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Radiation and Thermal Ageing of Nuclear Waste Glass

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Abstract

The radioactive decay of fission products and actinides incorporated into nuclear waste glass leads to self-heating and self-radiation effects that may affect the stability, structure and performance of the glass in a closed system. Short-lived fission products cause significant self-heating for the first 600 years. Alpha decay of the actinides leads to self-radiation damage that can be significant after a few hundred years, and over the long time periods of geologic disposal, the accumulation of helium and radiation damage from alpha decay may lead to swelling, microstructural evolution and changes in mechanical properties. Four decades of research on the behavior of nuclear waste glass are reviewed.

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Keywords: radiation effects; helium behavior; alpha decay; beta decay

1. Introduction

The radioactive decay of incorporated fission products and actinides in nuclear waste glass leads to self-radiation effects and self-heating that may affect the stability, structure and performance of the glass in a closed system [Weber et al. 1997, Weber et al. 2009]. The principal heat-generating radionuclides in glasses for high-level waste (HLW) are the short-lived fission products, ^{90}Sr and ^{137}Cs, and if incorporated in glass waste forms, these radionuclides cause significant self-heating for about the first 600 years [Weber et al. 2009]. Over the long time periods of deep geologic storage, self-radiation of nuclear waste glass is primarily due to the alpha decay of the

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actinides. In the present paper, the effects of thermal history and self-radiation effects on the evolution of structure in nuclear waste glasses are reviewed, with emphasis on glasses for the immobilization of HLW from the reprocessing of commercial nuclear fuel and glasses for the immobilization of plutonium (primarily ^{239}Pu) or minor actinides (primarily Np, Am, and Cm) separated during reprocessing or recovered from the production of nuclear weapons.

In nuclear waste glasses, ionization from gamma rays and beta particles emitted in beta decay of fission products can cause covalent and ionic bond rupture, valence changes, localized electronic excitations, and significant changes in ionic mobility. At elevated temperatures during the initial thermal period, the high ionization rates may cause decomposition, phase separation, and bubble formation [Weber et al. 1997, Weber et al. 2009]. The effects of ionization have been largely studied using electron beams or gamma sources [Weber et al. 2009], and the relevant data and implications from these studies are discussed. The effects of self-radiation due to alpha decay on the structure and properties of glasses have been studied over the past four decades using samples containing short-lived actinides, primarily ^{238}Pu or ^{244}Cm [Weber and Roberts 1983, Muller and Weber 2001, Weber et al. 1997]. Ion-beam irradiation has also been used to explore radiation effects from alpha decay. A wide range of experimental methods has been employed to characterize the changes in density, stored energy, local structure, bonding, and valence states as a function of radiation dose and temperature. Over very long time periods, helium accumulation from alpha decay may lead to the formation of helium bubbles that may cause additional swelling and changes in mechanical properties. The current state of knowledge on such self-radiation effects will be discussed.

2. Thermal Ageing

2.1. Radiation Heat Sources

The primary source of heat nuclear waste glass is from the electronic energy deposition or ionization from gamma rays, beta particles, and alpha particles. In high-level waste (HLW) glass, the principal heat-generation is from beta decay of the short-lived fission products, ^{90}Sr and ^{137}Cs, which cause significant self-heating for the first 600 years [Weber et al. 2009]. However, in nuclear glasses intended to immobilize plutonium or minor actinides, heat generation is primarily from the high-energy alpha particles emitted by alpha decay of the actinides. The ionization dose as a function of time is shown in Fig. 1 for HLW glass (25 wt% waste loading) [Weber and Roberts 1983] and glasses containing either plutonium (^{239}Pu) or minor actinides.

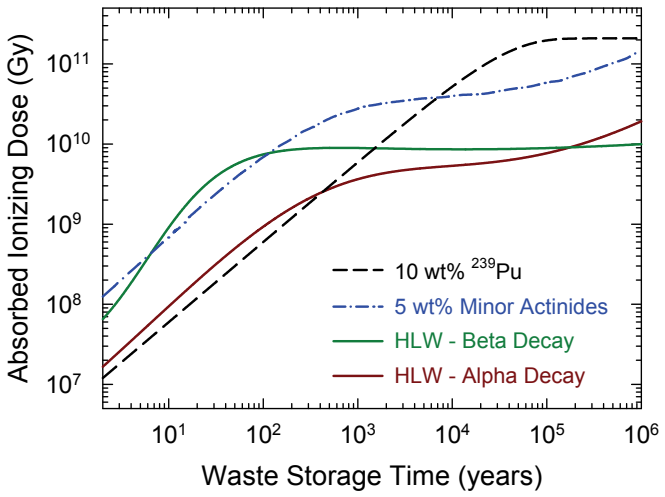

Fig. 1. Ionization dose from beta and alpha decay in nuclear waste glasses.

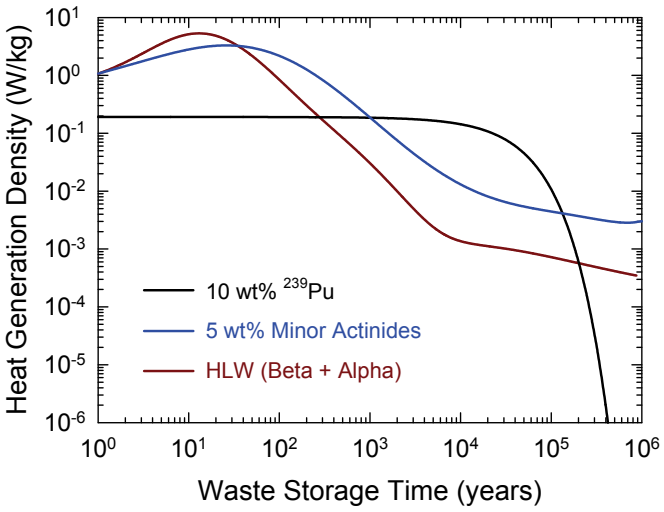

Fig. 2. Heat generation from beta and alpha decay in nuclear waste glasses.

2.2. Heat Generation and Temperature

The heat generated in nuclear waste glasses from the high ionization dose rates is shown in Fig. 2. The peak in heat generation for HLW and minor actinides is due to the ingrowth of ^{241}Am from beta decay of ^{241}Pu at short times. The expected centerline temperature and temperature at the outside diameter for a canister (32 cm diameter) containing HLW glass are shown in Fig. 3. In general, the current generations of nuclear waste glass compositions for HLW are formulated to minimize the precipitation of crystalline oxide phases during glass processing and from thermal ageing during interim storage periods (Fig. 3). Likewise, there has been considerable work over the past 20 years on specialized glass formulations that can immobilize higher concentrations of plutonium and minor actinides [Muller and Weber 2001]. One driver, particularly for minimizing the precipitation of crystalline oxide phases that

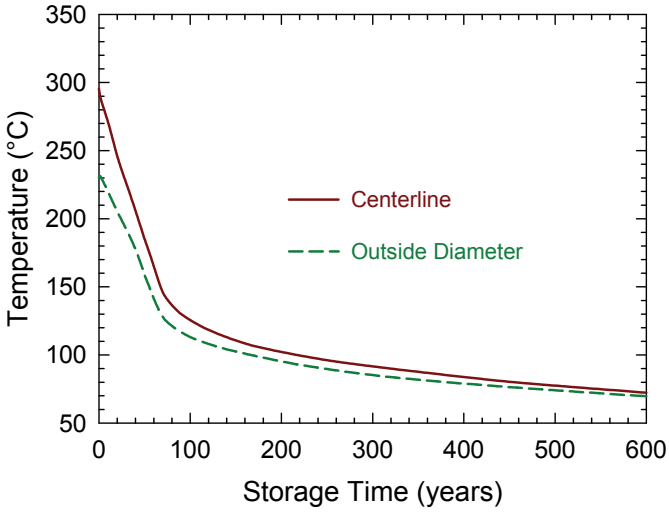

Fig. 3. Temperature history for HLW glass contained in a 0.32 m diameter canister.

contain actinides, is that it was shown early on [Weber and Roberts 1983] that radiation damage from alpha decay of actinides may lead to amorphization and significant swelling of such crystalline phases, which can result in microfractures. Thus, as a result of advances in glass formulations over the past 30 years or more, thermal ageing during interim storage times (Fig. 3) is not expected to cause significant changes in glass structure, and during this thermal period, radiation effects, such as stored energy [Weber and Roberts 1983], might be somewhat reduced.

3. Radiation Effects

Self-radiation from the decay of the radionuclides incorporated in nuclear waste glass can affect microstructural evolution, phase stability, and thermodynamic properties. The primary sources of radiation effects in nuclear waste glasses are beta-decay of the fission products (e.g., ^{137}Cs and ^{90}Sr) and alpha-decay of the actinide elements (e.g., U, Np, Pu, Am and Cm). Both fission products and actinides are present in current generations of nuclear waste glasses. In future advanced nuclear fuel cycles, fission products and actinides may be immobilized separately in glass or ceramic waste forms. Likewise, existing inventories of excess plutonium could be immobilized in specially tailored glasses. Beta-decay produces energetic beta-particles (~0.5 MeV), low energy recoil nuclei, and gamma-rays; alpha-decay produces energetic alpha-particles (4.5 to 5.5 MeV), energetic recoil nuclei (70 to 100 keV), and some gamma-rays. These decay particles and gamma-rays interact with glasses primarily through energy transfers to electrons via ionization processes, producing electron-hole pairs, or to atomic nuclei via ballistic collisions, displacing atomic nuclei in the glass network. In addition to energy transfer, the transmuted recoil nuclei emitted in radioactive decay must be readily accommodated in complex glasses. Helium atoms, formed from the capture of two electrons by the alpha particles, are also produced in actinide-bearing waste forms and must be accommodated.

3.1. Beta decay effects

Beta decay in HLW waste glasses produces very few direct atomic displacements from ballistic collisions of beta particles with atomic nuclei, and atomic displacements from gamma radiation are considered negligible. It has been estimated that beta decay produces less than one atomic displacement per decay event [Weber and Roberts 1983], resulting in atomic displacement rates in HLW glass that are two to three orders of magnitude less than that due to alpha decay. The cumulative ionization dose from beta decay in HLW glass, as shown in Fig. 1 as a function of storage time, is much higher than that due to alpha decay. These are relatively high ionization dose rates by human health standards, and elevated temperatures are expected during interim storage (Fig. 3). These high ionization rates and cumulative doses are difficult, if not impossible, to achieve in laboratory studies of bulk materials [Weber et al. 2009]. In general, available gamma sources have dose rates that are too low to achieve the necessary dose levels in reasonable laboratory time scales; whereas, doping with short-lived beta-emitting isotopes, such as ^{134}Cs, is limited by self-heating to low dopant levels, making it impossible to achieve the necessary doses or transmutation levels. The interaction of electron beams with glasses is the same as that for beta particles and fast electrons produced by gamma interactions, but the dose rates are orders of magnitude higher than those expected in HLW glasses. Electron-beams or highly ionizing ion beams (such as protons or helium) can locally achieve the necessary high ionization dose rates on microscopic to macroscopic scales to attain ionization doses over the range of interest (up to 10^{10} Gy) in relatively short laboratory time scales. However, decomposition, phase separation, and oxygen bubble formation have been observed to occur under these extremely high dose rates [Weber 1988, Weber et al. 1997, Sun et al. 2005], and whether or not such degradation processes occur at the orders of magnitude lower dose rates expected in actual HLW glasses has long been a critical question. A recent study [Ollier et al. 2006] on the effects of temperature and electron flux on oxygen bubble formation in electron-beam irradiated borosilicate glass has demonstrated that the limiting factor in bubble formation is the electron flux, and a threshold in the electron flux for bubble formation has been identified. This threshold electron flux increases with temperature, and the equivalent threshold in ionization dose rate is orders of magnitude higher than the actual ionization dose rate in HLW glasses. Thus, the decomposition, phase separation and oxygen bubble formation that have been previously observed at much higher ionization dose rates are not expected to occur in actual HLW glasses, and beta decay is not expected to significantly degrade HLW glass performance.

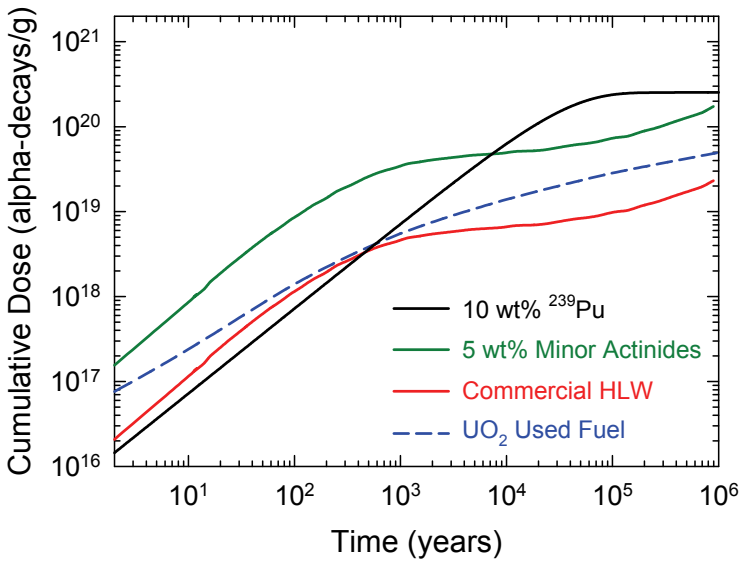

Fig. 4. Cumulative alpha decay events in HLW glass [Weber and Roberts 1983] and glasses incorporating plutonium [Weber et al. 1997] or minor actinides [Weber and Ewing 2013]. The cumulative number of alpha decay events in UO_2 used fuel [Weber and Ewing 2013] is included for comparison.

Another effect of radioactive decay is the change in chemical elements or transmutations that occur as a result of radioactive decay in HLW glasses. The primary transmutations of concern in HLW are from the beta decay of the relatively abundant fission products, ^{137}Cs and ^{90}Sr, which are accompanied by changes in both ionic radius and valence state. The Cs^{1+} decays to Ba^{2+} with a decrease in ionic radius of 20%, and Sr^{2+} decays to Y^{3+}, which in turn decays to Zr^{4+} with a final ionic radius decrease of 29%. One way to mitigate the effects of such transmutations is through the use of multiple cation waste forms with one or more variable valence cations [Vance et al. 1982], which are plentiful in HLW glasses. Thus, unlike crystalline waste forms, transmutations products can be readily accommodated in complex glasses.

Ground water exposed to gamma radiation will form free radicals and molecular species that may affect the dissolution or alteration rate of HLW glasses in the event of water intrusion into the geologic repository. Increases in dissolution rates of up to a factor of 3 have been observed in borosilicate waste glasses due to gamma radiolysis [McVay et al. 1981, Weber et al. 1984]. A very recent study on the effects of gamma radiation on the French SON68 HLW glass has demonstrated limited effects of gamma radiation on the residual alteration rates of the glass [Rolland et al. 2013], with negligible effects on alteration rate from realistic gamma dose rates expected at the time of water intrusion into the geologic repository.

3.2. Alpha decay effects

Self-radiation damage in glasses containing short-lived actinides, ^{238}Pu and ^{244}Cm, has been an active area of research for several decades; thus, radiation effects from alpha-decay in many glasses of interest are fairly well known [Weber and Roberts 1983, Muller and Weber 2001, Weber et al. 1997]. The cumulative numbers of alpha decay events expected as a function of waste storage time are shown in Fig. 4 for commercial HLW glass [Weber and Roberts 1983] and high-actinide glasses for the immobilization of ^{239}Pu or minor actinides [Weber et al. 1997, Weber and Ewing 2013], as well as for commercial UO_2 used fuel [Weber and Ewing 2013] for comparison. In glass waste forms, the effects of alpha-decay are generally small at the ambient temperatures expected over the decay times for actinides. At low doses, there is an increase in stored energy associated with the formation of defect

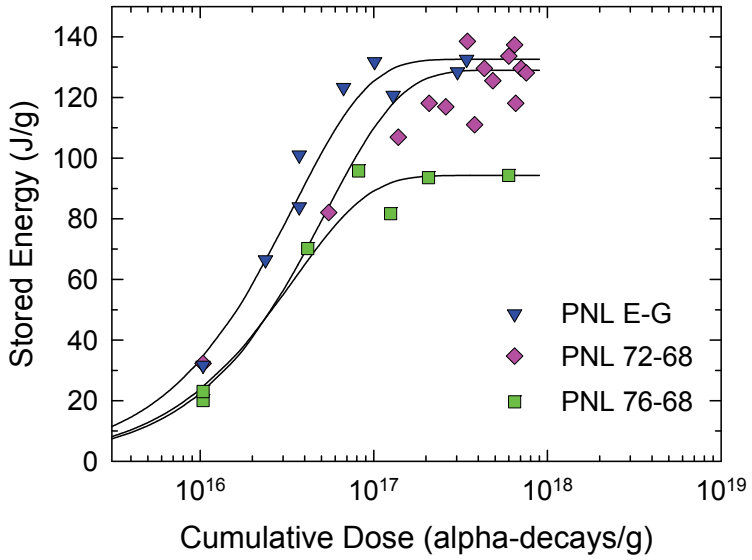

Fig. 5. Stored energy in borosilicate glasses doped with ^{244}Cm [Weber et al. 1997].

centers in the glass structure that saturates at values of 90 to 140 J/g and doses of 1 to 3 x 10^{17} alpha-decays/g [Weber et al. 1997], as shown in Fig. 5. In a recent study, of the French R7T7 glass doped with ^{244}Cm, the stored energy increased with dose to similar saturation stored energy values but at a somewhat higher dose of 10^{18} alpha-decays/g [Maugeri et al. 2012, Peuget et al. 2014]. In all these glasses, there are global rearrangements of the glass network that manifest as volume expansion or compaction at higher doses; these volume changes generally saturate at doses above 2 x 10^{18} alpha-decays/g, as illustrated in Fig. 6. The maximum volume expansion or compaction from alpha-decay damage is on the order of 1% [Weber et al. 1997, Muller and Weber 2001]. In comparing the stored energy (Fig. 5) and volume changes (Fig. 6), the stored energy saturates at much lower doses than the volume

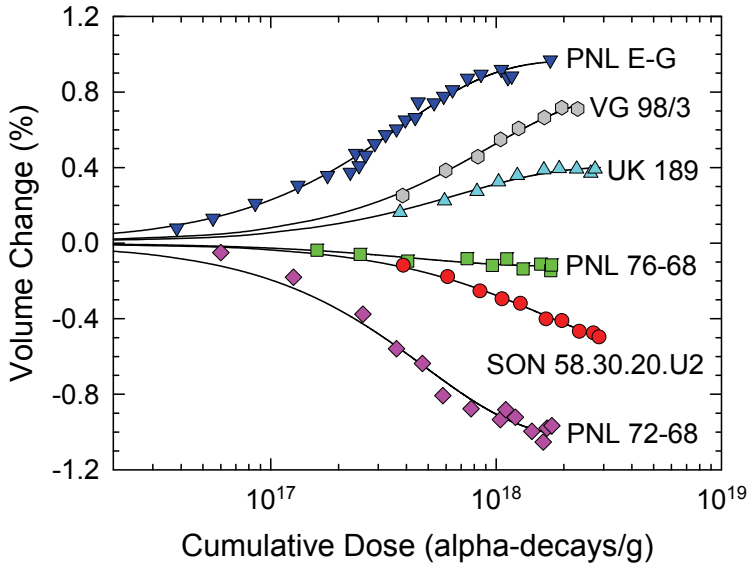

Fig. 6. Volume swelling and compaction in borosilicate glasses doped with ^{244}Cm [Weber et al. 1997] and ^{238}Pu glasses [Muller and Weber 2001].

changes, which is consistent with behavior observed for a broad range of glasses [Weber et al. 1997, Maugeri et al. 2012, Peuget et al. 2014]. This behavior suggests that the origin of the stored energy, such as defects from alpha particle damage, is different from the global network rearrangements associated with the volume changes, which is generally attributed to damage from the alpha-recoil nucleus. Another important point is that regardless of whether a glass undergoes swelling (PNL E-G), compaction (PNL 72-68) or negligible volume changes (PNL 76-68), there is significant positive stored energy, which supports different origins for stored energy and volume changes. Maugeri et al. [2012] have applied the concept of fictive temperature to the analysis of radiation damage due to alpha decay in ^{244}Cm-doped R7T7 glass, where the fictive temperature is the temperature at which the glass structure would be in equilibrium [Tool 1946], and they found that the fictive temperature closely followed the dose dependence of the volume changes. The interpretation of these results is that the alpha recoil nucleus causes ballistic melting and fast quenching of the glass [Griscom and Weber 2011]; this melt-quench process locally creates a new higher enthalpy glass structure that increases in volume fraction with increasing alpha-decay dose.

In a number of ^{238}Pu and ^{244}Cm doped glasses results, self-radiation damage from alpha decay results in decreases in hardness and Young's modulus, but increases in fracture toughness [Weber et al. 1997]. Similar decreases in hardness and Young's modulus have been reported in ^{244}Cm-doped R7T7 glass [Peuget et al. 2006, Peuget et al. 2014].

In a unique study, three compositionally identical Pu-containing borosilicate nuclear waste glasses were prepared, each containing 1 wt% PuO_2 [Weber et al. 1985]; however, the ^{238}Pu/^{239}Pu isotopic ratio was different in each glass. As a result, the alpha-decay rates (dose rates) in the as-prepared glasses varied by nearly a factor of 200. These glasses were studied over a 20 year period [Weber et al. 2003, Griscom and Weber 2011]. The macroscopic volume changes measured for these glasses are shown in Fig. 7 as a function of cumulative dose. The swelling exhibits the typical exponential rise to saturation as observed in other glasses (Fig. 6). However, the results demonstrate that for an order of magnitude difference in alpha-decay rates there is no significant effect of the dose rate on the swelling within this range of alpha activities, similar to negligible dose rate effects previously reported for stored energy in ^{244}Cm-doped glasses [Weber and Roberts 1983]. Analysis of extended x-ray absorption fine structure (EXAFS) spectra for these Pu-containing glasses indicates that the local environment around the cations exhibits different degrees of disorder due to the accumulated alpha-decay dose [Hess et al. 1998]. In general, cations with short

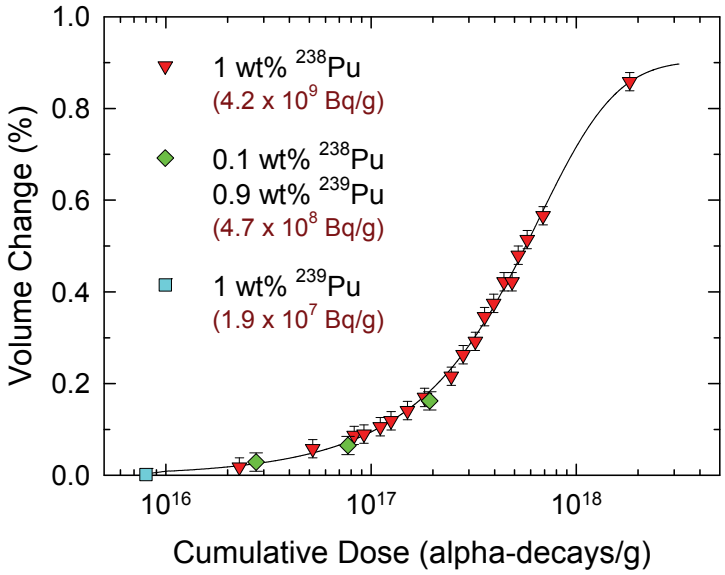

Fig. 7. Volume swelling in a borosilicate nuclear waste glass containing 1 wt% Pu with different ratios of ^{238}Pu/^{239}Pu [Weber et al. 2003]; the corresponding alpha decay rates are shown.

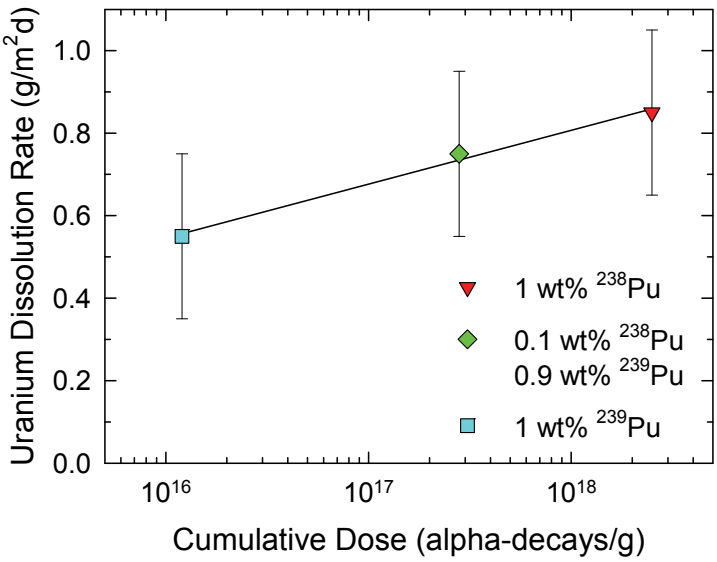

Fig. 8. Uranium dissolution rate in a borosilicate glass containing 1 wt% Pu with different ratios of ^{238}Pu/^{239}Pu [Wellman et al. 2005].

cation-oxygen bonds show little effect from self-radiation, whereas cations with long cation-oxygen bonds show a greater degree of disorder with accumulated alpha-decay dose, consistent with a global network rearrangement. Electron spin resonance was also used to characterize the structure changes from self-radiation damage in these Pu-containing glasses [Griscom and Weber 2011], and revealed for the first time evidence that the alpha recoil nucleus causes local "super vitrification" through a fast ballistic melt and quench process that achieves higher local temperatures than used in the glass melting. Similar processes have been confirmed experimentally and computationally in amorphous silica under ion irradiation [Toulemonde et al. 2011].

A comprehensive set of dissolution kinetics experiments have been performed on these Pu-doped glasses using the single-pass flow-through (SPFT) test method to measure the forward reaction rate as a function of temperature and pH [Wellman et al. 2005]. The forward rate is the most conservative estimate of the radionuclide release rate from the glasses. Because of the high flow-through rate (60 mL/day) used in these tests, the solution in contact with the glass remains very dilute in dissolved glass components, and the buildup of alpha-radiolysis products, which can enhance dissolution rates in these glasses, is minimized. Using dilute pH buffers also controls solution pH. With this test method, it is possible to unambiguously compare the results between glasses with different radiation damage levels and quantitatively assess differences in their dissolution rate. While the elemental concentrations of most elements were above the detection threshold, the Pu concentrations were just at the detection threshold and could not be quantified. Release rates based on release of Al, Cs, and Na cluster between 1 to 2 g m^{-2} d^{-1} for the three glass specimens. Release rates based on B and U are slightly lower (0.5 to 1 g m^{-2} d^{-1}) compared with the other elements. The normalized elemental release rates for U as a function of cumulative dose are shown in Fig. 8 for experiments conducted at 83°C and pH(25°C) = 9. Within experimental error, there is no significant effect of cumulative dose on the forward dissolution rate over the range from 1.2 x 10^{16} to 2.5 x 10^{18} alpha-decays/g. Similar negligible effects on initial dissolution rates in glasses doped with ^{238}Pu and ^{244}Cm have been reported for doses up to 4 x 10^{18} alpha-decays/g [Deschanels et al. 2007, Peuget et al. 2007, Peuget et al. 2014]. Thus, the effects of self-radiation damage from alpha decay on the dissolution rate appear to be negligible, consistent with the relatively minor changes in the glass network structure.

At extreme doses (greater than 8 x 10^{18} alpha-decays/g), helium bubble formation in glasses doped with ^{238}Pu and ^{244}Cm has been suggested from scanning electron microscopy of plastic replica specimens from fractured surfaces, and the measured volume change was 0.51% [Inagaki et al. 1992]. However, in more recent transmission electron microscopy studies, helium bubbles larger than 10 nm are not observed in ^{244}Cm-doped glass after a dose of

8 x 10^{18} alpha-decays/g [Peuget et al. 2014]. When helium is introduced by (n,α) reactions or by ion implantation, helium bubble formation is observed, but only after annealing at high temperatures, and the threshold annealing temperature for the formation of helium bubbles decreases with increasing helium concentration [Weber et al. 1997]. Recent helium implantation studies indicate that large helium concentrations (up to 3 at%) can be accommodated without any significant microstructural changes [Peuget et al. 2014].

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