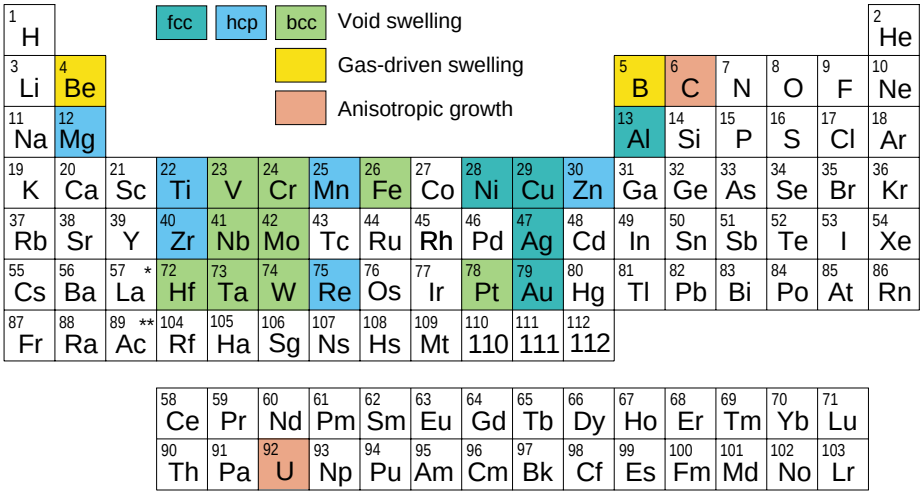

Figure 1. Elements Known to Undergo Radiation-Induced Dimensional Changes

All of the elements highlighted undergo radiation-induced volume changes due to void swelling, helium bubble (gas-driven) swelling, or anisotropic growth. The elements without highlighting have not yet been investigated for radiation-induced swelling or growth. Elements that exhibit void swelling are also identified by their crystal structure: face-centered cubic (fcc), hexagonal close-packed (hcp), or body-centered cubic (bcc).

Our models also predict macroscopic effects, such as the length of time before swelling begins and the steady-state, or linear, growth of swelling that eventually occurs. The time required for the onset of swelling is the most difficult to predict because it depends on detailed properties of the host metal, including its microstructure, its composition, and the presence of impurities. (For discussions of microstructural defects and dislocations, see the articles “Plutonium and Its Alloys” on page 290 and “Mechanical Behavior of Plutonium and Its Alloys” on page 336.)

There are a number of pressing questions concerning radiation effects in plutonium. For example, how long will a plutonium pit retain its size, shape, strength, and ductility given the self-irradiation damage that occurs throughout its volume? Will the material undergo a phase change from the δ phase to the denser, more-brittle α phase? Will the accumulation of defects and dislocations cause it to become weaker and more susceptible to fracture? We would like to apply our models to plutonium to answer those questions, but unfortunately, there are scant ambient-temperature experimental results and a substantial lack of data on the structure and properties of radiation-produced defects in plutonium.

We have made a preliminary assessment of radiation damage in gallium-stabilized δ-phase plutonium by assuming it is a member of the family of “normal” face-centered-cubic (fcc) metals. That is, we assume it has lattice defect properties consistent with those of other fcc metals such as nickel, copper, and austenitic stainless steels. We then use our models to predict the likely course of radiation damage.

This analysis identifies void swelling as the most likely effect of radiation damage. It also identifies specific defect properties and parameters that need to be obtained for a more realistic assessment. Furthermore, this analysis has guided us to plan and design experiments using plutonium that is enriched in the short-lived isotope plutonium-

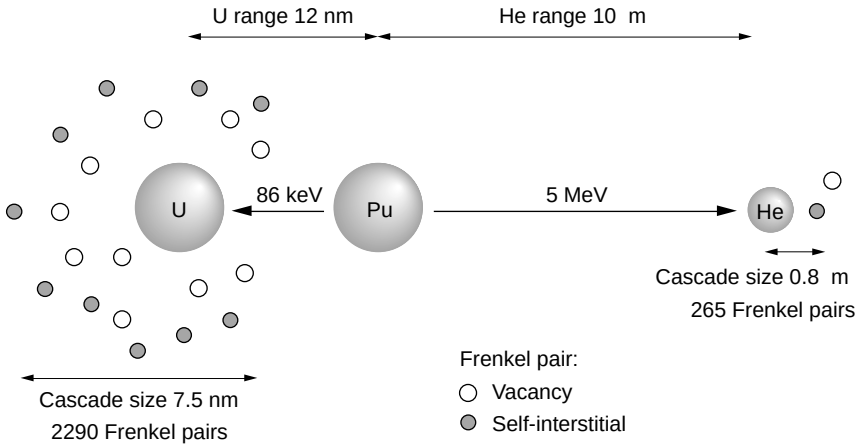

Figure 2. Plutonium Decay and the Generation of Defects

Plutonium decays to uranium by α emission. The α particle (a helium nucleus) takes away most of the energy and has a range of about 10 μm through the plutonium crystal. The heavy uranium nucleus recoils as a result of the α emission. It receives far less energy, and its range is only about 12 nm. Both particles produce displacement damage in the form of Frenkel pairs, namely vacancies and interstitial atoms, predominantly at the end of their ranges. Most of the damage results from the uranium nucleus and is confined to the collision cascade region of the size indicated.

238. Because the α-decay rate of plutonium-238 is nearly 300 times faster than that of plutonium-239, it is possible to accumulate helium nuclei and radiation damage within a sample at an accelerated rate. This acceleration of the processes will help us to obtain data on plutonium aging in just a few years.

Primary Radiation Effects in Plutonium

Radiation damage accumulates in metals after a complicated sequence of events that evolve over various time and distance scales. Figure 2 gives a schematic overview of the initial processes as they occur in plutonium. The plutonium nucleus decays to a uranium nucleus and a helium nucleus. Those daughter nuclei fly off through the lattice in opposite directions. Along their paths, and particularly toward the end of their range, they initiate a collision cascade wherein they transfer energy and momentum to the electrons and atoms of the material. Plutonium atoms can become displaced, or knocked from their lattice positions, thus creating numerous vacancies. A displaced plutonium atom will eventually come to rest at an interstitial site (between the normal lattice sites), becoming a "self-interstitial." Each displacement therefore creates a so-called Frenkel pair consisting of a vacancy and a self-interstitial, and each decay event creates many Frenkel pairs.

Many of the processes involved in this sequence have not been studied in plutonium, but the initial accumulation of helium atoms and Frenkel pairs can be estimated fairly reliably. Those estimates change depending on the plutonium isotope.

A typical isotopic composition of weapon-grade material is listed in Table I, which among other things lists the contribution of each isotope to the rate of helium generation per year. The cumulative rate of helium production is moderate—about 41.1 atomic parts per million (appm) per year. For example, during 10 years, 411 helium atoms will accumulate for every million plutonium atoms, making a helium concentration of about 0.04 atomic percent.

The last column in Table I lists our estimates for the rate at which the decay products displace plutonium atoms from their normal lattice sites, in units of displacements per atom (dpa) per year. To obtain the rate, we first estimate the number of displacements per decay event (normalized to the number of plutonium atoms in the material), and then divide by the number of decay events per year. The latter is obtained from the half-life of plutonium.

The Linhard-Scharff-Schiott theory of energy dissipation, combined with a procedure outlined by Robinson (1994), enables us to evaluate the number of displaced atoms per plutonium decay. Table II summarizes this evaluation for the isotope plutonium-239. In principle, both the helium and uranium decay products can damage the crystal lattice through collisions with plutonium atoms and with the bound and free electrons. However, the highly energetic helium nucleus loses all but 0.1 percent of its 5-million-electron-volt (MeV) energy through collisions with electrons. Only when it moves with low energy at the end of its range will it produce displacements. In contrast, nearly 75 percent of the uranium nucleus' 85 kilo-electron-volt (keV) kinetic energy results in the displacement of plutonium atoms.

To calculate the number of displacements, we need to know the so-called displacement energy, E_d, which is the minimum kinetic energy an atom must receive to be dislocated from its stable lattice site. The E_d for plutonium has not yet been measured, but an empirical rule provides the basis for a reasonable estimate:

$$E_d \cong 175\, k_B T_m \ , \qquad (1)$$

where k_B is the Boltzmann constant and T_m is the melting point of the metal. Using the melting temperature of plutonium, T_m = 913 K, gives the value $E_d \cong 14$ electron volts. The recoil nucleus needs to impart at least this

Table II. Radioactive Decay Characteristics of Plutonium Isotopes

Isotope Mass	Half-life (yr)	Abundance (at. %)	Decay Energy (MeV)	He Energy (MeV)	Recoil Energy (keV)	Helium Rate (appm/yr)*	Displacement (dpa/yr)
238	86.4	0.02	5.50	5.39	92.2	1.6	0.0043
239	24,390	93.6	5.15	5.04	85.8	26.6	0.0679
240	6580	5.9	5.16	5.07	85.9	6.2	0.0159
241	14.98	0.44	4.89	4.84	82.0	6.7	0.0152
242	388,000	0.04	4.90	4.85	81.9	0	0
					Total:	41.1	0.1033

*Determined by the half-life.

Table II. Electronic and Nuclear Energy Losses and Displacements for the Decay Products of ^{239}Pu

Product	Kinetic Energy (keV)	Fraction of energy Lost to Electrons	Available Energy (keV)	Number of Displacements
^{4}He	5040	0.9985	7.56	265
^{235}U	85.8	0.253	64.1	2290
		Total number of displacements per decay:		2555

amount of energy to dislodge a plutonium atom. That occurs in about half of all collisions, since often the two nuclei only graze each other (Robinson 1994). Dividing the available collision energy by approximately $2 \times E_d$ yields the total number of displaced plutonium atoms per plutonium decay, which is given in Table II as 2555.

Referring back to Table I, the displacement rate is then calculated to be about 0.068 dpa per year for plutonium-239, leading to a total rate of 0.1033 dpa per year for all isotopes in weapon-grade material. Each atom in the crystal lattice of plutonium is displaced from its stable site once every decade. During this same period, about 400 appm of helium is produced and retained within the material.

To summarize, the immediate result of self-irradiation in plutonium is the buildup of helium and uranium decay products, and the accumulation of vacancy and self-interstitial lattice defects. These primary defects, which are on the atomic scale, eventually diffuse and create a new microstructure of defect clusters, which ultimately control the macroscopic properties (Kiritani 1994). In the sections that follow, we estimate the ultimate effects of these initial atomic-scale defects.

Helium Bubble Formation

Following a radioactive decay event in a host material, the energetic helium nucleus races through the metal lattice and loses energy through collisions with electrons. It captures two electrons and eventually comes to rest as a helium atom.

What is the likely fate of the helium that accumulates in plutonium? Helium has an extremely low solubility in condensed matter because it does not bind. If the helium atom comes to rest at an interstitial site, it can easily diffuse through the lattice until it becomes trapped in one of the vacancies it created as it came to rest. The helium atom is only about half the size of a metal atom, however, so the vacancy it occupies is not eliminated.

Helium diffusion, both by association with vacancies and by dissociation from vacancies, has been studied in several metals, particularly in nickel (Adams and Wolfer 1988). These studies suggest that short-range diffusion of helium will certainly occur in plutonium at ambient temperatures and that the helium mobility is on the order of the vacancy mobility. The helium atoms/vacancies will likely cluster together, and these clusters can act as precursors to helium bubbles. We suggest therefore that helium bubble formation will take place in plutonium.

But how fast and to what extent?

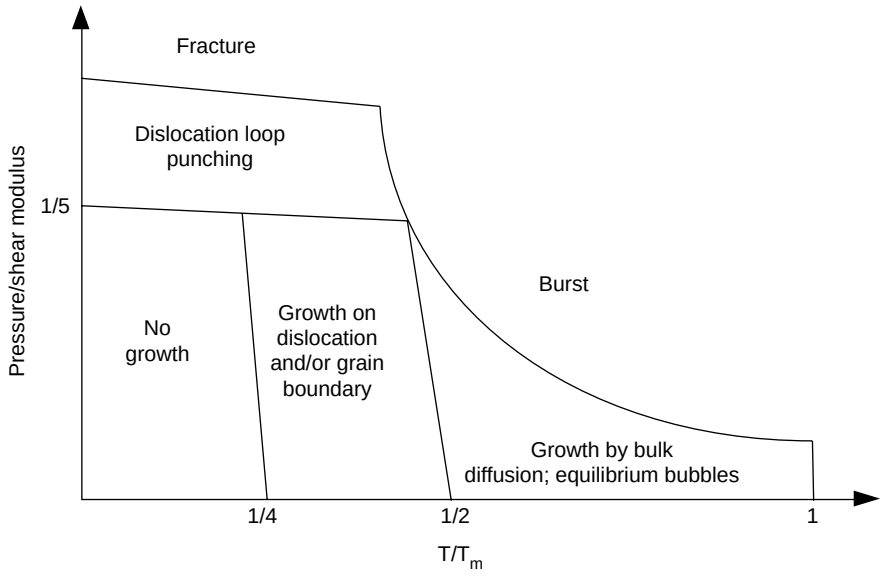

Figure 3. Helium Bubble "Growth Map" for Metals

The operative mechanism depends on the temperature of the material relative to the melting temperature, T/T_m (the homologous temperature). The helium pressure required to activate the growth depends on the mechanism and on the metal. However, the dependence on the metal is eliminated when the pressure for growth is scaled with the shear modulus. At low temperatures, bubble growth occurs by dislocation loop punching when the helium pressure exceeds one-fifth of the shear modulus. At temperatures at or above one-half the melting point, bubble pressure remains close to the value determined by the surface tension of the host material, and growth occurs by the absorption of thermal vacancies. In the intermediate temperature range, growth can also take place for bubbles on dislocations and grain boundaries.

In the absence of direct studies on plutonium, we turn to models we have developed from studies of other materials.

Helium bubble formation and growth in the absence of radiation damage can be studied in metal tritides, that is, metal–tritium compounds. In this case, helium accumulates in the material from tritium decay, and the recoil energy from the decay is so small that no displacements are produced.

Several mechanisms for bubble growth, including bulk diffusion and the formation of dislocation loops, were identified in those studies. We found that bubble growth depends on the temperature of the metal relative to its melting temperature (the homologous temperature) and on the helium pressure. Usually, one growth mechanism dominated over the others within a given temperature/pressure regime, as seen in the schematic bubble-"growth map" of Figure 3. We also developed quantitative models for the various growth mechanisms and compared our predictions with the experimental swelling results for metal tritides. We can apply those models to derive lower and upper bounds on helium-induced swelling in plutonium.

The lower bound is given by the dislocation-loop-punching mechanism, which requires no thermal activation but a very high helium pressure inside the bubble. The studies on metal tritides show that helium bubbles produced by this mechanism have a density of roughly two helium atoms per host atom, that is, every two helium atoms take up the volume, V, of one vacancy. Therefore, the minimum volume expansion induced by the formation of the helium bubbles is given by (Wolfer 1989)

$$\left(\frac{\Delta V}{V}\right)_{\min} \cong 0.5\,[\mathrm{He}]\ , \qquad (2)$$

where the helium concentration [He] is given in atomic parts per million. This model predicts that as helium accumulates, the swelling will increase linearly in proportion to the helium concentration.

To develop an upper bound for helium-induced swelling, we consider the growth of equilibrium bubbles by bulk diffusion, which occurs when the temperature is greater than or equal to one-half the melting temperature. Bubbles are in equilibrium when the helium pressure inside the bubble equals the surface tension of the host material, that is,

$$p = \frac{2\gamma}{r}\ , \qquad (3)$$

where p is the helium pressure, γ is the surface energy of the host metal, and r is the bubble radius. If the density of bubbles per unit volume is N_B and we assume an ideal gas law, then the helium-induced swelling is given by

$$\left(\frac{\Delta V}{V}\right)_{\max} = N_B\left(\frac{4\pi}{3}\right)r^3 = \sqrt{\frac{3}{4\pi N_B}}\left(\frac{[\mathrm{He}]k_B T}{2\gamma}\right)^{\frac{3}{2}}\ . \qquad (4)$$

We can evaluate helium-induced swelling from equilibrium bubbles using Equation (4), but first we need to know the bubble density, as well as the surface energy in the host material. The latter can be estimated from the value for liquid plutonium at the melting point, γ_{lm} = 0.55 joule per square meter, which is the only value of the surface energy for plutonium reported in the literature. We use an empirical formula given by Murr (1975),

$$\gamma \cong 1.2\gamma_{lm} + 0.45\left(T_m - T\right)\ . \qquad (5)$$

The bubble density N_B depends mainly on the temperature, or more precisely on the helium mobility. At

temperatures at which vacancy migration is possible, small helium–vacancy clusters form at very low helium concentrations of about 10 appm or less. After nucleation, the existing clusters or bubbles capture any newly generated helium atoms. Thus the bubbles grow.

At one-half the melting point of a metal with helium concentrations of 100 to 1000 appm, the observed bubble densities range from 10^{12} to 10^{14} bubbles per cubic centimeter (bubbles/cm^3). The density, however, increases sharply with decreasing temperature as the mobility of vacancies declines. At one-fourth the melting point, bubble densities increase by about four orders of magnitude and range from 10^{16} to 10^{18} bubbles/cm^3. Noting that lower bubble densities give higher bubble swelling according to Equation (4), we conservatively estimate bubble densities in plutonium to be about 2×10^{14} bubbles/cm^3 at an ambient temperature of 70°C. (See Figure 4.)

We are now in a position to compute the swelling caused by helium bubbles using Equations (3) and (4) for a lower and upper estimate, respectively. The results are shown in Figure 5 for a temperature of 70°C. Helium bubble swelling for lower aging temperatures falls within the range limited by the upper and lower bounds. Even though these estimates are crude, the volume changes caused by helium bubbles alone are small. Even after 60 years, the swelling is substantially less than 1 percent. It is therefore important to look into other mechanisms for density changes, such as void swelling.

Radiation-Induced Void Swelling in Metals

Void swelling is a byproduct of processes that occur during the recovery of the damaged crystal lattice. We have already mentioned that a decay event causes the heavy daughter nucleus to recoil through the material, and the subsequent displacement damage creates thousands of Frenkel pairs consisting of vacancies and self-interstitials. In fcc materials, the self-interstitials are mobile even at cryogenic temperatures and, during their random migration, become absorbed at dislocations and grain boundaries. But the self-interstitials will also recombine with vacancies, thus annihilating a Frenkel pair and restoring the perfect crystal structure. About 100 picoseconds after the decay of a plutonium atom, up to 70 percent of the Frenkel pairs have recombined.

As seen in Figure 6, the remaining defects have rearranged in the form of defect clusters, mainly small dislocation loops. A few isolated vacancies also remain. The vacancy clusters eventually lead to the nucleation and subsequent growth of voids.

The macroscopic swelling of the metal that is due to the growth of voids—void swelling—has been studied extensively. It became clear that the phenomenon occurs in all metals within a well-defined temperature range and as the result of any irradiation process that produces displacement damage,

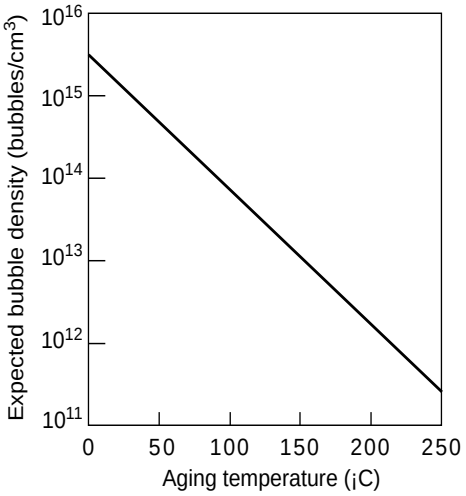

Figure 4. Estimate of the Helium Bubble Density

The helium bubble density is needed to predict an upper limit to the extent of helium swelling. The curve shows the expected density for plutonium as a function of temperature. At 70°C, the helium bubble density is approximately 2×10^{14} bubbles/cm^3.

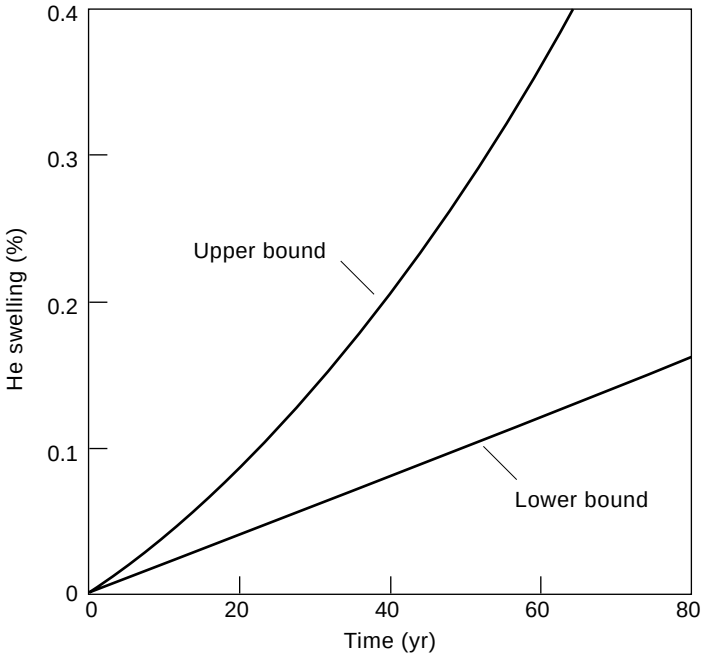

Figure 5. Helium Bubble Swelling in Plutonium at 70°C

The lower bound assumes the loop-punching mechanism for bubble growth; the upper bound, the mechanism of equilibrium bubble growth. At 70°C, helium bubble swelling is expected to fall between the two bounds.

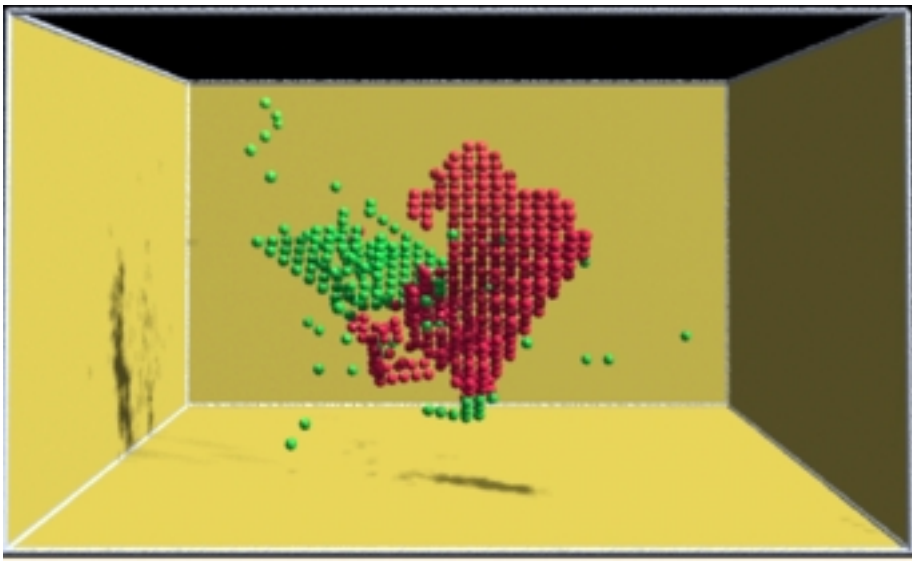

Figure 6. Cluster Formation in Lead

This output from a simulation shows the final damage in one part of a collision cascade in lead. Red balls are interstitial atoms and green balls are missing atoms, or vacancies. In this frame, 183 ps after one atom received a recoil energy of 80 keV, two large clusters have formed, one made of dislocation loops, or planar arrangements of interstitials (red cluster), and the other made of vacancies (green cluster). Note that very few of the vacancies or interstitials are not in clusters.

(Unpublished results by M. J. Caturla and T. Diaz de la Rubia, Lawrence Livermore National Laboratory, January 2000.)

including electron irradiation. (Electron irradiation does not generate collision cascades, but only isolated Frenkel pairs.)

Given the general occurrence of void swelling, there is no reason to believe that it will not happen in plutonium. In order to assess the temperature range and the magnitude of the swelling for plutonium, it will be useful to review the fundamental aspects of this phenomenon.

Void formation requires favorable conditions for nucleation and subsequent growth. Vacancies must be mobile, and a minimum amount of helium has to have accumulated. The minimum temperature necessary to initiate vacancy mobility determines the lower temperature limit for void formation. The upper temperature limit results from a supersaturation of the irradiation-induced vacancy concentration. As opposed to thermally induced vacancies, the clusters formed from the collision cascade are not thermodynamically stable. They slowly dissolve over time. Once the concentration of thermal vacancies exceeds the irradiation-induced vacancy concentration, void swelling ceases. Both temperature limits are weakly (that is, logarithmically) dependent on the displacement rate.

Table III contains a list of parameters that characterize different radiation environments where void swelling has been observed. The estimates for plutonium (last column) were obtained from scaling relationships that were derived from the theoretical models of void swelling.

The two irradiation methods, accelerators and reactors, often lead to very similar results, but there are differences. Accelerators implant metal ions (self-ion implantation) or helium ions (helium implantation) into the samples. The displacement damage occurs in a very thin (a few micrometers) layer, close to the surface and confined by the surrounded material. Reactors irradiate the sample with neutrons and the damage is uniformly spread over much larger dimensions. As a result, the two methods produce a different ratio for the void and dislocation densities, resulting in a different swelling rate. Neutron irradiation more closely resembles the self-irradiation damage in plutonium.

A typical swelling curve, derived from neutron irradiation studies (Wolfer 1984), is shown in Figure 7. There are three stages: the first stage is an incubation period during which void nucleation takes place; the second stage is a transient period during which an optimum ratio is established for the densities of voids and dislocations; and the third stage is a steady-state swelling period, which will eventually terminate at a saturation swelling value. The saturation value is usually too large to be of any practical concern.

Although these general features are observed in many metals and alloys (Garner 1994), large variations exist, particularly in the length of the transient period, which can range from a few to 100 dpa. Metals of high purity and fcc structure swell almost immediately. In well-annealed materials with an initially low dislocation density, large grain size, and low precipitate density, swelling commences immediately after the incubation period. The latter has been found to be roughly the time needed to accumulate 5 to 10 appm of helium (Wiedersich and Hall 1977) from nuclear transmutations. However, oxygen impurities in excess of the solubility limit can also shorten the incubation time (Zinkle et al. 1987).

Cold-worked materials with initially high dislocation densities have, in general, long transient periods. However, the initial dislocation density does not remain constant. It evolves during irradiation and reaches a new steady-state density determined by the irradiation conditions (Wolfer and Glasgow 1985). When this steady-state density is reached, void swelling can proceed at its steady-state rate.

Table III. Comparison of Radiation Environments

Irradiation Parameter	Accelerator		Breeder Reactor	Thermal Reactor	Plutonium
	Self-Ion Implantation	Helium Implantation			
Dose rate (dpa/sec)	10^{-3}	10^{-3}	10^{-6}	10^{-7}	10^{-9}
He accumulation rate (appm/dpa)	sequential[a]	1000	1	15	400
Homologous temperature range	0.45–0.65	0.45–0.65	0.35–0.55	0.3–0.4	0.25–0.45

[a]He ions are implanted in the material prior to heavy ion bombardment.

A characteristic feature of this steady state is that parity exists between the dislocation density and the void density. The dislocations are in fact the primary cause of void swelling. They attract self-interstitials slightly more than vacancies and therefore possess a "bias" for preferential absorption of self-interstitials. In contrast, voids, which also absorb self-interstitials, are more neutral or less biased.

The difference in these biases, the net bias ΔB, is the fundamental driving force for void swelling. The maximum possible separation of vacancies and self-interstitials—as determined by net bias—is realized only when voids and dislocations are of equal abundance. This state is referred to as the state of sink parity. During the initial stages of void swelling, the dislocations are more abundant and the swelling rate is less than what it could be. For very large values of swelling, and hence void sizes, the voids are the most abundant sink, and the swelling rate diminishes again.

It has been shown by Garner and Wolfer (1984) that an upper bound to the void swelling rate can be derived when sink parity exists. As a result,

$$\frac{\mathrm{d}}{\mathrm{d}\tau}\left(\frac{\Delta V}{V}\right) \leq 1.25\,\Delta B\ , \qquad (6)$$

where τ is the time measured in units

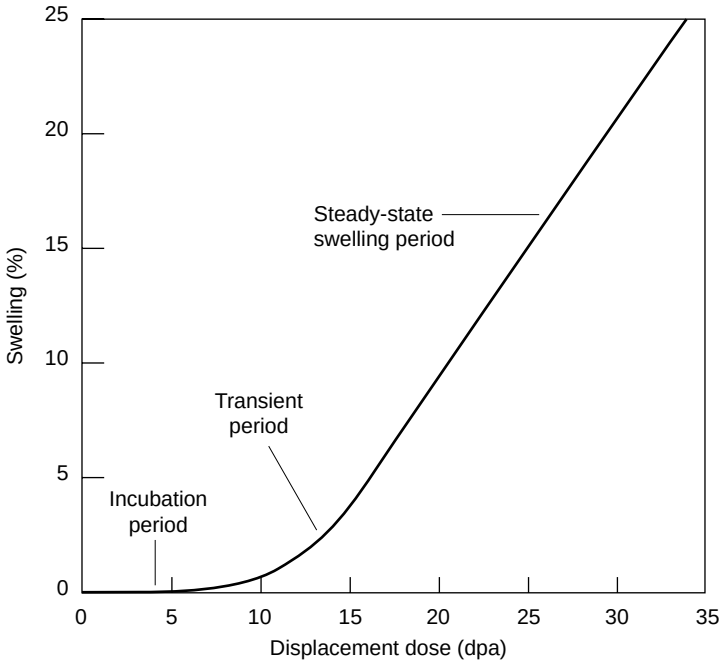

Figure 7. The Different Stages for Radiation-Induced Void Swelling
The fractional volume increase due to void swelling is plotted as a function of irradiation exposure time measured in displacements per atom. The incubation period is, in general, determined by the time required to accumulate about 5 to 10 appm of helium by nuclear transmutation reactions. The transient period can extend from a few to 100 dpa, depending on the crystal structure, alloy composition, initial dislocation density, precipitate evolution, and irradiation temperature. The steady-state rate is mainly determined by the fundamental properties of the crystal lattice and of the vacancy and self-interstitial.

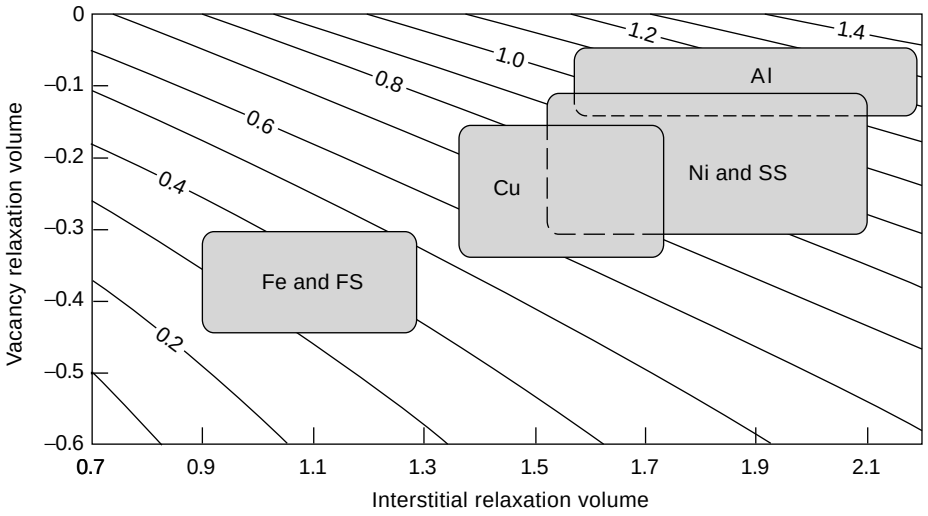

Figure 8. Net Bias for Radiation-Induced Void Swelling

Dislocations attract and absorb self-interstitials slightly more than vacancies. The different rate of absorption is known as the net bias, Δ*B*, and is the fundamental driving force for void swelling. The contour lines are lines of constant net bias value as indicated. The boxes circumscribe the range of predicted values for the net bias using measured relaxation volumes and their uncertainties. The materials are aluminum (Al), nickel and austenitic stainless steels (Ni and SS), copper and copper-nickel alloys (Cu), and body-centered-cubic iron and ferritic steels (Fe and FS).

Table IV. Swelling Rates of Some Metals and Alloys

Metal	Measured Steady-State Swelling Rate[a]	Estimated Upper Bound[a]
Al	1.0	1.1–1.7
Ni and SS	1.0	0.8–1.5
Cu	0.5	0.7–1.2
Fe and FS	0.2	0.35–0.6

[a]Measured in units of percent volume change per dpa, %/dpa

of dpa. (In a reactor, the neutron flux produces damage at a rate that is expressed in dpa/sec. For a constant flux, dpa is therefore proportional to the time.)

Sniegowski and Wolfer (1984) have developed a theory to derive the net bias from the fundamental properties of the crystal lattice and the defects. The two most influential parameters are the so-called relaxation volumes of the self-interstitial and of the vacancy, which can either be measured or determined from atomistic defect calculations. Figure 8 shows the theoretical results of Sniegowski and Wolfer for the net bias.

The comparison shown in Table IV of measured steady-state swelling rates with the estimates of the upper bounds indicates that both exhibit the same trends, that the steady-state swelling rate is close to the maximum possible, and that metals with the more open body-centered-cubic (bcc) structure swell less than fcc metals do.

The fcc phase of plutonium, δ-phase plutonium, exhibits some unusual properties, including a negative thermal expansion coefficient. This behavior could be indicative of a very soft interatomic repulsive force between plutonium atoms in the δ phase. If so, the relaxation volumes of both vacancies and interstitials would be low, resulting in a small net bias.

However, the addition of an alloying element such as gallium to stabilize the δ phase may change the net bias in two ways. First, the electronic band structure could be affected sufficiently to alter the interatomic forces and hence the relaxation volumes. Second, radiation-induced segregation of the alloying element can significantly alter the net bias (Wolfer 1983). If this segregation occurs, microscopic regions in the crystal could possibly become sufficiently depleted of the stabilizing element and be converted to the denser α phase. This would result in a negative volume change of $\Delta V/V = -0.14f$, where f is the volume fraction of the transformed material.

Void Swelling in δ-Stabilized Plutonium

Figure 9 compares the predictions for helium bubble swelling and void swelling in δ-phase plutonium. A reliable prediction of void swelling is not possible at the present time without knowledge of the relaxation volumes for vacancies and interstitials. A conservative assumption is to adopt the typical values for fcc metals, and hence, the steady-state swelling rate of 1 percent volume change per dpa. With regard to the incubation dose for void nucleation, the required amount of helium is available after about 4 months or a dose of about 0.025 dpa. The temperature range for swelling is expected to be from –30°C to 150°C.

There is a large uncertainty, howev-

er, in the length of the transient period. To obtain an accurate prediction, it would be necessary to perform extensive model calculations, both of the dislocation evolution and of the void evolution. Theoretical predictions, performed by Wolfer and Glasgow and by Wehner and Wolfer (1985), and experimentally observed transient periods seem to indicate that a minimum dose of at least 1 to 2 dpa is needed before steady-state swelling is approached. Based on these assumptions, void swelling in stabilized δ-phase plutonium could in principle begin as early as 10 years after pit fabrication. However, there is no evidence for void swelling even after 30 years; it is entirely possible that the transient period is much longer, and void swelling may not occur within the next 100 years.

Nevertheless, the swelling predictions shown in Figure 9 already provide an important result: Swelling due to displacement damage is potentially a more serious problem than swelling due to helium.

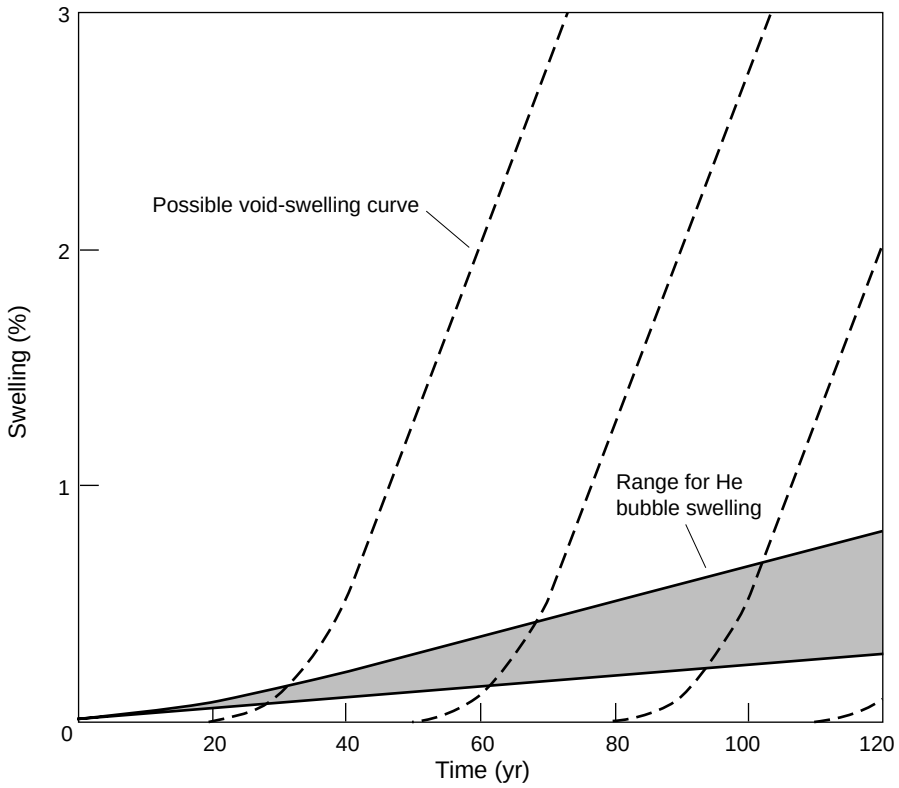

Figure 9. Predictions for Radiation-Induced Damage in Plutonium
The figure shows the predicted contributions to volume distortion in stabilized plutonium (aged at 70°C). Distortions due to void swelling are likely to be much larger than those due to helium-bubble formation. However, the large uncertainty in the transient period prevents us from estimating when the void swelling should begin its linear growth rate. The figure shows several possible swelling curves.

Radiation-Induced Creep and Stress Relaxation

Radiation-induced creep has been recognized as a phenomenon even before the discovery of void swelling. First reported as a deformation process accompanying fission in uranium fuel (Roberts and Cottrell 1956), it has been found to occur in graphite, ceramic nuclear fuel, steels, and zirconium alloys, as well as in glass. The primary cause is again the displacement damage and the diffusion of the self-interstitial and, at higher temperature, the diffusion of the radiation-produced vacancies. Radiation-induced creep is distinct from thermal creep in that it exhibits a rather weak temperature dependence and an (approximately) linear dependence on stress and displacement damage rate. It occurs below the homologous temperature, which defines the upper temperature limit for void swelling. In the case of plutonium, this means below a temperature of 150°C.

Radiation-induced creep will plastically deform a material and so is capable of relaxing stresses. It is therefore a beneficial mechanism in conjunction with void swelling because it limits the amount of stress that can be generated. For example, when swelling takes place in a thin layer of material attached to a rigid substrate, stresses build up in this layer according to the equation

$$\frac{1}{M}\frac{d\sigma}{dt} + \frac{d}{dt}\left(\frac{\Delta V}{V}\right) + \psi\sigma = 0 \ , \qquad (7)$$

where σ is the lateral stress component in the swelling layer, $M = E/[3(1 - \nu)]$ with E the Young's modulus and ν the Poisson ratio, and ψ is the compliance coefficient for radiation-induced creep. It is seen from Equation (7) that a saturation stress is reached that is given by

$$\sigma_{sat} = -\frac{1}{\psi}\frac{d}{dt}\left(\frac{\Delta V}{V}\right) \ . \qquad (8)$$

This saturation stress is found to be about half the initial yield strength of the annealed material. However, as mentioned above, secondary processes lead to an increase in the dislocation density of annealed materials, and their yield strength increases rapidly with the irradiation dose and reaches values two to three times higher after about 5 dpa. The increase in yield strength is accompanied by a reduction in ductility, and it is therefore all the more important to have an additional stress relaxation mechanism such as radiation-induced creep.

The prediction of the radiation-in-

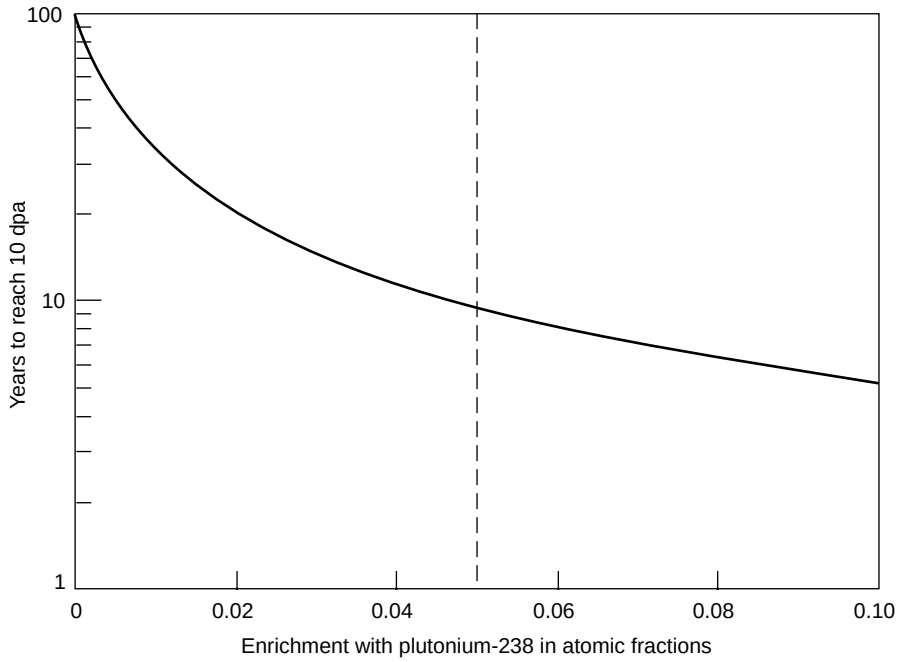

Figure 10. Accelerating Aging

By enriching a sample of weapon-grade plutonium with ^{238}Pu, the time to accumulate a radiation damage dose of 10 dpa can be significantly reduced. Without enrichment, it takes 100 years to acquire such a dose. Upping the ^{238}Pu fraction by as little as 5 percent will reduce the time to ten years. However, the effects of radiation dose rate on aging are not known. It is not clear how to translate the data from accelerated aging experiments to the study of aging in pits.

duced creep rate, or more precisely of the creep compliance ψ, is in principle possible, as demonstrated by Wolfer (1980) and by Matthews and Finnis (1988). However, in addition to the defect parameters and microstructural data required for void swelling predictions, other defect parameters are needed that are more difficult to determine experimentally or by computer simulations. These are the so-called elastic polarizabilities for both the vacancy and the self-interstitial.

Summary and Recommendations for Further Research

The analysis presented here of self-irradiation damage effects in plutonium reveals that both helium accumulation and displacement damage must be considered. Although helium accumulation alone is unlikely to pose a serious plutonium aging problem, the displacement damage effects can lead to significant dimensional changes as a result of the following two phenomena.

First, void swelling is expected to occur in the temperature range from –30°C to 150°C and may reach a rate of about 0.1 percent per year. However, the transient period, or the time it takes to reach this steady-state swelling rate, may be anywhere from 10 to 100 years after pit fabrication. The length of the transient period depends on many variables, including alloy composition, dislocation and grain structure, and the evolution of the latter two with time. It is unlikely that these metallurgical variables can be specified with sufficient accuracy to predict the transient period in δ-phase plutonium.

Second, radiation-induced phase transformation from δ to α in marginally stabilized plutonium alloys can result in significant shrinkage. (The α phase has a specific volume that is 20 percent less than the δ phase.) This phenomenon is closely coupled to the processes that occur in void swelling, and the two mechanisms for dimensional changes may in fact take place simultaneously. Experimental x-ray diffraction studies on phase compositions in aged plutonium can easily reveal the occurrence of this process.

The analysis presented in this article relies to a large extent on the assumption that δ-phase plutonium can be viewed as a typical fcc metal with regard to radiation and helium effects. Clearly, this assumption must be replaced with actual data on radiation-induced dimensional changes and with measurements and theoretical calculations of critical defect properties for stabilized δ-phase plutonium.

To accomplish these two goals, an accelerated-plutonium-aging research program has been initiated using material with a higher content of the isotope plutonium-238. This isotope decays at a much faster rate than plutonium-239 (refer to Table I), and by its addition, the rate of radiation damage can be accelerated.

For example, suppose one wants to study radiation effects up to a total dose of 10 dpa. For plutonium metal with an isotope composition as listed in Table I, it would take 100 years to accumulate such a dose. But as seen in Figure 10, that time is reduced to 10 years with a 5 percent enrichment of plutonium-238.

Accelerating the rate of radiation damage, however, raises several other issues. One is the dissipation of the thermal energy and the temperature control of the irradiation experiments. The other is the effect of dose rate on the evolution of the defect morphology, on void nucleation, and on the transient period. The void-swelling results from neutron and ion irradiations give a clear indication that the steady-state swelling rate is independent of the rate of damage production, but the incubation and transient periods do depend on dose rate.

It is therefore necessary to develop detailed models for all the important processes involved in the damage production, including the evolution of the microscopic defect structure and the nucleation and growth voids, together with the evolution of the dislocation structure. With these models in hand and verified with experimental data, we can translate the results obtained from the accelerated-aging experiments to the lower dose rates in stockpile materials. ■

Further Reading

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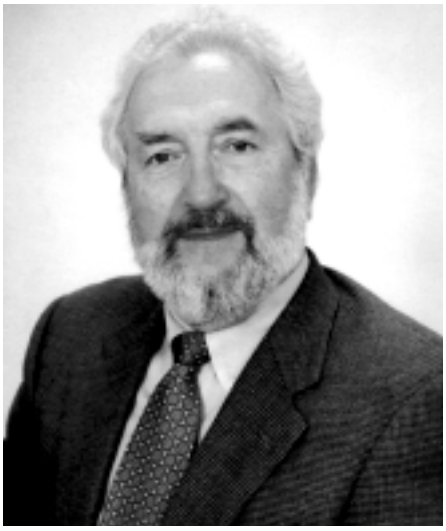

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