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BOOK REVIEW

URANIUM: MINERALOGY, GEOCHEMISTRY AND THE ENVIRONMENT. Peter C. Burns and Robert Finch, Editors, Reviews in Mineralogy, 1999, Vol. 38, 679 p. Mineralogical Society of America, Washington, D.C. $32 ($24 for MSA members).

Uranium: Mineralogy, Geochemistry and the Environment is based on a short course offered on October 22 and 23, 1999. This tome represents a uniquely valuable and generally up-to-date source of information on uranium mineralogy, ore deposits, the geomicrobiology of uranium, radioactive wastes, and chemical, isotopic and other analytical methods. For those interested in these topics, the book should become an essential reference. The fourteen chapters and their authors include (1) *Radioactivity and the 20th Century*, R.C. Ewing; (2) *The Crystal Chemistry of Uranium*, P. C. Burns; (3) *Systematics and Paragenesis of Uranium Minerals*, R. Finch and T. Murakami; (4) *Stable Isotope Geochemistry of Uranium Deposits*, M. Fayek, and T.K. Kyser; (5) *Environmental Aqueous Geochemistry of Actinides,* W.M. Murphy and E.L.Shock; (6) *Uranium Ore Deposits: Products of the Radioactive Earth*, J. Plant, P.R. Simpson, B. Smith and B.F. Windley; (7) *Mineralogy and Geochemistry of Natural Fission Reactors in Gabon*, J. Janeczek; (8) *Geomicrobiology of Uranium*, Y. Suzuki and J.F. Banfield; (9) *Uranium Contamination in the Subsurface: Characterization and Remediation*, A. Abdelouas, W. Lutze and H.E. Nutall; (10) *Uranium Mineralogy and the Geologic Disposal of Spent Nuclear Fuel*, D. Wronkiewicz and E. Buck; (11) *Spectroscopic Techniques Applied to Uranium in Minerals*, J.M. Hanchar; (12) *Infrared Spectroscopy and Thermal Analysis of the Uranyl Minerals*, J. Cejka; (13) *Analytical Methods for the Determination of Uranium in Geological and Environmental Materials*, S.F. Wolf; and (14) *Identification of Uranium-bearing Minerals and Inorganic Phases by X-ray Powder Diffraction,* F.C. Hill.

In the Preface the editors note that “more than five percent of minerals known today contain uranium as an essential constituent.” This number has exploded through time from two (pitchblende and torbernite) in 1800, to 20 in 1900, 70 in 1950 and 135 in 1978 (Steacy and Kaiman 1978) to about 200 in 1995. Since the 1970s, the purpose of study of uranium mineralogy and geochemistry has shifted from exploration for and characterization of uranium ore deposits (cf. Kimberley 1978), to environmental and human health concerns related to the disposal of uranium mine and mill tailings and of nuclear wastes.

Chapter 1 is an excellent introduction to the book and will appeal to the widest audience. Rod Ewing tells the fascinating history of the isolation of elemental uranium (by Peligot in 1841), the discovery of radioactivity (by Henri Becquerel in 1896), the law or radioactive decay (Rutherford and Soddy 1902) and radiometric dating of the earth, and the concept of secular equilibrium (Boltwood 1905). Also addressed are nuclear weapons, nuclear reactors (of which there are about 1000 globally, and growing), and ongoing issues related to the disposal of nuclear wastes, environmental radioactivity and contamination.

Chapters 2 and 3 offer an exhaustive, updated view of uranium mineral crystal chemistry and mineral paragenesis for specialists in the field. These chapters update and expand on the earlier work of Frondel (1958) and Smith (1984). In Chapter 2, Burns notes that crystal structures have been determined and refined for only about a third of uranium minerals. Finch and Murakami observe in Chapter 3 that the uranyl silicate uranophane is possibly the most common U mineral after uraninite, as well as being a likely alteration product of spent nuclear fuel (which is chiefly UO_2) at the potential Yucca Mountain geological repository in Nevada.

Finch and Murakami (Chapter 3) report measured and estimated thermodynamic data for 14 uranium silicate and carbonate minerals. They then present solubility calculations and phase diagrams, and relate these diagrams to the occurrence of natural uranyl minerals and the probable occurrence of the same or similar minerals from the alteration of nuclear wastes. Clearly, a great deal remains to be done to obtain reliable thermodynamic and solubility data on uranium minerals. Such data is quite limited and most is of questionable reliability. Interested readers will find an exhaustive bibliography of what has been published on this subject elsewhere (see Grenthe et al.1995).

Fayek and Kyser (Chapter 4) have written an excellent introduction to stable isotope geochemistry and then have applied it to explain the origin and paragenesis of uranium deposits. They consider all important deposit types in this well written chapter.

In Chapter 5, Murphy and Shock give a useful overview of actinide geochemistry, but don’t say enough about uranium, the topic of the book. The authors perpetuate errors in Grenthe et al. (1992) by greatly overestimating the stability of the complexes $UO_2(OH)_2^0$, $U(OH)_4^0$, and $U(OH)_5^-$ in their calculations as did Shock et al. (1997), although these errors were acknowledged by Grenthe et al. (1995), and are admitted by the authors in their appendix (cf. Langmuir 1997). Thus, their phase diagrams and discussion involving these species are of questionable value. No natural water compositions are discussed or plotted on the mineral phase diagrams given by Murphy and Shock, or by Finch and Takashi (Chapter 3) making those diagrams of limited use to readers. The value of Chapter 5 is in its development of high temperature thermodynamic properties for Am species using the revised HKF equation of state (cf. Shock et al. 1992). A follow-up paper on americium and uranium, cited as Shock et al. (2000) had not been published as of January 2001.

Chapter 6 by Plant et al., which details at length the origin and history of uranium ore deposits, is nicely complemented in the book by Chapter 2 with its discussion of the paragenesis of uranium minerals and Chapter 4 on stable isotopes in uranium deposits. Plant et al. note that the unconformity-related deposits (e.g., those of the Athabasca Basin, Canada, and northern Australia) include the largest high-grade deposits in the world. The sandstone-type deposits among which are the Oklo natural fission reactors of Gabon and the roll front deposits of the western United States are economically second in importance.

In Chapter 7, Janeczek has probably written the definitive report on what is known about the history, mineralogy and geochemistry of the unique natural fission reactors at Oklo, Gabon, Africa. He notes that there are (or were) 15 natural fission reactors at Oklo. These are "zoned bodies...of high-grade U ore (the reactor core) enveloped by a mantle of clay minerals," making them physically analogous to proposed nuclear waste burial sites below the water table that would surround spent fuel with a bentonite clay backfill. Interestingly, migration of radionuclides (e.g., U, Pu, Np, and Tc) and fission products from the Oklo reactors from the time of primary mineralization 2 billion years ago, has been limited to a distance of from a few tens of cm to 10 m.

In Chapter 8 on the microbiology of uranium, Suzuki and Banfield note that the actinides including U are hard acid cations, and therefore they bond most strongly with hard acid functional groups on and in biomolecules. Uranium (as uranyl), which has no essential biological function, is adsorbed in minutes by cells, and in acid waters is 20 to 40 times more toxic to cells than Cu or Ni, although U toxicity decreases with continued exposure.

Chapters 9 and 10 deal with actual and potential environmental contamination resulting from uranium mill tailings, and as a consequence of the geological disposal of spent nuclear fuel. Abdelouas et al. (Chapter 9) focus on subsurface contamination derived from uranium mill tailings, which retain about 85% of the radioactivity of the ore. They begin with a global view of the problem (there are more than 230 and 300 million tons of U mill tailings in the U.S. and Canada, respectively). They then focus on their own studies of groundwater contamination by tailings at Tuba City, AZ, and propose the application of in-situ remedial methods involving reduction, which they note are far more cost-effective than pump and treat methods.

Wronkiewicz and Buck (Chapter 10) discuss the history of the radionuclides and fission products associated with spent nuclear fuel as a function of time after geological disposal. They also examine the effects of water and an oxidized environment on the paragenetic sequence of alteration products which in an oxidizing environment are likely to begin as hydrated uranyl hydroxide phases, and later become uranyl silicates such as uranophane and boltwoodite. These authors note that the general absence of V, P, As, Pb, and CO_3 in spent fuel reduces the number of possible alteration mineral phases to about 40. The Nopal I uranium deposit in the Pena Blanca Mining District of Mexico is considered the best analog for the proposed nuclear waste disposal site at Yucca Mountain, NV. At this deposit, uranophane is apparently the long-term alteration product of weathered uraninite.

Chapters 11–14 are in-depth and up-to-date reports that describe methods of identification of uranium and uranium-bearing minerals and their chemical and isotopic analysis. Hanchar (Chapter 11) details the state of the art applications of spectroscopic techniques including EXAFS, XANES, XAS, and XPS to uranium mineral identification and characterization. These methods allow the investigator to determine for example, the oxidation state of U and the speciation of adsorbed U. In Chapter 12, Cejka offers IR spectra and thermogravimetric data for uranyl minerals, including some data that have not been previously published. He discusses a wide variety of thermal analytical methods and presents results for many U minerals. Wolf (Chapter 13) fairly exhaustively describes the many analytical methods that have been used for the determination of U in geological and environmental materials. Early applied methods of U analysis included AA spectroscopy and fluorimetry. Isotopic ratios may be determined using mass spectroscopy. Extremely low detection limits have recently been obtained by FI-ICPMS (0.3 pg/mL or 0.3 μg/L) and high resolution ICPMS (<1 fg/mL or $<10^{-7}$ μg/L). In Chapter 14, Hill points out that the unusual size of the U atom results in intense X-ray diffraction peaks for U-bearing minerals. He notes that disparities among published X-ray powder diffraction results for U minerals often reflect differences in sample preparation. Hill presents 15 tables of calculated *d*-spacings for the 5 most intense diffraction peaks along with lattice parameters and space group symbols for 136 uranium-bearing phases that include 64 minerals.

In this book, the individual authors have with few exceptions created uniquely up-to-date and critical reports in their respective areas, which are exhaustively referenced. The book is an excellent reference source, particularly in areas of mineralogy, ore deposits and chemical and isotopic analytical methods. It contains many valuable tables related to uranium crystal chemistry and mineralogy. However, the chapters largely stand alone, rarely cite each other, and disagree (without acknowledging it) on such topics as the selection of thermodynamic data for uranium. Unfortunately, the book does not offer a critical review of the uranium geochemistry of natural waters. Editorial errors are few. The most notable are "words" such as "Pbing," "misPbing" and "transU" in Chapter 7, undoubtedly a result of the "find and replace" word-processing command.

Regardless, the book is an outstanding, one-of-a-kind reference work, particularly because of its thorough, updated coverage of uranium mineralogy, ore deposits and analytical methods, all of which are considered in multiple chapters. No other book comes close to its coverage and currency in these areas. Single chapters on radioactivity and the 20th century, the geomicrobiology of uranium, uranium contamination of the subsurface, and the geological disposal of spent fuel are also well written and highly informative. Given its bargain price and unique content, the book should be on the shelf of all of those interested in uranium mineralogy and geochemistry.

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