HLWYM HEmails

From: Vijay Jain [vjain@swri.edu]
Sent: Tuesday, November 23, 2004 10:07 AM
To: Tae Ahn; Richard Codell
Cc: 'David A Pickett'; jmcmurry@swri.edu
Subject: RE: notes for a reply to Tae

Tae and Dick,

We still feel that there are several significant issues about how DOE has constructed its in-package chemistry model, particularly with respect to the codisposal waste packages.

First, let's recall that the focus of all three of these KTI agreements specifically is the in-package chemistry model and how it is conceptualized.
So, what does the in-package chemistry model actually do, and why is that important?

The in-package chemistry model is to designed to simulate the changes in solid and aqueous chemistry that would occur over time inside a breached waste package. The main purpose of the model, ultimately, is to provide several bits of information to other waste form submodels:

. The calculated in-package pH value is supplied directly or indirectly to the wasteform submodels to determine the waste package source term.
. In addition to being used directly in determining the commercial SNF degradation rate and the HLW glass degradation rate, the pH also is an important direct input for calculating the solubility of radionuclides after they are released from the SNF or the glass wasteform. Also, colloid concentrations and cladding degradation are influenced by the pH of the water. All of these, not just the degradation rates of the wasteforms, comprise the waste package source term that is supplied to other models.
. The other main outputs of the in-package chemistry model are ionic strength (which is a direct input for the colloid model, indirect input for solubility calculations), and carbonate and fluoride concentrations.

As DOE states, "The pH is the most important output parameter from the in-package chemistry abstraction because it has the greatest influence in determining rates of radionuclide release from waste packages." We agree.
Our evaluation of the KTI agreements involving the in-package chemistry model can all be traced back to questions about whether or not the DOE model provides the appropriate range of pH values, to be supplied to other models as input terms or conditions.

There are two components in the waste package that appear to be capable of producing extreme-pH conditions inside the waste package-carbon steel (potentially very low pH) and glass (potentially very high pH). If the corrosion of the carbon steel is modeled simplistically, without accounting for accompanying reactions like surface complexation on corrosion products, then pH values lower than 3 are predicted by the model. DOE recognized that this representation was unrealistic, however, so in their model the pH is calculated separately, to include surface complexation effects, during the period of time that DOE estimates that carbon steel corrosion would be an important process.

It is the other end of the pH range, the dissolution of the glass wasteform, where we have the most concerns about DOE's development of the in-package chemistry model:
. In our comments about CLST.3.02, we noted that DOE selected only one type of codisposal package to represent all of the codisposal packages in the repository. But this selected package type contains proportionately much less glass than the other types of codisposal packages. More glass per package, more opportunity for high-pH conditions to develop inside the waste package. There also

would be a considerably greater number of the "glass-rich" type of waste packages compared to the type DOE selected as "representative". The main concern here is how glass dissolution affects the pH inside the waste package -- has DOE represented the full range of possible alternatives?

. Similarly, we think that DOE may have underestimated the amount of glass surface area that would be available for reaction inside the canister. Greater glass surface area has the potential to generate higher pH excursions inside the waste package.

. In our comments about ENFE.3.04 and CLST.3.05, we noted that the DOE's EQ3/6 calculations of glass dissolution predicted a maximum pH of about 10. However, we also noted that there is a large body of experimental evidence for glass dissolution where pH values of 10 to 12 routinely are observed. The validation of the in-package chemistry model did not address the experimental evidence for higher pH values, or why the EQ3/6 calculations were lower than this. However, if there indeed are circumstances in which the pH of the effluent inside the waste package would be larger than the maximum value that is currently predicted by the in-package chemistry model, then the DOE wasteform submodels would not be receiving the full range of information that they need for solubility calculations, colloid formation, and so forth.

We've also looked again at the wording of Agreement CLST.3.05. It appears that the NRC's intent in reaching this agreement was to be able to follow DOE's plans for any additional experimental work they might undertake related to in-package chemistry. DOE's restatement of the request was that they would inform NRC of plans for any additional experimental work, if they decided to do any. In their response, DOE notes that they instead chose to develop the model further with sensitivity analyses and an expanded set of input parameters, and so there was no further experimental work to provide the plans for. In that respect, it could be argued that DOE has satisfied the agreement item, and it can be closed.

The concern that we noted in our reply to CLST.3.05 focused on the fact that DOE's in-package chemistry model does not appear to have considered the large body of existing experimental evidence for glass dissolution with pH values greater than 10. Given that we have expressed this concern elsewhere, in our comments about validation of the model in ENFE.3.04, closing agreement item CLST.3.05 seems reasonable on the basis of the DOE response. However, following the "Guidance for Reviewing and Responding to KTI Agreements" that was issued by NRC on September 29, 2004, for the reasons we have discussed above (plus the other concerns about validation of the model that are described in ENFE.3.04), we think that the NRC response to CLST.3.02 and ENFE.3.04 should be to supply comments and feedback on why these items could not be closed.

Bottomline we don't think that the DOE is on the right track on this one like the other CLST agreements where we have seen DOE plans that will resolve our concerns.

Let me know if we need to have a meeting to further discuss this.

Vijay

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