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CENTER FOR NUCLEAR WASTE REGULATORY ANALYSES

TRIP REPORT

SUBJECT: Trip report for Mineralogical Society of America (MSA) Short Course – Uranium: Minerals, Chemistry and the Environment, and Geological Society of America (GSA) Annual Meeting

DATE/PLACE: October 21-23, 1999 Golden, CO (MSA Short Course) 20.01402.158
October 24-28, 1999 Denver, CO (GSA Annual Meeting) 20.01402.158

AUTHOR: F. Paul Bertetti

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PERSONS PRESENT:

The MSA short course was well attended with about 80 presenters and registrants representing many nations and a variety of organizations. An attendance and contact list is attached. Personnel from both the CNWRA and NRC (P. Bertetti, B. Leslie, and L. Veblen) attended the course as students.

The annual GSA meeting is one of the foremost and well attended conferences in the geosciences. The meeting represents a broad range of disciplines and interests. Typically, the meeting has several hundred technical sessions given over the four to five day conference period. Session contents range from theme oriented subjects such as this year's group on the exploration and study of Mars, to general topic sessions in areas such as hydrology, geochemistry, paleontology, and geoscience education, to name just a few. This year's meeting was attended by over 6,000 people.

PURPOSE OF TRIP:

P. Bertetti attended the MSA short course on uranium and the GSA Annual meeting as part of his professional development for 1999.

SUMMARY OF ACTIVITIES:

The MSA short course on uranium was organized by Peter Burns (Univ. of Notre Dame) and Bob Finch (Argonne National Laboratory). The course addressed fundamental issues such as uranium crystal chemistry, systematic uranium mineralogy, aqueous chemistry of uranium and actinides, uranium-ore genesis, and isotopic systems. Other subjects like microbial influences on uranium chemistry, remediation of uranium contaminated sites, applications to radioactive waste disposal, and the natural fission reactors in Gabon, Africa were also included. The short course notes, published as a volume in the *Reviews in Mineralogy* series, were provided as part of the course materials. The review volume also includes chapters on a variety of analytical techniques for the study of uranium and uranium bearing minerals. A course agenda is attached.

The MSA short course lectures generally followed the structure and content of the material provided in the review volume. As expected, each lecture was well presented, with five lectures given each day. Overall, the

course subject material emphasized an understanding the behavior of uranium and uranium minerals in the environment. Most lectures focused on the fate of uranium with respect to the disposal of spent fuel and other nuclear wastes. The short course was supplemented by three dedicated technical sessions (two oral, one poster) at the GSA meeting the following week.

An example of interesting and relevant information that was disseminated during the course is provided by the materials presented by R. Finch and D. Wronkiewicz (Finch and Murakami, 1999; Wronkiewicz and Buck, 1999). Finch spoke at length about observed paragenesis of uranium ore mineralization and alteration in natural systems, while Wronkiewicz presented results of long duration uranium oxide and spent fuel dissolution experiments. These long term experiments show a progression of secondary phase uranium mineralization similar to that observed at the Nopal I mine at Peña Blanca, a natural analog for the proposed Yucca Mountain repository. The general sequence of alteration minerals includes schoepite, metaschoepite, boltwoodite, uranophane, and soddyite. Finch suggested there is some evidence of reversibility between schoepite and metaschoepite formation due to availability of water. Another key point was that the secondary phases will also likely influence the migration of actinides and fission products. Several of the secondary phase display an affinity for inclusion of elements such as Cs, Sr, Tc, and Np.

The short course also offered an opportunity for informal discussion with participating researchers. One significant source of information was Peter Burns' group at Notre Dame, where research has been focused on the characterization of uranium mineral crystal structures. To support this work, the group has succeeded in development of methods for synthesis of several uranyl phases including schoepite, boltwoodite, and uranophane. These phases are of interest because they are common alteration products of uraninite (and spent fuel), but their thermodynamic properties are not well known, in part due to the common difficulty of obtaining pure mineral separates from field samples. The exchange of information offered a potential solution to recent difficulties encountered in attempts to consistently synthesize uranophane for use in thermodynamic studies at CNWRA.

As previously mentioned, there are a great number of topical sessions at the annual GSA meeting each year, and many of these sessions contain presentations relevant to the disposal of HLW. Readers are encouraged to review the abstracts associated with the meeting to follow up on specific areas of interest. Three examples of research relevant to the HLW program include: research utilizing analytical techniques with improved spatial resolution, research describing secondary uranium mineral formation and behavior, and research investigating secondary uranium mineral phase (and other alteration minerals) influence on the migration of radionuclides.

In studies of natural analogs of the proposed HLW repository, one of the difficulties in characterization stems from the fine-grained nature of uranium secondary mineralization. Often, secondary phases such as schoepite, uranophane and boltwoodite are intergrown and very closely associated spatially, making chemical analyses (and possible dating of the rock) of the individual phases difficult. Some presentations offered new techniques that might begin to address these analytical scale problems. M. Fayek of UCLA (Fayek et al., 1999) reported on successes in using ion microprobe techniques to measure O and Pb isotopes in a uraninite. Specifically, Fayek suggested that spatial resolution on a scale of $15 \times 30\ \mu m^2$ was achievable with good resolution and accuracy. Ideally, similar techniques could be used to analyze natural samples from the Peña Blanca region. In a study of rare earth element concentrations in whole rocks, Patino et al. (1999) have developed techniques using laser ablation and ICP-MS to gather chemical compositional data over small sample areas ($\sim 20 \times 20\ \mu m^2$) and with very good detection limits (ppb range). Patino et al. used whole rock

glass disks also used for XRF but are extending their work to studies of individual mineral grains on polished sections.

The fate of uranium oxide (uraninite) in an oxidizing environment was discussed in several presentations. Wronkiewicz et al. (1999) discussed results of long duration experimental results on uranium oxide and spent fuel. He described evidence for incorporation of Np into schoepite and incorporation of Tc into the structure of uranophane. Additional information on secondary phase development was presented by Finch (1999). Finch suggested that although accumulation of Pb may indeed strain the host uraninite crystal structure, causing it to break apart, the production of Pb-bearing uranium minerals and the possibility of generating nucleation sites for other secondary U mineralization may have the effect of mitigating the overall release of U.

In related studies of secondary uranium minerals, several studies investigated the inclusion or retardation of radionuclides such as Cs, Sr, U, and Np as a result of mineral alteration. Burns et al. (1999) and Hill and Burns (1999) presented results for studies in which Sr and Cs were incorporated into uranyl silicates. In general, different alteration and secondary phases have differing affinities for elements. For example, schoepite seems to incorporate Np, whereas uranophane does not. Likewise, boltwoodite and uranophane readily incorporate Cs and Sr. Reeder et al. (1999) discussed the potential for uptake of U in calcite and aragonite. Differences in uptake between the two $CaCO_3$ minerals was attributed to differences in uranyl coordination within the crystal structures. Giammar and Hering (1999) presented preliminary results of U migration influences due to uranyl mineral dissolution and desorption from iron oxide. Of particular interest is the predicted formation and sequence of occurrence of secondary uranyl phases and other common alteration minerals and their influence on the migration of radionuclides. These results suggest that it may be important to account for source terms of various radionuclides by understanding the behavior of these alteration minerals.

CONCLUSIONS:

The MSA short course and Annual GSA meeting provided an opportunity to learn and exchange information with a number of researchers on important and relevant topics.

PROBLEMS ENCOUNTERED: None

PENDING ACTIONS: None

RECOMMENDATIONS:

The developments in analytical techniques for investigation and characterization of minerals at very small spatial scales should be pursued. DOE plans to use data from Peña Blanca to support their license application. These data are highly uncertain. By utilizing new characterization techniques, CNWRA and NRC can improve their understanding of uranium mineralization at Peña Blanca. This could be of benefit in resolving issues related to DOE source term estimates. The relationship between secondary uranium mineralization and the migration of radionuclides should also be investigated further. For instance, the tendency of radionuclides such as Np and Tc to be released (or retarded) during alteration of secondary uranium minerals (transition from schoepite to uranophane, for example) should be investigated. By using information on the synthesis of uranyl minerals and applying the appropriate experimental protocols, an improved understanding of retardation of important low-sorbing elements such as Tc and Np could be achieved. This would do much

to improve our understanding of the release and migration potential of these radionuclides, and would provide a basis for probing DOE assertions.

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ATTACHMENT 1

Short Lecture Schedule

URANIUM: MINERALS, CHEMISTRY AND THE ENVIRONMENT
Short Course Lecture Schedule

Friday, Oct. 22, 1999

9:00-10:00 Uranium Ore Deposits: Products of the Radioactive Earth	Jane Plant, Peter Simpson, Barry Smith & Brian Windley
10:00 - 10:30 *COFFEE BREAK*	
10:30 - 11:30 Stable Isotope Systematics of Uranium Deposits	Mostafa Fayek & T. Kurtis Kyser
11:30 - 12:30	T. Kurtis Kyser
12:30 - 1:1:30 *LUNCH*	
1:30 - 2:30 Mineralogy and Geochemistry of Natural Fission Reactors in Gabon	Janusz Janeczek
2:30 -3:00 Environmental Aqueous Geochemistry of Actinides	William M. Murphy & Everett L. Shock
3:00 - 4:00 *BREAK*	
4:00 - 5:00 Geomicrobiology of Uranium	Yohey Suzuki & Jillian F. Banfield

Saturday, Oct. 23, 1999

9:00-10:00 Uranium Contamination in the Subsurface: Characterization and Remediation	Abdessalam Abdelouas, Werner Lutze & H. Eric Nuttall
10:00 - 10:30 *COFFEE BREAK*	
10:30 - 11:30 The Crystal Chemistry of Uranium	Peter C. Burns
11:30 - 12:30 Systematics and Paragenesis of Uranium Minerals	Robert Finch & Takashi Murakami
12:30 - 1:1:30 *LUNCH*	
1:30 - 2:30 Uranium Mineralogy and the Geologic Disposal of Spent Nuclear Fuel	David Wronkiewicz & Edgar Buck
2:30 -3:00 *BREAK*	
3:00 - 4:00 Radioactivity and the 20th Century	Rodney C. Ewing
4:00 - 5:00 *OPEN DISCUSSION*	

Saturday Night Banquet: 7:00 PM

ATTACHMENT 2

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