

AUTOMATIC SOFTWARE PROCESSING FOR INVENTORIES OF NUCLIDES (ASPRIN)

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INTRODUCTION

EQ6 users have difficulties tracking isotopic concentrations in a system with chemical and nuclear decay reactions. The capability to track isotopic concentrations is particularly important for simulating the degradation of the spent nuclear fuels. Isotopic tracking is necessary to evaluate the criticality risk associated with fissile material relocation inside of a disposal container. A code called ASPRIN (*A*utomatic *S*oftware *P*rocessing, *I*inventories of *N*uclides) was created to allow isotopic tracking using the results of EQ6 simulations. (Ref 1 and Ref 2)

DESCRIPTION

During the degradation process the fissile isotope is degraded from solid to solution. When the fissile isotope is in solution, two chemical paths can follow. One path is generating a mineral, and after that degraded again to solution or from solution can be flush out to the waste package.

The isotopic uranium inventories are computed at time "k" by a direct proportionality to the elemental inventory in a chemical phase.

$$N_{iej}^k = m_{iej}^k N_{ej}^k \quad (\text{Eq. 1})$$

m_{iej}^k = mole fraction of isotope "i" of element "e" in phase "j"
 N_{iej}^k = inventory of isotope "i" of element "e" in phase "j" (moles)
 N_{ej}^k = inventory of element "e" in phase "j" (moles)

The elemental inventory (N_{ej}^k) is read directly from a EQ6 binary output file.

For all phases, the isotopic mole fraction is computed by scaling,

$$m_{iej}^k = \frac{N_{iej}^{k'}}{\sum_{i \in e} N_{iej}^{k'}} \quad (\text{Eq. 2})$$

where

$N_{iej}^{k'}$ = inventory of isotope "i" of element "e" in phase "j" calculated with a predictor / corrector method (moles)

The predictor step accounts for radioactive decay or in-growth, and all contributions from solid phases to the isotopic inventory in aqueous solution. The corrector step accounts for all isotopic donations from solution to solids, and includes any isotopic losses due to flow-through flushing of the waste package.

The general equation to represent the predictor/corrector inventory of the isotope "i" in phase "j" is,

$$N_{iej}^{k'} = N_{iej}^{k''} + \gamma_{iej}^k - \zeta_{iej}^k \quad (\text{Eq. 3})$$

where

$N_{iej}^{k''}$ = prior estimate of isotopic inventory (moles)
 γ_{iej}^k = cumulative isotopic contributions resulting in additions to aqueous solution and radioactive decay (moles)
 ζ_{iej}^k = cumulative isotopic contributions resulting in losses from aqueous solution and flushing out of the waste package (moles)

The contributions comprising the term γ_{iej}^k in Eq. 3 are performed in the predictor step, and the contributions from ζ_{iej}^k are assessed in the later corrector step.

RESULTS

An evaluation test case was implemented for ASPRIN. The test case conforms to idealized waste form and waste package configuration that is modified from previous EQ6 evaluation cases.

This case tests the reliability of the data read from the EQ6 binary output. It furthermore verifies the isotopic exchange, mole fraction, system mole balance and fissile enrichment calculations without implementation of ASPRIN's Pu decay accounting.

This case configuration involves 2 U-Th-C fuel compacts of identical mole inventory and chemical composition. The first fuel compact is altered at a rate of $6 \cdot 10^{-16}$ (mole/s), and the U in this compact is assumed to be pure ^{235}U . The fuel in the second compact is altered at the faster rate $6 \cdot 10^{-15}$ (mole/s), with the U composed of the isotope ^{238}U . Various species and minerals are suppressed in the EQ6 simulation, so that the only U-bearing mineral formed is schoepite.

Comparison results are generated with an independent, case-specific implementation of the ASPRIN solution methodology in the Microsoft Excel 97 SR-2.

Figure 1 shows the concentration of the fissile material (^{235}U) remaining in solid (fuel), fissile material accumulated in aqueous solution, schoepite, and the amount of fissile material flushed out from the waste package in function of the time.

Figure 2 shows the transient fissile enrichments in solution and aggregated solid EQ6 elemental. The absolute error of the fissile enrichment in solution is also shown. The error is below 10^{-3} most of the time.

REFERENCES

1. CRWMS M&O. User's Manual for EQ6 V7.2bLV, DTN: 10075-UM-7.2bLV-00. Las Vegas, Nevada (1999).
2. CRWMS M&O. Software Routine Report for PP Version 1.10, 10194-SRR-1.10-00(C). Las Vegas, Nevada (2000).

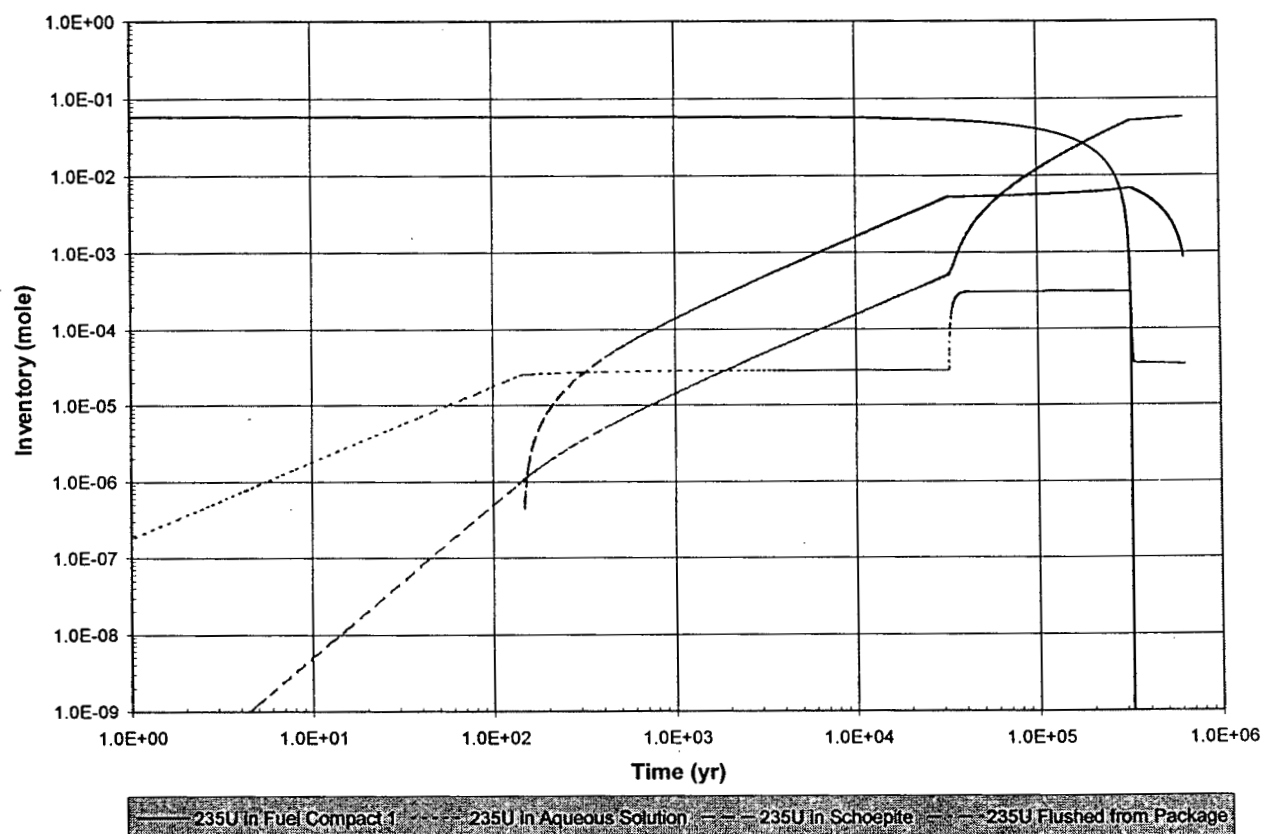


Figure 1. Fissile material (^{235}U) inventories as a function of time.

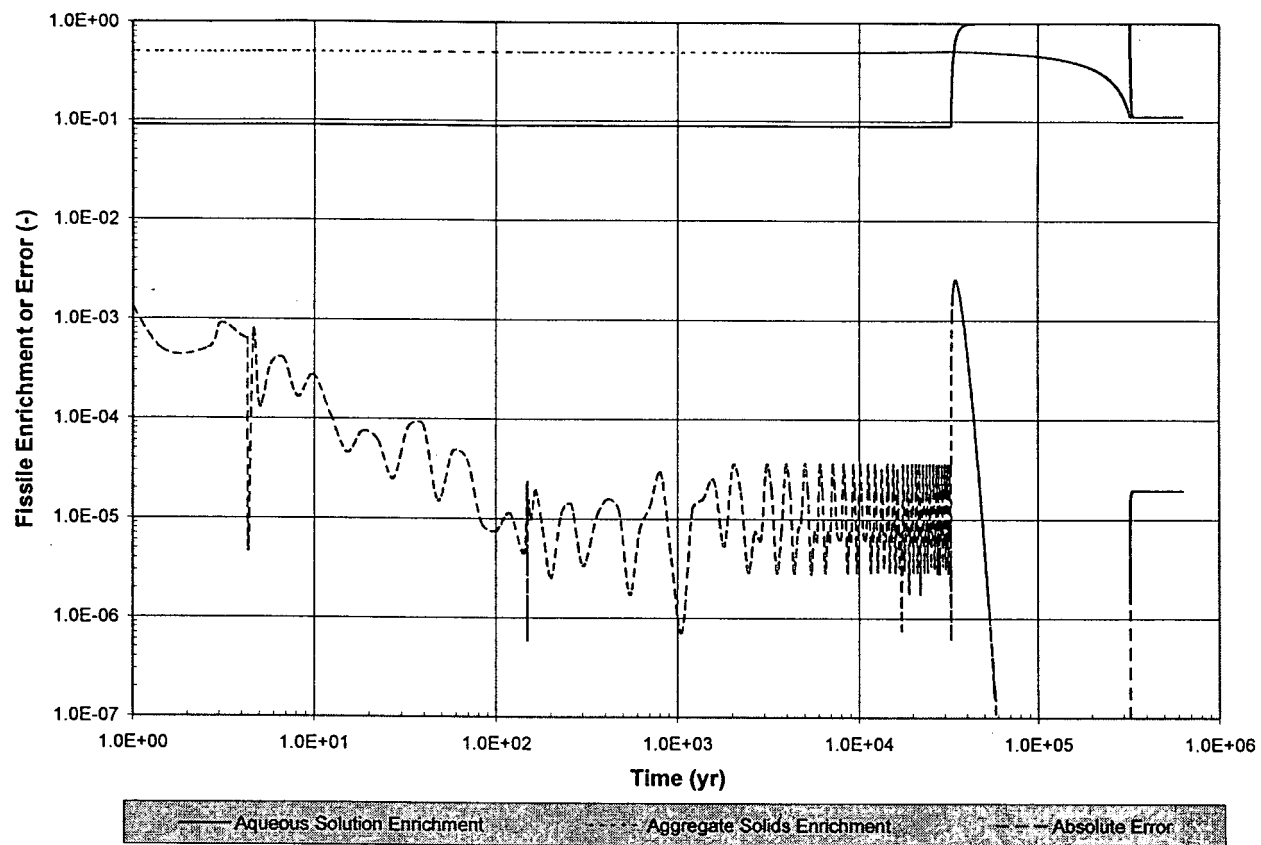


Figure 2. The transient fissile enrichments and absolute error.