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Mineralogical and Geochemical Aspects of the Disposal of Nuclear-Fuel Waste

PREFACE

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In Canada, investigations into the safe disposal of radioactive waste, produced in the generation of nuclear power, began in the late 1950s with the formulation and production of a leach-resistant nepheline syenite glass. When nuclear generating-systems came into full operation in the 1970s, renewed interest in this topic led to the establishment of the Canadian Nuclear Fuel Waste Management Program by Atomic Energy of Canada Limited (AECL). This federally funded program encompasses research into the immobilization of intact bundles of used fuel, formulation of advanced waste-forms for chemically separated radionuclides, construction of a disposal vault in plutonic rock, radionuclide transport in the geosphere and food chain, and assessment of the environment impact of nuclear-waste disposal. In addition, other organizations direct programs dealing with uranium-mine tailings and with nuclear-reactor wastes characterized by low- to medium-level activity.

To bring together researchers involved in these studies and scientists in the geochemical discipline, a special half-day symposium dealing with the mineralogical and geochemical aspects of nuclear-fuel-waste disposal was held in conjunction with the annual GAC/MAC conference in Winnipeg in May of 1982. The symposium was organized by Dr. F.P. Sargent of AECL's Whiteshell Nuclear Research Establishment (WNRE) at Pinawa, Manitoba, and sponsored by the Mineralogical Association of Canada. Eight papers were presented in a session ably chaired by Dr. G.M. Anderson of the University of Toronto. Five representative papers from this symposium are published in this issue of *The Canadian Mineralogist.* They illustrate the research typical of that being carried out to minimize the environmental impact of the use of nuclear power.

The first paper deals with the environmental impact of uranium mining. Low concentrations of uranium and its daughter products, in particular radium, remain in discarded mine-tailings. Murray, Brown, Fyfe and Kronberg report on a study carried out at the University of Western Ontario on the use of phosphate to stabilize these elements, based on the observation that naturally occurring apatite has the ability to retain actinides. The authors show that extremely low leach-rates for radium can be obtained by treating mine tailings and mill-discharge wastes with a potassium phosphate solution. This work suggests that the potential environmental impact of mine tailings can be alleviated by a relatively simple chemical process.

Development of an advanced waste-form, a glass-ceramic, is discussed in the paper by Hayward, Doern, Cecchetto and Mitchell. This work, carried out at WNRE, is based on observations that naturally occurring titanite is able to accept a wide variety of foreign ions in its structure. Using thermodynamic calculations, the authors show that titanite is expected to be stable with respect to the groundwater compositions expected in a deep disposal-vault in the Canadian Shield. They produced a glass-ceramic, a partly crystalline material (containing 0.5–5 μm titanite crystals in an aluminosilicate glass matrix) by controlled crystallization in the system Na_2O–Al_2O_3–CaO–TiO_2–SiO_2 and studied its resistance to leaching. Their results indicate that this promising waste-form is very durable.

The role of fracture-filling material in a plutonic rock-formation in retaining radionuclides was discussed by Kamineni, Vandergraaf and Ticknor. This study, carried out jointly at WNRE and the Geological Survey of Canada, involved the sorption of various radionuclides from solutions on petrographically analyzed thin sections of plutonic rock containing alteration minerals and fracture-infilling materials. The extent of sorption was determined qualitatively by autoradiography. The results confirm the ability of titanite and apatite to sorb actinides and demonstrate the enhanced ability of alteration and fracture-filling materials to retard the movement of radionuclides toward the biosphere. Even technetium, which exists under normal laboratory conditions as a nonsorbing anionic species, was found to sorb on some minerals.

Nonsorbing radionuclides (especially ^{129}I) are not retained by minerals in a rock formation. However, they may diffuse into micropores and fissures in the rock. Consequently, an understanding of the micropore structure of plutonic rocks and the effects of alteration is needed to quantify the diffusional effects for these nuclides. Katsube and Kamineni of the Geological Survey of Canada report their investigations pertaining to the pore structure and alteration effects of samples taken at various depths from a highly fractured pluton near Atikokan, Ontario. They note a general increase in pore tortuosity with decrease in degree of alteration and a corresponding decrease in connecting porosity and permeability. These phenomena are explained in terms of the mass transport of dissolved matrix-material to increase pocket porosity and reduce overall permeability.

Information on the behavior of trace elements and, in particular, naturally occurring radionuclides in the geosphere can be used in a predictive analysis of their behavior in and around a disposal vault. The geochemistry associated with the formation of uranium deposits can often be used in this respect. In the final paper of this series, Lobato, Forman, Fuzikawa, Fyfe and Kerrich discuss various processes that may have led to the establishment of uranium in ore-grade zones in regionally developed granite gneiss in Brazil. These include oxidation–reduction reactions, selective dissolution and precipitation of individual elements. A partial depletion of uranium from granulitic rocks and a subsequent enrichment of the metasomatic rocks appear to have been caused by fluids under oxidizing conditions, during tectonic processes that raised the basement rocks to the surface. This enrichment was accompanied by an enrichment in the rare-earth elements, lead and vanadium. Unless similar conditions can be ruled out in or around a waste-disposal vault, actinide transport must indeed be considered.

These five topics then, from mine tailings to uranium deposits, reflect some of the many mineralogical and geochemical aspects of the program on nuclear-fuel-waste disposal. By publishing these papers together in this issue of *The Canadian Mineralogist*, the editor is providing a wider forum to those of us involved in this vital program; on behalf of the authors, I thank him and his staff for making this possible.