

HLWYM HEmails

From: Vladimir Cvetkovic [vdc@avat09.ce.kth.se]
Sent: Friday, April 20, 2001 9:02 AM
To: dpickett@swri.edu
Subject: Re: Pu desorption (fwd)
Attachments: letter to Vladimir

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david --

enclosed is the reply I got from ningping for your record. Unfortunately I am not able to open the attached file and I have no one in the vicinity with a Mac. Would it be possible for you to help me, by converting the attached file to a Word file and sending it back to me? I am anxious to read her letter.
Thanks in advance!

vladimir

----- Forwarded message -----

Date: Wed, 18 Apr 2001 17:58:50 -0600
From: Ning Ping Lu <ningping@lanl.gov>
To: vdc@avat09.ce.kth.se
Subject: Re: Pu desorption

Dear Dr. Cvetkovic:

Thank you very much for your e-mail of April 17. Attached file is my answers to your questions in the e-mail, including the desorption data of Pu from Pu-loaded colloids of hematite, goethite, smectite, and silica as a function of time. The hematite and goethite data were mentioned in the article in Radiochim. Acta, 1998. The smectite and silica data did not published. In my letter, I have made an explanation about the estimated Kd values after desorption in the report. If you have more question about the report LA-UR-00-5121, please let me know and I will try my best to answer it.

Sincerely

Ningping Lu

At Tue, 17 Apr 2001 22:03:25 +0200 (CEST) you wrote:

Dear Dr. Lu --

I am working together with D. Pickett, D. Turner and S. Painter from CNWRA at SWRI on colloidal transport at Yucca Mnt. As part of this work, I am currently studying in detail your report LA-UR-00-5121. I have a few questions.

1. I believe I understand how to relate the fraction of Pu adsorbed (Figure 1) and Kd values from Table 2, by using eq. 5 (equilibrium assumption) and correcting for sorption in the equipment. What I do not understand is how to relate your fractions desorbed (after 293 days) and your estimated Kd value (Table 6). For instance, for silica/natural water the adsorbed fraction is 0.62 and corresponding Kd=8.1 L/g. For the desorption, you state (p.26) that 20% of the adsorbed Pu was desorbed after 293 days. Doesnt this mean that after 293 days, $0.2 \times 0.62 = 0.12$ (i.e.

12%) of the original Pu mass is in solution, and $0.62 - 0.12 = 0.5$ (i.e. 50%) is adsorbed on silica? The partitioning coefficient is then $0.5/0.12 = 4.2$ which would imply $K_d = 4.2/0.2 = 21$ L/g against your 6.3 L/g (Table 6). Can you see where there is a flaw in my reasoning?

2. In your article in *Radiochim. Acta*, 1998, you state (p.169, sec.5) that desorption was allowed to proceed from 2 to 150 days, and that extraction samples were collected at 2, 50, 100 and 150 days. Do you have this data available? In the report you provide only the 293 day desorption value. It would help my analysis very much if I could obtain more desorption values, ie for different times, whatever is available. I am interested only in Pu for natural waters, for all three minerals (silica, mont., hematite), if available.

3. In the report you present the dependence of K_d on colloidal concentration (table 16), after 240 days. Do you have any values for intermediate times (as given in Table 2) for colloidal concentration other than 0.2 g/L, in particular for low values (0.01, 0.05 g/L)?

Thanks in advance for taking your time and responding to these questions.
I look forward to receiving your response.

Sincerely

Vladimir Cvetkovic
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Dr. Ningping Lu
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Los Alamos, NM 87545
April 18, 2001

Dr. Vladimir Cvetkovic
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Dear Dr. Vladimir Cvetkovic:

Thank you very much for your e-mail and your comments. My answers to your questions in the e-mail are:

1. In the report of LA-UR-00-5121, the percentage Pu desorbed and the K_d were calculated based on the data collected from desorption experiments. In the report, I described the Desorption method as follows: "Desorption experiments were conducted in duplicate under the same conditions as the kinetics experiments. The ^{239}Pu -, ^{243}Am -, ^{237}Np -, and ^{238}U -loaded colloid particles were collected from the 240 hours sorption in the kinetics experiments. At the end of the sorption process, the colloid particles were separated from the liquid phase by centrifuging at 38,300 g for 2 hours. Most of the supernatant was carefully removed, and the wet weights of the colloids were obtained. The colloid samples were then extracted with 10 ml aliquots of the **radionuclide-free J-13** and SYN.J-13 water. Desorption of ^{239}Pu , ... was allowed to proceed 293 days for ^{239}Pu ,...". Here, the K_d values after desorption were calculated using the following equation in the radionuclide-free J-13 and SYN.J-13 water. The assumption was that desorption of actinides occurred in the radionuclide-free water when Pu-loaded colloids had migrated in to natural water system.

$$K_d\text{-value (ml g}^{-1}\text{)} = \{[(A_s + A_R) - A_d]/M\}/A_f$$

where A_s is the total Pu sorbed onto colloids, which obtained from sorption experiment; A_R is the total residual Pu remaining in the solution that was the residual solution after colloid particles were separated from solution at the end of sorption process; A_d is the total Pu desorbed in to water after 293 days of desorption; A_f is the amount of Pu per ml solution after desorption; and M (g) is the mass of colloids used in the desorption process. Therefor the K_d value after desorption was not estimated by your method described in your e-mail.

2. The exactly desorption data related to the article in Radiochim.Acta., 1998 are:

Percentage of Pu-239 Desorbed from Pu-loaded Colloids of Hematite and Goethite
as a Function of Time in Natural Groundwater

Time day	Hematite J.13.Pu(V) (%)	Hematite J.13.Pu(IV) (%)	Goethite J.13.Pu(V) (%)	Goethite J.13.Pu(IV) (%)
2	0.003 ± 0.002	0	0.1 ± 0.003	0.1 ± 0.02
50	0.01 ± 0.004	0	0.5 ± 0.01	0.3 ± 0.04
100	0.01 ± 0.003	0	0.7 ± 0.01	0.5 ± 0.04
150	0.02 ± 0.003	0	0.7 ± 0.02	0.6 ± 0.05

Percentage of Pu-239 Desorbed from Pu-loaded Colloids of smectite and Silica as a Function of Time in Natural Groundwater (the data did not published)

Time day	smectite J.13.Pu(V) (%)	smectite J.13.Pu(IV) (%)	Silica J.13.Pu(V) (%)	Silica J.13.Pu(IV) (%)
2	0.06 ± 0.01	3.2 ± 0.3	4.5 ± 1.1	9.3 ± 0.9
50	0.26 ± 0.01	5.3 ± 0.1	6.2 ± 1.0	12.8 ± 0.2
100	0.4 ± 0.003	6.9 ± 0.2	6.6 ± 0.9	14.7 ± 0.2
150	0.5 ± 0.03	8.3 ± 0.2	7.1 ± 0.9	16.2 ± 0.2

3. I have 4 hours sorption data for all colloidal concentrations of 0.01, 0.05, 0.1, 0.15, 0.2 and 1.0 g/L. I did not conducted time dependent test with colloid concentrations < 0.02 g/L.

Sincerely

Ningping Lu, Ph.D
Environmental soil chemistry