
Radiation and Thermal Effects on Used Nuclear Fuel and Nuclear Waste Forms

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ABSTRACT

Used nuclear fuel and nuclear waste forms will be exposed to continued radiation and thermal effects during interim storage and permanent disposal that can result in significant restructuring of the materials. During near-term interim storage (up to several hundred years), both used nuclear fuel and waste forms will be subjected to a high-temperature thermal environment produced primarily by beta-decay of short-lived fission products. After this, radiation damage and helium accumulation from alpha decay of actinides will be dominant for millions of years. In the case of used fuels, the radiation damage processes and environment are completely different than in-reactor. As a result, the structure and properties of aged nuclear fuel in a thousand or several hundred thousand years may not be well represented by today's characterization of used fuel. In the long-term, the accumulation of helium from alpha decay is expected to result in helium bubble formation in both used fuel and waste forms, or the potential growth of existing fission gas bubbles in used fuel. While the conditions for helium bubble formation are not well known, existing data suggest that the onset of helium bubble formation could occur during interim storage in high burn-up nuclear fuels and high actinide waste forms.

During the near-term thermal period, the combined effects of radiation and high temperatures from beta and alpha decay can lead to phase instabilities, charge imbalance, redistribution of fission products and actinides, and the onset of helium bubble formation. Predicting the structural and chemical response of used nuclear fuel and waste forms during this high thermal and radiation stage are critical for establishing the benchmark structures on which to model long-term evolution due to continued self-radiation damage and helium accumulation from alpha decay. In order to develop the necessary understanding and predictive models, well-designed experiments to investigate separate and integrated effects are critical. Since timescales of interest range from several hundred to a million years, irradiation response and degradation models must be validated using laboratory-scale experiments that are highly accelerated in time. The experimental approach for this proposed work is based on separate, sequential and simultaneous irradiations with energetic helium ions, heavy ions and electrons, over a wide range of temperatures under highly controlled conditions to investigate the separate and integrated effects of irradiation and helium accumulation on microstructure evolution in model used fuel and nuclear waste form materials. Physics-based constitutive models of the response of used fuel and waste forms during interim storage and geologic isolation will be developed and experimentally validated. Based on thermal and radiation histories, these constitutive models will provide structural evolution boundaries that control radionuclide release to the environment once canisters are breached and interactions with the geological environment commence.