HLWYM HEmails

From: David Pickett
Sent: Thursday, February 17, 2005 5:59 PM
To: Richard Codell
Cc: English Pearcy
Subject: RE: Thoughts on Np solubility

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Dick,

Here's where my thinking stands on Np solubility limit. Feel free to forward to interested parties. (Note: calculations and analyses recorded in Scientific Notebook 172. DOE reports used were mainly TBDoc 7, ANL-WIS-MD-00010 Rev 2, and ANL-EBS-MD-000033 Rev 2.)

First, as you probably heard at the latest team meeting, DOE told the NWTRB that they are currently revising the Np solubility limit abstraction. I'll keep looking for the slides at the NWTRB website, but Bret seemed to say that they will be lowering their values. It would be nice if this were a topic in a future NWTRB meeting, but we'll have to wait and see. One of the documents I used - "Dissolved Concentration Limits of Radioactive Elements," ANL-WIS-MD-000010 - has been revised but the revision has not been released.

As I've mentioned, in the most recent DOE documents, they concluded that the basis for a secondary retention model for Np concentration limit was insufficient. For instance, one recent set of experiments showed little to no Np incorporation into schoepite, which DOE considers to be the dominant secondary U phase. I'm sure they're still looking at this, and perhaps new data will be coming out. (Note that Bill Murphy agrees that the data and model are not good enough at this point.)

For now, what I've done is to come up with a new proposed distribution based on the DOE solubility models and in-package chemistry models. We don't have independent models at this point, but I hope to before the end of the year. The DOE abstraction is a function of pH and f(CO2), so I used the most current TSPA abstracted ranges of those parameters and looked to see what Np solubility limits they would predict for those ranges. For instance, the DOE in-package chemistry model for CSNF after 600 years samples pH from a uniform distribution from 4.5 to 7.0. I've examined the bases for this distribution, and it seems sound, though pH up to 8 can be attained if water flux rates are higher than 50 L/yr per waste package. Because pH is sampled uniformly, the log of Np solubility is linear with pH, and f(CO2) has little effect in this pH range, the resulting [Np] distribution is log-uniform.

DOE considers two solubility-controlling Np pure phase solids: Np2O5 and NpO2. The former is observed in low-temperature solubility experiments, while the latter is only observed in higher-temperature experiments. Thermodynamic data suggest that NpO2 should be the more stable phase, but it may be kinetically disfavored at laboratory time scales. Therefore, DOE uses Np2O5 because it gives higher Np concentrations and the lab data are not compelling enough to go with NpO2. I've incorporated the uncertainty in solid phase by using the Np2O5 maximum and the NpO2 minimum for my distribution. (The lower minimum for NpO2 also indirectly accounts for the unrelated possibility of pH up to 8 at higher fluxes leading to lower concentrations.)

The resulting log-uniform distribution, with [Np] in kg/m3:

minimum = -3.95 (1.1e-4 kg/m3)
mean = -2.09 (8.1e-3 kg/m3)
maximum = -0.23 (5.9e-1 kg/m3)

Compared to the current log-triangular TPA distribution, the new maximum is about twice as high, the new minimum is an order of magnitude lower, and the new mean is about one-third. So, overall, the distribution is lower, but because it's uniform instead of triangular, there's the possibility of greater influence of high values.

For comparison to that Figure 6-12 in TBDoc 7 (note units change), this proposed range is from 0.1 to 590 mg/L. So even though the minimum is an order of magnitude lower than the existing TPA minimum, the numbers are still above the majority of the leach test results. But, as I've said before, that's not unexpected for a solubility limit.

I consider this an ongoing process. I am very curious to know what DOE has been up to lately, particularly with regard to Np incorporation.

David

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