HLWYM HEmails

From: David Pickett
Sent: Wednesday, February 09, 2005 1:03 PM
To: Richard Codell
Cc: Andy Campbell; Timothy McCartin
Subject: RE: Thoughts on Np solubility

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Dick,

I'm sympathetic with the desire to avoid over-predicting concentration, and we can talk more about this. You hinted at one distinction we need to make when looking at the data: are they solubility experiments or fuel leaching experiments? One doesn't necessarily expect leach test concentrations to reflect a solubility limit. A good solubility limit should act as an upper bound to the leaching experiments, rather than an average fit. (Analogously, TPA may not always calculate a Np concentration that's at the solubility limit.)

The Pu data in Fig 6-11 are both types. Only the Wilson data are from leaching, and the bulk of them are below the mean curve. The curves do a good job of fitting the solubility data (Nitsche, Rai, Efurd).

For some reason, they didn't show Np solubility data in Fig 6-12 - it's all from leach tests. The leach results are, as a whole, much lower compared to the solubility curve than was the case for Pu. As you suggest at the end of your second paragraph, we should compare the model curve to pure Np solubility data. I'm working on that.

But you're right that we need to consider that pure phase solubility may not be the controlling process for Np concentration. DOE addresses this as an alternative conceptual model in Section 6.6.4 of "Dissolved Concentration Limits...," ANL-WIS-MD-000010 Rev 02. (Note that there is a Rev 03, cited in TBDoc 7, that I don't think has been released yet.) But in TBDoc 7 they decline to use the alternative model due to data uncertainties. We may not have sufficient data to implement it ourselves.

I'll keep in touch.

David

> -----Original Message-----
> From: Richard Codell [mailto:RBC@nrc.gov]
> Sent: Wednesday, February 09, 2005 10:08 AM
> To: dpickett@cnwra.swri.edu
> Cc: Andy Campbell; Timothy McCartin
> Subject: Re: Thoughts on Np solubility
>
>
> Dave:
> Thanks for your assessment. I need to study you explanation, but on
> first look, I don't think I agree completely with it.
> I say that if the themo model was a good model, it would go through
> the middle of the data. Sure it bounds it, but the point is that Np is
> likely to be the most important radionuclide, and bringing the
> solubility model into compliance with the data takes on more
> significance given that the "safety factor" for the calculations has
> less room for error for a million year calculation. The estimate for
> Np solubility needs to be taken with the same degree of conservatism

> or realism as the many other important factors in our and DOE's
> analyses. If you look at figure 6-11 in TBD-7, you see that the model
> for Pu solubility is much more convincing when compared to the data.
>
> I think I can speak for Tim McCartin as well. We jointly feel that its
> best when the model and data agree well with each other, but if we
> have to make a choice between the two, we should always take the data
> over the model. I feel that something is going on with Np solubility
> that isn't being explained well by the thermo model, probably that the
> Np doesn't exist as a pure phase (maybe all of the uranium around). It
> would be interesting if the solubility of Np could be compared against
> data for a pure phase without U present.
>
> I don't know where to point you next because I am sure you are more
> familiar with the literature than I. I hope something turns up about
> the solubility. Perhaps we can force DOE to do some more experiments
> on this matter?
>
> Dick Codell
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>
> >>> David Pickett <dpickett@cnwra.swri.edu> 02/08/05 11:10AM >>>
> Dick,
>
> I was just looking at Figure 6-12 of TBDoc 7: In-Package Environment
> (page 6-19). I think DOE is overstating the case in the paragraph
> above the figure when they say, "...the measured neptunium
> concentrations in spent fuel corrosion experiments are 4 to 6 orders
> of magnitude lower than the [modeled] concentrations...." For the
> sake of argument, let's disregard the ANL Low Drip results (brown
> squares], one of which already exceeds the model solubility limit in
> Figure 6-12. If you lower the model curve by one order of magnitude,
> the curve will barely exceed the highest Wilson data point at about pH
> 8.2. My quick ruler-and-calculator estimate is that the curve value
> will be about twice the highest measured value in that pH region. The
> ANL High Drip result at about pH 6 would be about one order of
> magnitude below the lowered curve. [It appears they used the fCO2 =
> 10e-3 curve.]
>
> This tells me that their current curve does a pretty good job of
> serving as a model of a concentration limit. And it is, of course,
> thermodynamically based. It seems to me that, if we intend our
> solubility distribution to represent a concentration limit (which we
> do), our values should reflect values similar to the DOE model at
> appropriate pH-CO2 ranges, as expected in the package.
>

> David
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Hearing Identifier: HLW_YuccaMountain_Hold_EX
Email Number: 636

Mail Envelope Properties (dpickett@cnwra.swri.edu20050209130300)

Subject: RE: Thoughts on Np solubility
Sent Date: 2/9/2005 1:03:10 PM
Received Date: 2/9/2005 1:03:00 PM
From: David Pickett

Created By: dpickett@cnwra.swri.edu

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