Plutonium: Aging Mechanisms and Weapon Pit Lifetime Assessment

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Planning for future refurbishment and manufacturing needs of the U.S. nuclear weapons complex critically depends on credible estimates for component lifetimes. One of the most important of these components is the pit, that portion of the weapon that contains the fissile element plutonium. The U.S. government has proposed construction of a new Modern Pit Facility, and a key variable in planning both the size and schedule for this facility is the minimum estimated lifetime for stockpile pits. This article describes the current understanding of aging effects in plutonium, provides a lifetime estimate range, and outlines in some detail methodology that will improve this estimate over the next few years.

INTRODUCTION

Systematic aging studies on pits for nuclear weapons were initiated a few years ago after the shutdown of the Rocky Flats manufacturing plant in Colorado. During the past 60 years, pit designs, materials, and processes have changed dramatically, resulting in pits that are more robust, safer, and suited for longer storage times. Modern pits consist of hollow, metallic shells containing fissile material at their core. The outer, non-nuclear materials used in pits are selected for properties such as mechanical robustness and integrity as well as corrosion resistance. These materials remain remarkably pristine over decades. Further, modern designs rely on the boost process—the introduction of deuterium/tritium mixtures into the interior—as an essential element of weapon function. Hence, the integrity of pits as gas-pressure vessels is another important element of weapon function. In this respect as well, the surveillance program has proven that pits are demonstrably robust over decades. Given this positive history with the non-nuclear materials in pits, most concerns with pit aging focus on the behavior and possible degradation of the plutonium.

At the end of this year, the U.S. Nuclear National Security Administration (NNSA) Enhanced Surveillance Campaign has a key deliverable to provide a pit lifetime assessment based on old pit data. In 2006, this assessment will be updated based on further collection and review of old pit data as well as new information expected from the accelerated aging program (described later in this article). Between now and 2006, further experiments, modeling, and design sensitivity calculations on different systems are required to reduce uncertainties regarding lifetime estimates.

EVALUATION OF THE AGING PROCESS

The approach used to address the aging of pits starts with an identification of the key plutonium properties required to ensure safe and reliable weapon function. These properties (such as density) are selected by knowledgeable design physicists and engineers who will ultimately use them in computer simulations as part of the certification process of a given weapon. This process is quite complicated because for years designers relied largely on testing the devices at the Nevada Test Site to assess performance. Although a substantial amount of work was done to relate performance to specific materials properties, a better understanding is needed as to how key properties influence weapons performance using advanced tools such as improved codes. Once these properties have been identified, diagnostic tools can be developed to measure them with sufficient precision as determined by the weapon designer. An important aspect of the aging program is the execution of experiments to measure baseline properties of new (zero-aged) material.[1]

Next, materials scientists and chemists identify the aging mechanisms that could potentially alter these properties over time. The three most important potential aging effects in plutonium are the radioactive decay of the various plutonium isotopes (and the impact of this decay on the chemistry, structure, and properties of the material), the thermodynamic phase stability of the plutonium alloy itself, and the corrosion of the plutonium during both storage and function. In many cases, these aging effects accumulate slowly over decades and not necessarily in a linear fashion. Only when key properties have sufficiently changed would a measurable impact on weapon safety or performance be expected. Through the experiments, modeling of the age-related changes, and design sensitivity studies, the designers attempt to specify the limits of acceptable change for each of these properties by evaluation of the margins associated with each system. By combining these limits with the measured or predicted rates of change due to aging effects, estimates for pit lifetimes are derived.

Each of the three principal aging mechanisms identified is under intensive examination within the Enhanced Surveillance Campaign. This program has four key elements/objectives: measurement of actual properties and trends from the newest to the oldest

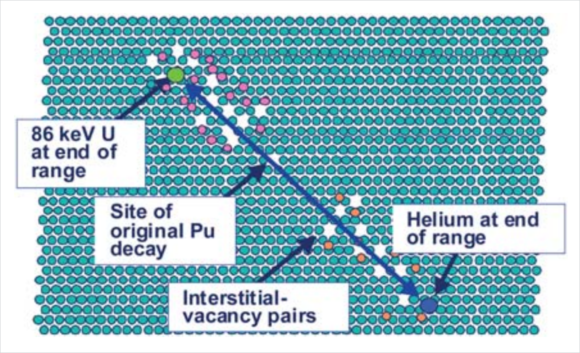

Figure 1. An illustration of alpha decay and recoil-particle damage in δ-Pu.[2]

materials available from the stockpile; acceleration of the aging where possible and subsequent measurement of material properties; modeling of aging effects for insertion into design sensitivity analyses; and the development of new diagnostics to identify the signatures of aging as early as possible in order to provide lead time for refurbishment. In parallel, the NNSA Primary Certification Campaign and the Accelerated-Strategic Computing program are developing the computational tools required to address design sensitivity, acquiring the test data (e.g., sub-critical experiments) to quantify key parameters, and developing the expertise to complete the design-sensitivity assessment.

DAMAGE MECHANISMS AND APPLICABILITY TO EVALUATION OF OLD PITS

The oldest plutonium made in the United States and available for analysis is approximately 40 years of age. This plutonium was manufactured by processes somewhat different from the materials in the enduring stockpile. As a result, a direct comparison of this oldest plutonium to modern alloys may invoke uncertainty, but has provided substantial insight into the aging behavior. Extensive, but incomplete evaluations of this material over the past three years have shown only modest changes in key properties. Nonetheless, these small changes are invaluable in helping to calibrate and refine existing aging models. This oldest plutonium has been crucial in another respect: It exhibits no void-swelling, one of the potentially most troublesome manifestations of self-irradiation damage.

A fundamental aspect in the accumulation of radiation damage in materials is the existence of a threshold beyond which further damage results in rapid swelling and density change. Experience from all materials in reactor environments of similar crystal structure (face-centered cubic [fcc]) to the plutonium alloys in the stockpile shows that the damage results initially in little change in density, but after an incubation period, void swelling begins. The length of this incubation is unknown for weapon-grade plutonium and cannot be predicted.

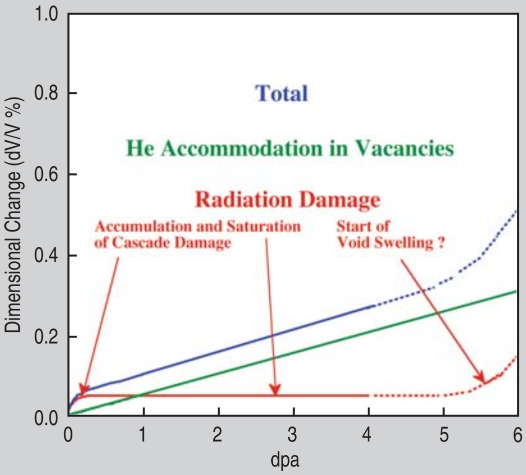

Figure 3. An illustration of combined effects from radiation damage on potential density change in plutonium. Note the incubation period associated with void swelling. The duration of the period is currently unknown for plutonium metal.[2]

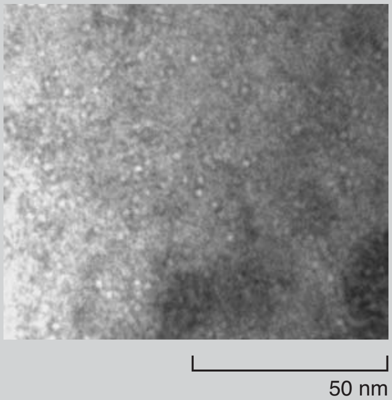

Figure 2. A bright-field transmission electron micrograph of helium bubbles in 35 year old plutonium alloy. The image was taken using the Fresnel fringe technique at –1.2 mm defocus. The bubbles appear as a dark fringe surrounding a light dot.

The principal decay mechanism for most plutonium isotopes is alpha-particle decay. The parent atom spontaneously decays into a doubly charged helium nucleus (i.e., alpha particle) and a uranium atom (Figure 1). Both of these particles are highly energetic. This initial decay event is rapid and results in considerable local disruption of the crystalline lattice. Based on theoretical considerations, this single decay energizes roughly 20,000 other atoms and displaces approximately 2,400 atoms from their lattice sites. Within the first 200 nanoseconds, about 90% of these displaced atoms return to a normal lattice position. The remaining 10% of these atoms are retained in the lattice, where an atom now sits between regular positions on the lattice (known as an interstitial) and the regular lattice positions are empty (known as a vacancy).[2] The ultimate disposition of these more permanent defects is the principal concern in this evaluation. This accumulation of damage is significant within the time frames of interest: On average, each atom of plutonium has been displaced once every ten years. In addition to the generation of alpha particles that ultimately lead to the formation of helium atoms and helium bubbles, an atom of uranium is also created in each radioactive decay event. Hence, an aging plutonium material becomes enriched in uranium and also americum, which may have implications for long-term phase stability.

Positron annihilation data indicate that the newly formed helium atom immediately fills an unfilled vacancy.