HLWYM HEmails

From: William Murphy [wmurphy@swri.edu]
Sent: Friday, June 09, 2000 2:01 PM
To: Bret Leslie
Cc: English Pearcy (epearcy); James Prikryl (jprikryl); David Pickett (dpickett)
Subject: RE: uranophane experiments

Follow Up Flag: Follow up
Flag Status: Flagged

Bret,

Thanks for your acknowledgment.

To your questions: I think that incomplete accounting for aqueous speciation, nonstoichiomentry of the solid, and lack of reversal are all important problems with the Perez et al. data. The speciation problem could be corrected if they reported the complete chemistry and we did EQ3 analyses. Maybe we'll get some of those data in response to my email, but they didn't even measure Ca or SiO2 in most cases. The stoichiometry and reversibility problems probably result in their data providing at best a limit on the solubility, which could be used possibly to confirm our data.
Our experiments were designed and conducted to attempt to avoid all these problems, but we'll probably encounter others.

Bill

-----Original Message-----
From: Bret Leslie [mailto:BWL@nrc.gov]
Sent: Friday, June 09, 2000 12:29 PM
To: wmurphy@swri.edu
Cc: EPEARCY@swri.edu
Subject: Re: uranophane experiments

Bill,

Thanks again for your careful reading of the Perez and Bruno paper and your diplomatic attempts to obtain info that will help our own experiments.
Cheers,
Bret

>>> William Murphy <wmurphy@swri.edu> 06/09 10:44 AM >>>
Dear Jordi and Dr. Perez,

I read with great interest your recent paper in GCA on uranophane dissolution. I have long been impressed with the significance of the problems you attack, perhaps because I desire to attack many of the same ones. Just this week I was reading old papers of Jordi*s on coprecipitation while contemplating a GSA abstract. I have several questions about your experiments which are listed below. Your responses would be appreciated. We are conducting some similar uranophane dissolution experiments and had similar difficulty synthesizing a stoichiometric material. Eventually by adjusting concentrations in the starting solutions and following a procedure like that used by Cesbron et al. (Min. Mag., v. 57, p. 301-308), we succeeded in synthesizing a phase that corresponds well to the ideal stoichiometry and XRD pattern. I can give you more information if you are interested.

Best regards,

Bill Murphy

Questions about "The thermodynamics and kinetics of uranophane..." by Perez et al. GCA, v. 64, p. 603-608.

1. You note that the uranophane is Ca deficient and state that *ICP-MS analyses show that this ion could have been partially substituted by sodium* (p. 603). Can you report the Na data?

2. Reference is made to a few Ca and Si analyses of the reactant solutions, and it is stated that these data showed the assumption of congruent dissolution to be correct. Can you report these data?

3. Carbonate speciation was purported to have been calculated. Can you report these results? I guess that the calculated activities of aqueous species were not used in the equilibrium constant determinations. It is asserted that for certain experiments the tris-carbonato-uranyl species is dominant, and I presume that all dissolved uranium was attributed to this species for the calculations of equilibrium constants. Is this true?

4. Variable Na bicarbonate solution matrices were used. What was the corresponding CO2 partial pressure? Was there a potential for CO2 degassing (or gassing) of the solutions? Did you analyze the bicarbonate?

5. Do rates of dissolution (rdissol) refer to rates of U release or rates of uranophane dissolution (which differ by a factor of 2 for the indicated uranophane formula)? Text indicates that the rates are based on data for U release normalized to surface area, suggesting they are U rates, but other text implies they are uranophane rates.

6. Batch experiment rates are an order of magnitude faster than flow-through reactor rates under the same conditions. Can you provide a possible explanation for this result?

7. Curves based on equilibrium constants in the plot of log U versus HCO3- are difficult to understand because equilibrium curves depend on Ca, Si, and pH, as well as uranium and bicarbonate. Perhaps Ca and Si values were estimated by assuming congruent dissolution and measured pH values were used. Is this true? Were the same values for Ca, etc., used to calculate the curves using the HATCHES and Nguyen equilibrium constants?

!
!
!

!
!
!

Hearing Identifier: HLW_YuccaMountain_Hold_EX
Email Number: 843

Mail Envelope Properties (000001bfd23c$9e3f6a10$30c8a281)

Subject: RE: uranophane experiments
Sent Date: 6/9/2000 2:00:36 PM
Received Date: 6/9/2000 2:01:14 PM
From: William Murphy

Created By: wmurphy@swri.edu

Recipients:
"English Pearcy (epearcy)" <epearcy@swri.edu>
Tracking Status: None
"James Prikryl (jprikryl)" <jprikryl@swri.edu>
Tracking Status: None
"David Pickett (dpickett)" <dpickett@swri.edu>
Tracking Status: None
"Bret Leslie" <Bret.Leslie@nrc.gov>
Tracking Status: None

Post Office: faerie.cnwra.swri.edu

Files	**Size**	**Date & Time**
MESSAGE	4243	6/9/2000 2:01:14 PM

Options
Priority: Standard
Return Notification: No
Reply Requested: No
Sensitivity: Normal
Expiration Date:
Recipients Received: Follow up