Preferential Radionuclide Release Due to Alpha Decay: Effects on Repository Performance

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The Concept

- Alpha decay-related, time-dependent preferential isotopic release can occur from radioactive waste forms and their alteration products (e.g., two papers from MRS Proc. Vol. 713, 2002: Poinssot et al., p. 615, and Murphy & Pickett, p. 867)
- Current performance assessment (PA) models do not incorporate such preferential release from spent nuclear fuel (SNF) waste forms
- Potential repository performance consequences were estimated by modeling to quantify the effects

Motivation: Isotope fractionation in nature from nuclear effects

- Best known examples: natural waters
- Groundwaters:
  - $^{234}U/^{238}U$ activity ratio typically exceeds one (isotopic equilibrium); majority of reported waters range between 1 and 10, concentrated in the range 1 to 5
  - Shorter-lived Ra and Th isotopes
- Nopal I natural analogue seep water $^{234}U/^{238}U$ of up to 5.1; (Pickett & Murphy, 1999, MRS Proc. Vol 556, p. 809)
- Cause: alpha decay effects (e.g., recoil damage)
- Preferential release is a cumulative reflection of alpha decay ancestry

Preferential release will affect performance of disposed SNF

- PA models used by the U.S. Department of Energy and U.S. Nuclear Regulatory Commission (NRC) for SNF at the proposed Yucca Mountain repository assume congruent release for most radionuclides
- Through time, the inventory for certain radionuclides increases by alpha decay, and the decay product component may be released at a higher rate than estimated by a congruent release mechanism
- Laboratory leaching tests do not reveal time-dependent preferential release, which will not become significant until years in the future
- Murphy & Pickett (2002, MRS Proc. Vol. 713, p. 867) tabulated SNF radionuclides that may be affected, specific to the proposed Yucca Mountain repository, on the basis of
  - proportion of inventory that grows in by alpha decay
  - potential dose effect
- ^{237}Np, ^{234}U, ^{230}Th, ^{226}Ra, and ^{210}Pb

Approach

- Use NRC TPA Version 4.1j computer code
  - SNF only
  - Calculates SNF degradation rate
  - Congruent release, subject to solubility limit
  - Gap fraction affects only Cs, I, Tc, Cl, C, and Se

Approach

- Alpha product inventory at a given time consists of two components:
  - Initial
  - Ingrown
- Ingrown component is a function of:
  - Time
  - Parent decay
- For efficiency, work within the congruent release framework of TPA to account for enhanced release of ingrown portion, while not affecting release of initial component

Approach

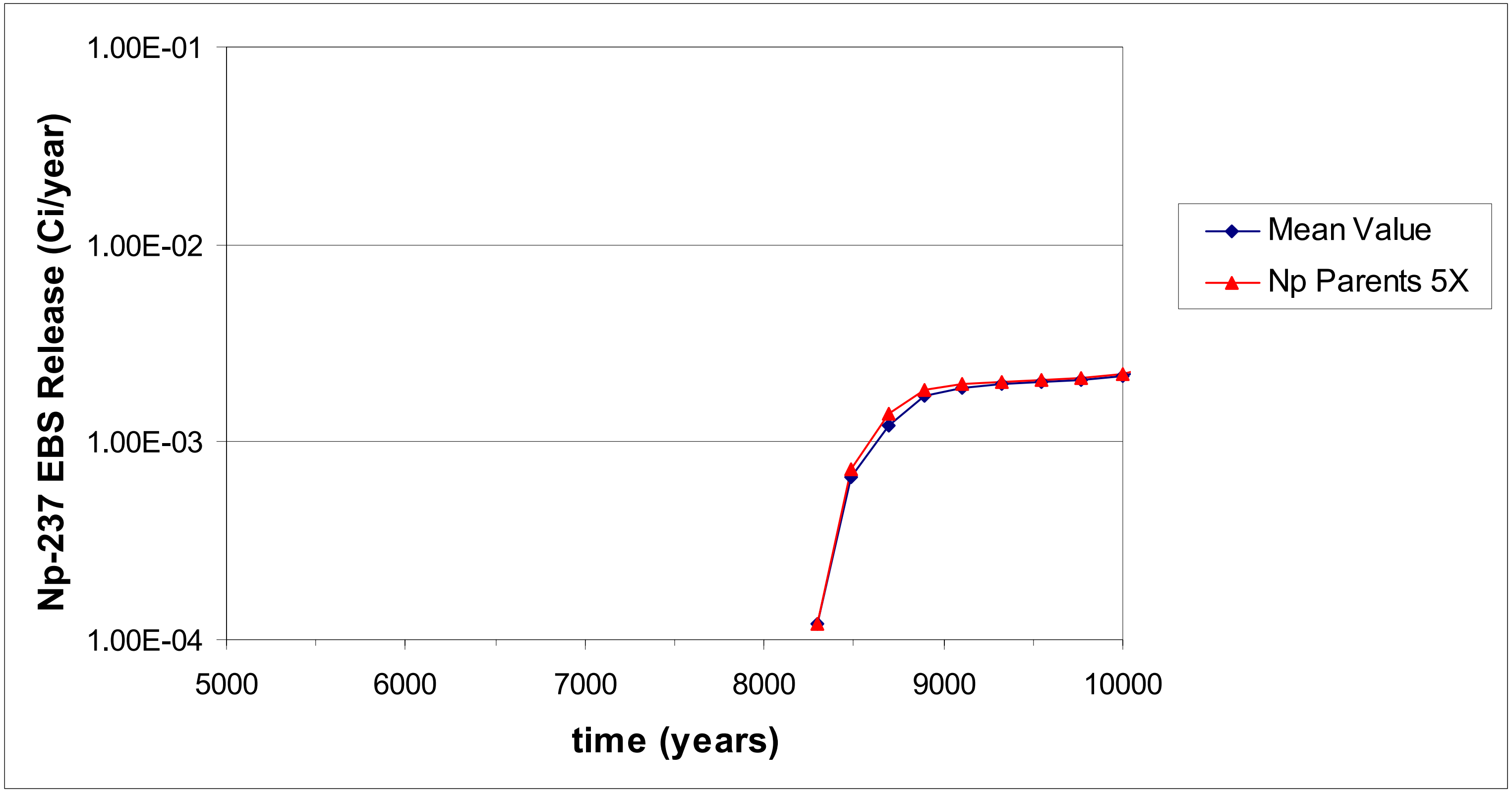
- Increase the initial parent inventory so that the daughter inventory – and, thus, its modeled congruent release – is increased in a time-dependent way
  - Only the ingrown portion is enhanced
- Use a factor of five
  - Based on observed preferential release factors in natural systems (e.g., $^{234}U/^{238}U$)
  - Five-fold daughter enrichment is a reasonable upper bound for a simple test of the effect
  - Apply to all up-chain parents that contribute substantially to alpha ingrowth

Other Modeling Considerations

- Retain solubility control
- Neglect artificially enhanced parent release and dose effects in interpreting results (not an issue if parent is solubility-limited or short-lived)
- Evaluate artificial effects on ingrowth due to altered parent release history
- Check that true parent and daughter inventory would not be exhausted during simulation time

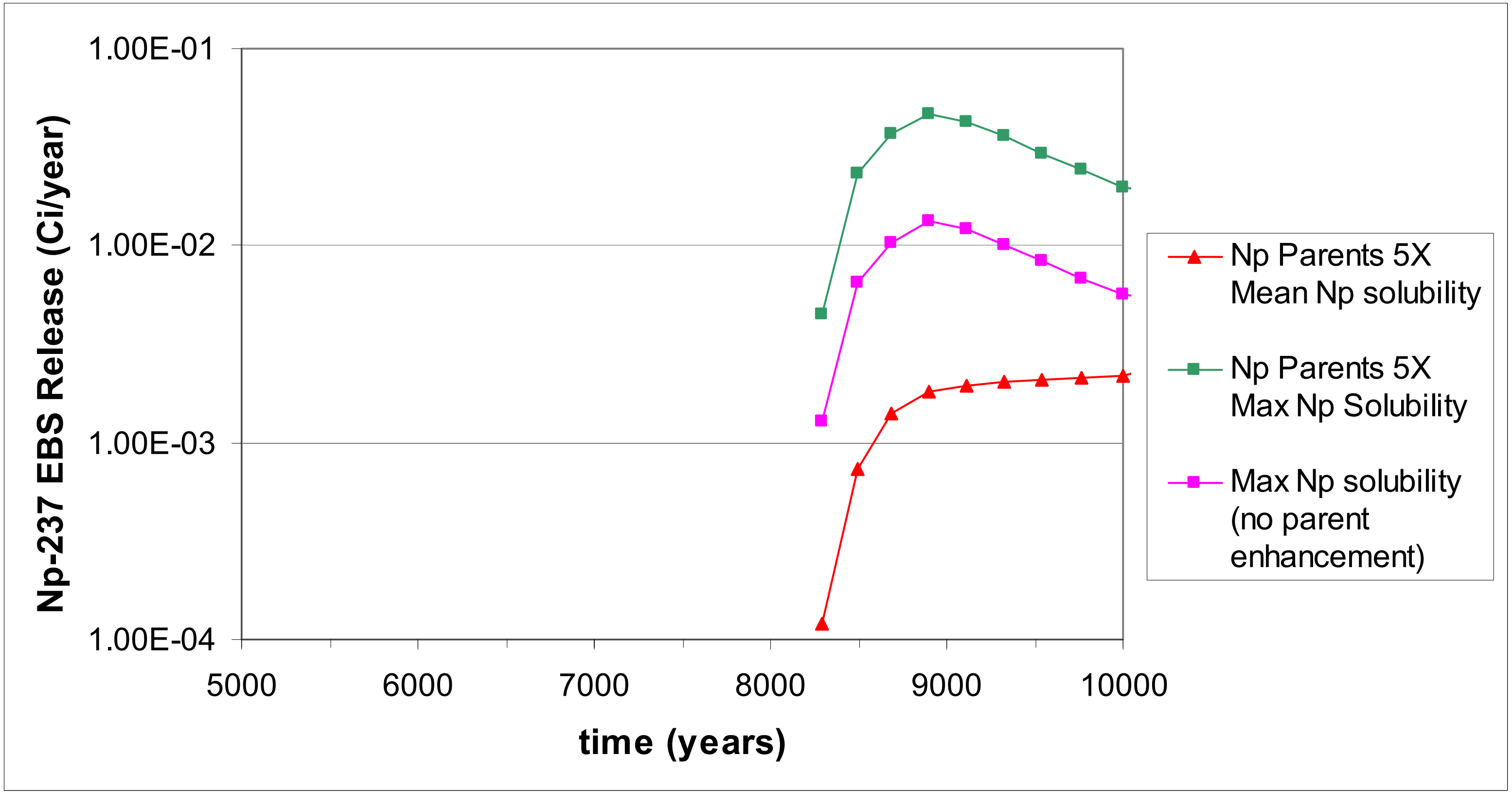
Results of ^{237}Np deterministic, mean-value simulations

Initial ^{241}Pu and ^{241}Am inventories increased five-fold



Solubility effect on ^{237}Np

Deterministic cases



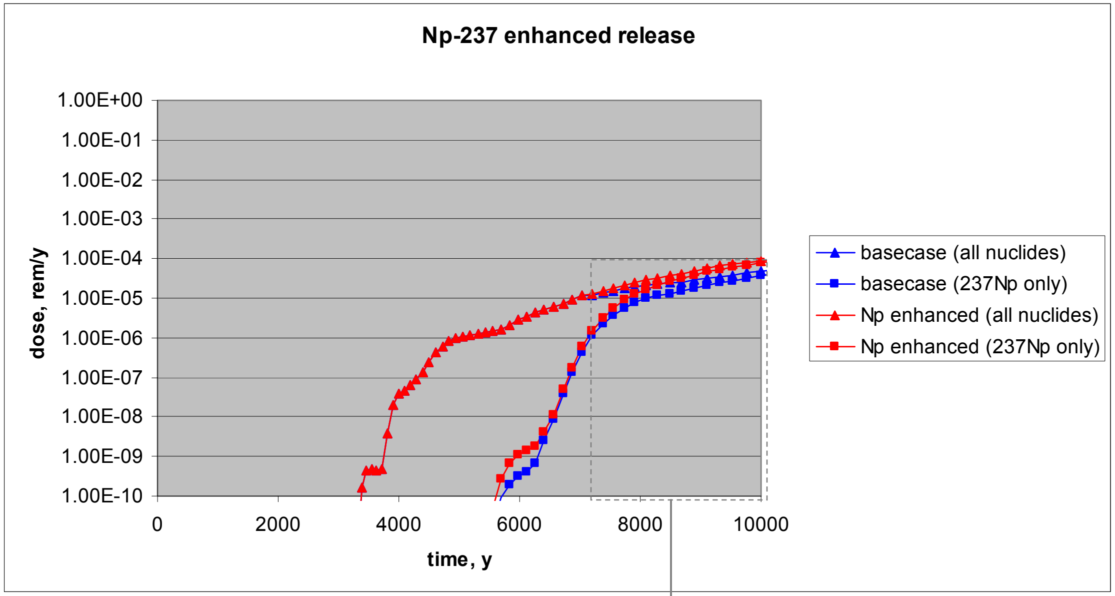
Results of ^{237}Np deterministic, mean-value simulations

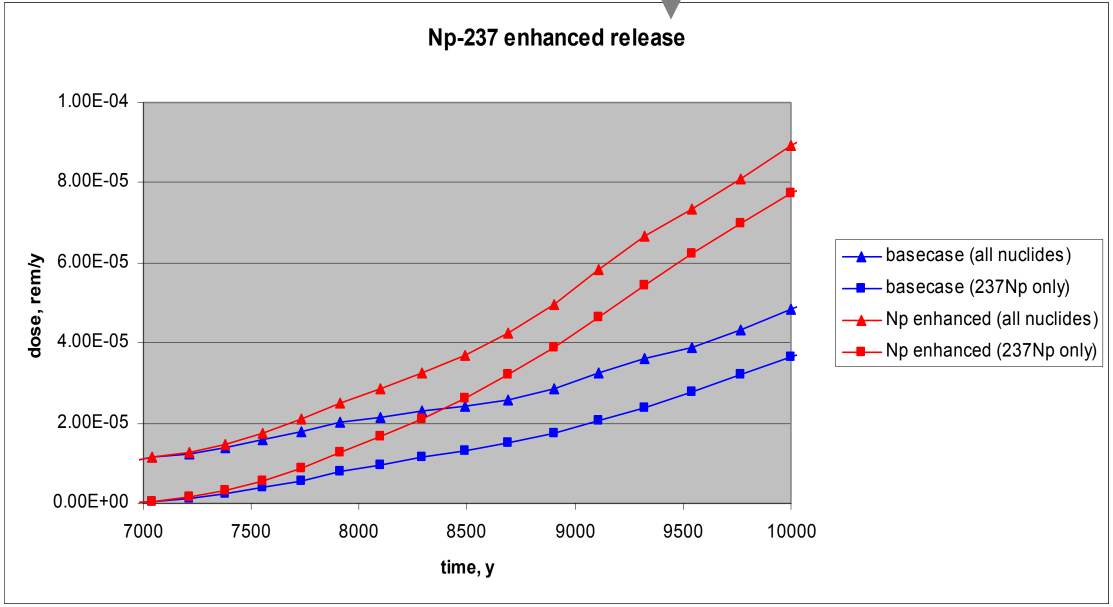
- Pre-10,000 year EBS release is relatively unaffected by parent enhancement. Np is solubility-limited. Confirmed by setting Np solubility limit at its maximum value: release increases.
- Post-10,000 year EBS release (not shown) increases by the correct factor (~3.5), implying that Np is not solubility-limited. Attributed to higher flow rates post-10,000 years.

Stochastic analyses

- TPA 4.1j default inputs (some solubility limits modified)
- 350 realizations
- 10,000-year groundwater dose results for Yucca Mountain
- Parent inventories increased 5X
  - $^{237}Np \rightarrow$ ^{241}Pu and ^{241}Am
  - $^{234}U \rightarrow$ ^{238}Pu
  - ^{230}Th, ^{226}Ra, and $^{210}Pb \rightarrow$ ^{238}Pu and ^{234}U (run separately from ^{234}U case)

Estimated Dose from 350-Realization Stochastic Runs





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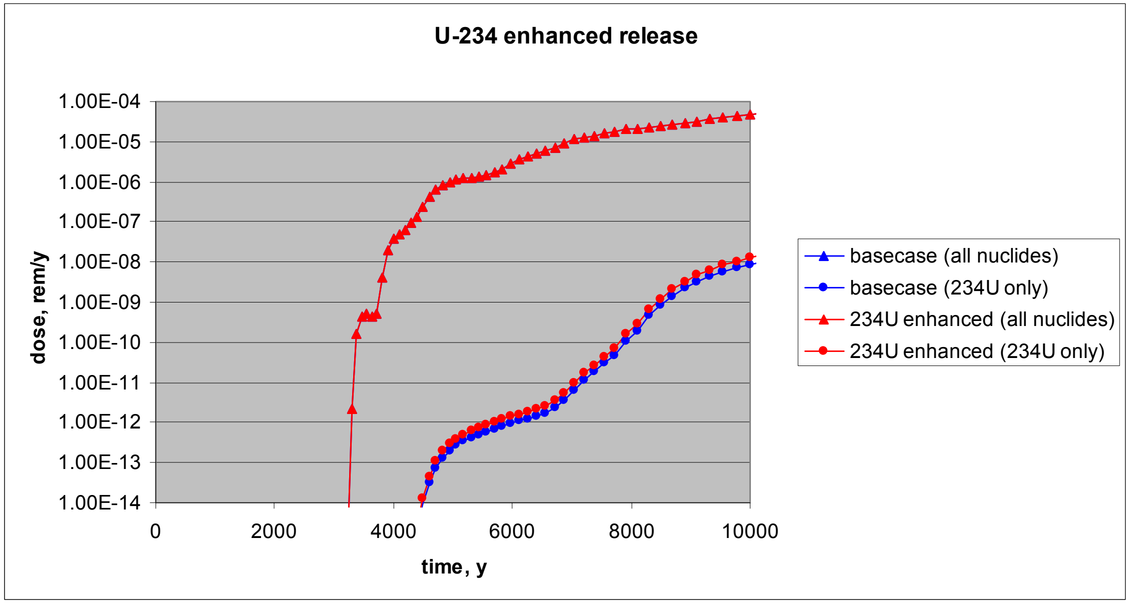

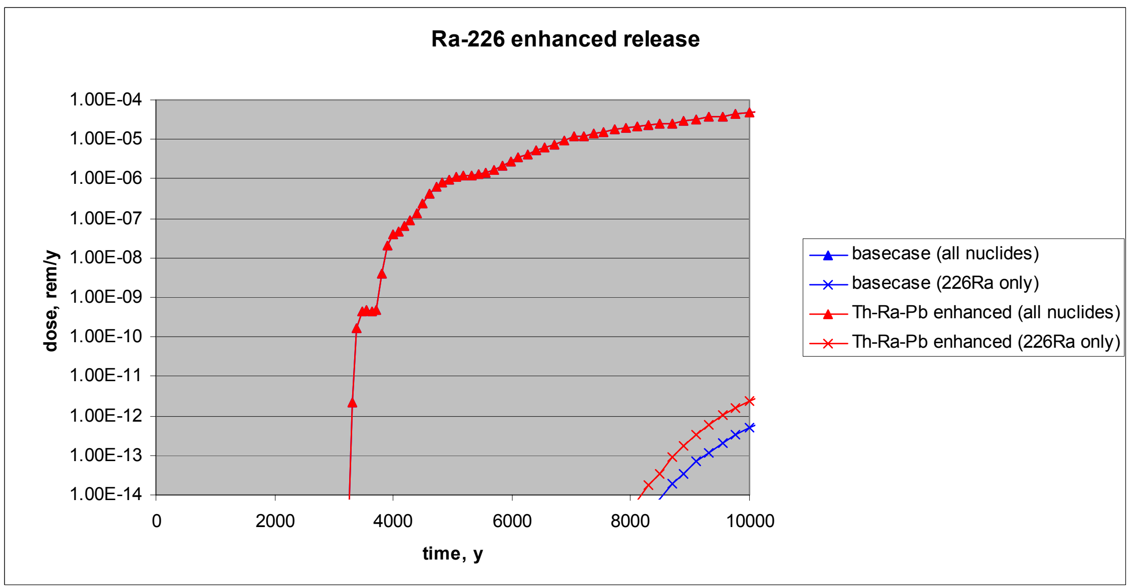

Results of stochastic TPA runs for Yucca Mountain case

- The dose effect of enhanced release is observable in all cases for individual radionuclides
- ^{237}Np effect is partially suppressed by solubility
- Dose effects are potentially significant for ^{237}Np; however:
  - still well below compliance limits
  - secondary U phase formation could negate the alpha daughter effect if phase formation largely post-dates ^{241}Pu and ^{241}Am decay (about 2000 years)

Conclusions

- Preferential release can affect repository performance
- For the Yucca Mountain simulations, the effect does not appear important at 10,000 years:
  - delay afforded by engineered and natural barriers
  - low initial inventory (^{234}U, ^{230}Th, ^{226}Ra, and ^{210}Pb)
  - potential suppression of ^{237}Np enhancement by secondary phase formation
- Potential effect is dependent on specific wastes, barriers, and geochemical conditions

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