HLWYM HEmails

From: Tae Ahn
Sent: Wednesday, June 29, 2005 8:07 AM
To: David Pickett; Paul Bertetti; Aladar Csontos; Allen Fetter; Bret Leslie; John Bradbury; Richard Codell; Timothy McCartin
Cc: Andy Campbell; Marissa Bailey
Subject: Ca and Si Effects: Re 2: recent papers on incorporation of neptunium into secondary uranyl minerals

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There was another accompanying paper by Douglas et al. on Np incorporation in uranosilicates. Cations and Iron (or its products, in a separate German paper) seem to stabilize Np better, although cation supplies may be limited.

Dick, I am the person who talked with Robert Finch at the DC Chapter Materials Meeting. At the Chapter monthly meeting I presented materials issue at YM last month. Robert sent these papers later to me and I forwarded them to you and others. The Center is conducting experiments (Pau Bertetti) on these, and I and Allen Fetter are working on the energetics of Np incorporation. Allen is planning to have a staff exchange at the Center on Np incorporation (precipitation or sorption) in iron products. Additionally I have feedbacks on those papers from Germany where these papers are also being evaluated. I encourage you to join our attempts. You have had always sparkling insights on important issues.

Tae

>>> Richard Codell 06/28/05 05:11PM >>>
I have read a series of papers dealing with the incorporation of neptunium and other nuclides like plutonium into some secondary uranyl minerals. This is an important topic because both neptunium and plutonium are significant dose contributors, so if they are tied up in the secondary minerals and not released, it would lead to lower doses.

There are three papers:
1. Corrosion of commercial spent nucliear fuel. 1. Formation of studtite and metastudtite by Hanson, McNamara, Buck, Rreise, Jenson, Krupka and Arey

2. Corrosion of commercial spent nuclear fuel. 2. Radiochemcial analysis of metastudtite and leachates by McNamara, Hanson, Buch and Soderquist Both published in Radiochimica Acta, vol 93, 2005

and
3. Neptunium (V) partioning to uranium (VI) oxide and peroxide, by Douglas, Clark, Friese, Arey, Buck and Hanson, pubished in Environmental Science and Technology (Either in press or published recently but date unknown)

I know I got these papers from someone rather than finding them myself, but I honestly can't remember who gave them to me.

All three papers deal with the formation of studtite and metastudtite, both uranyl peroxides, under conditions of radiolysis either because the spent fuel was radioactive or hydrogen peroxide was added. There was strong incorporation of neptunium and plutonium into these secondary minerals, particularly the more hydrated metastudtite, and that led to an apparently lower release rate to the water, particularly for neptunium. This appears to be good news for the release of troublesome neptunium from the fuel, but the news is tempered by other considerations mentioned in the papers:

1. Metastudtite and studtite, both uranyl peroxides, form under special conditions such as when the hydrogen peroxide concentration is high. Only certain experiments with irradiated spent fuel produced these minerals. In the experiments conducted for these papers, they used finely ground spent fuel with relatively small quantities of water with no exchange of water for at least 2 years, and at low temperatures. Similar conditions of low temperature in the K-basin storage sites at Hanford also led to the formation of uranyl peroxide minerals. Uranyl peroxides also seem to have formed in natural uranium deposits for which there was long exposures to peroxides formed by radiolysis. However, other experiments on irradiated spent fuel conducted elsewhere did not produce uranyl peroxides, but produced schoepite and metaschopite instead (minerals for which there is no appreciable capture of neptunium). These other experiments had probably lower concentrations of peroxide, more exposure to atmospheric air, and higher temperatures.

2. In the 3rd paper, the authors show that even though there is appreciable capture of neptunium into metastudtite, it seemed to be released at a faster rate than expected when the metastudtite dissolved. Therefore, even if all worked O.K. for the production of metastudtite, it does not seem to be a trustworthy material to capture the neptunium in the long term.

3. Unirradiated uranium dioxide never seems to form these uranyl peroxides. It seems therefore that the mechanism of formation depends (at least) on a strong enough radiation field to produce hydrogen peroxide. Therefore, if most waste packages are intact for many tens of thousands of years, and are relatively dry to start with, conditions for the formation of uranyl peroxides would not appear to be present in most cases.

Reading these papers makes me question my earlier feeling that neptunium concentrations would be lowered by incorporation into secondary uranyl minerals.

Please feel free to review these papers yourselves and comment on any of the conclusions I have drawn.

Dick Codell

Richard B. Codell, Ph.D.
Senior Hydraulic Engineer
Division of High-Level Waste Repository Safety Office of Nuclear Material Safety and Safeguards U.S. Nuclear Regulatory Commission
11545 Rockville Pike
Rockville MD 20852
Phone 301-415-8167
Fax 301-415-5399
EMail RBC@NRC.GOV

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From: Tae Ahn

Created By: Tae.Ahn@nrc.gov

Recipients:
"Andy Campbell" <Andy.Campbell@nrc.gov>
Tracking Status: None
"Marissa Bailey" <Marissa.Bailey@nrc.gov>
Tracking Status: None
"David Pickett" <dpickett@cnwra.swri.edu>
Tracking Status: None
"Paul Bertetti" <pbertetti@cnwra.swri.edu>
Tracking Status: None
"Aladar Csontos" <Aladar.Csontos@nrc.gov>
Tracking Status: None
"Allen Fetter" <Allen.Fetter@nrc.gov>
Tracking Status: None
"Bret Leslie" <Bret.Leslie@nrc.gov>
Tracking Status: None
"John Bradbury" <John.Bradbury@nrc.gov>
Tracking Status: None
"Richard Codell" <Richard.Codell@nrc.gov>
Tracking Status: None
"Timothy McCartin" <Timothy.McCartin@nrc.gov>
Tracking Status: None

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