Adaptive Waste Forms: The Concept and Its Implications

SAND2009-7226C

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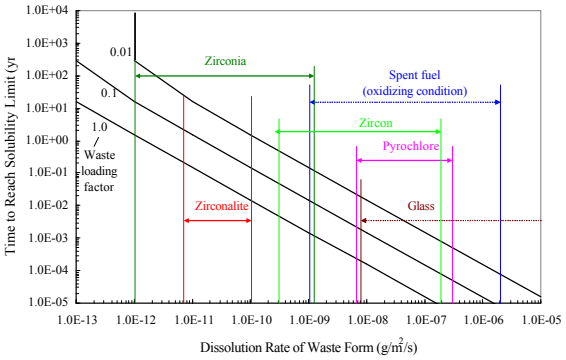
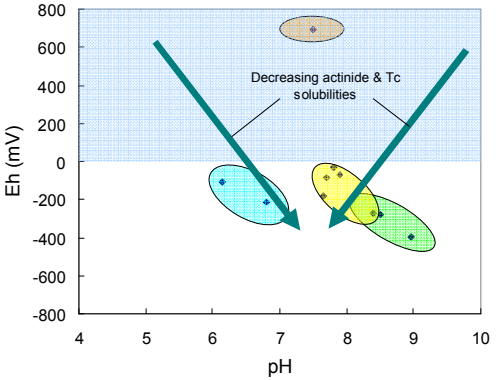
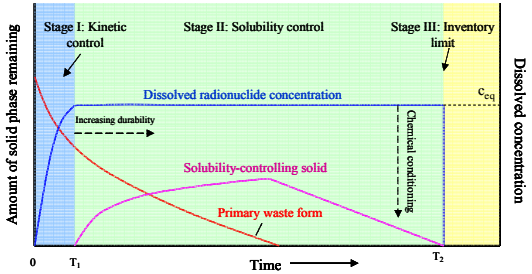
Abstract

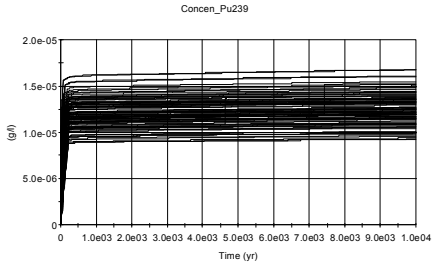
A great deal of effort has been devoted to creating durable waste forms ranging from borosilicate glasses to crystalline mineral phases based on the assumption that a slow waste form dissolution rate would effectively limiting radionuclide release from a geologic repository. From a perspective of long-term repository performance, however, this assumption may not be justifiable. Radionuclide release in a repository is divided into three stages: (1) The dissolved concentration of a radionuclide has not reached its solubility limit and the release of the radionuclide is partly controlled by a waste form dissolution rate; (2) the radionuclide concentration has reached its solubility limit as controlled by the stability of a degradation product and the release of the radionuclide then becomes independent of waste form dissolution rates; and (3), the dissolved radionuclide concentration drops to zero due to the disappearance of both the primary waste form and the solubility-controlling degradation product. The duration of stage 1 is estimated to be less than a few hundreds of years, which is negligible as compared with a typical repository regulatory time ranging from 10 K to 1 M years. Thus, the total release of a radionuclide from a repository is predominantly controlled by the stability of radionuclide-bearing degradation products, rather than by the dissolution rates of primary waste forms. This is mainly due to high solid/water ratios and slow groundwater flow rates in actual repository environments that cause the water contacting waste forms to rapidly reach chemical equilibrium with degradation products. One effective way to limit radionuclide release is to chemically condition waste forms or repository environments to ensure a specific set of degradation products to form that will immobilize radionuclides of concern. The choice of an appropriate waste form composition should be an important aspect of waste form development. A chemical component for repository conditioning can also be added to the repository separately as a backfill. A similar concept has been successfully demonstrated in the Waste Isolation Pilot Plant (WIPP). Therefore, future waste-form development should be increasingly focused on conditioning waste forms or repository environments to minimize radionuclide solubilities, rather than on striving for marginal improvements to the durability of primary waste forms.



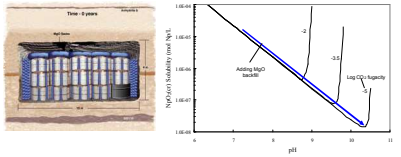




Model Results for Salt Repository: ^{239}Pu

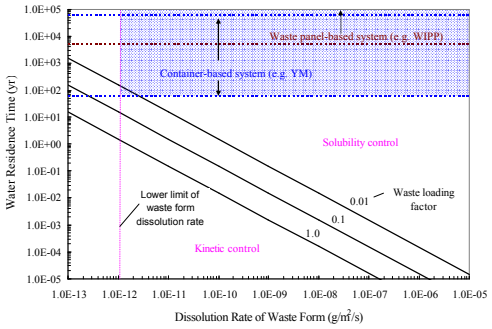


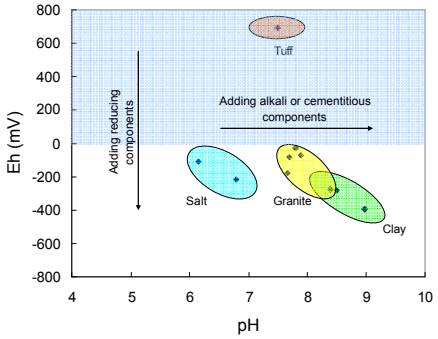
Chemical Conditioning WIPP Repository



A salt-based repository is ideal for chemical conditioning:

- Closed system
- Lack of interference by ambient rocks





Concluding Remarks

- The total release of radionuclides from a repository is primarily limited by the stabilities of waste form degradation products, rather than by the dissolution rates of primary waste forms.
- Waste-form development should be increasingly focused on conditioning waste forms or repository environments to minimize radionuclide solubilities, rather than on striving for marginal improvements to the durability of primary waste forms.
- Radiation damage studies should be directed to the evaluation of the potential effect of radiation damage on the stability of degradation products that will directly control radionuclide solubilities.
  - The effect may be small because alteration products will continuously dissolve and re-precipitate as radiation damage accumulates.