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ORNL

FOREIGN TRIP REPORT

229774

Date: September 26, 2005

Subject: Report of Foreign Travel to Russia — Team Report for M. Jonathan Haire, TA 229774; Charles W. Forsberg, TA 229335; and Les R. Dole, TA 230522

To: John L. Wengle, DOE Office of Radioactive Waste, Office of Science and Technology (RW-40)

From: M. Jonathan Haire, Charles W. Forsberg, and Les R. Dole

Purpose: The purpose of this foreign travel was to attend and participate in the 10th International Conference on Environmental Remediation and Radioactive Waste Management, ICEM 2005, in Glasgow, Scotland. Also, Oak Ridge National Laboratory (ORNL) and the All-Russian Scientific Research Institute of Experimental Physics (VNIIEF) had organized Conference Session 45 on depleted uranium (DU) reuse. Papers were presented at this session as a review of the progress on International Science and Technology Center (ISTC) Projects 2691, 2693, and 2694.

The primary objective was to learn more about the geologic repository plans of other countries and, in particular, the Russian work on sorption of radionuclides onto uranium dioxide (UO_2). Additionally, the Russians are conducting experiments concerning advanced DU composite shielding materials for casks, and the United States needs to stay abreast of this research.

Travel Dates: September 2–9, 2005

Summary: The Russians (Dr. Elena V. Zakharova et al.) reported that preliminary experimental results show that neptunium, Np(V) and Np(IV), are strongly sorbed onto UO_2. If true, this would lower the calculated site boundary radiation dose by a factor of ~10 at the U.S. Yucca Mountain (YM) spent nuclear fuel (SNF) repository. Early results show that technetium, Tc(IV), is also sorbed. To our knowledge, these are the first data on sorption of Np and Tc onto UO_2. These results could support the licensing basis of YM and may ultimately simplify repository design and operation by avoiding the need for various expensive engineered barriers such as the drip shield. The Russians (Drs. Vataliy T. Gotovchikov, Victor A. Seredenko, Vitaly Z. Matveev et al.) presented laboratory experimental data for new DU composite materials intended

for SNF transport and storage cask shielding. Early calculations show that the capacity of SNF casks may be nearly doubled with the use of these new materials, thus reducing by 50% the number of lag storage casks needed at YM repository.

Background

The 10th International Conference on Environmental Remediation and Radioactive Waste Management, ICEM05, was jointly sponsored by the American Society of Mechanical Engineers (ASME), the Institute of Mechanical Engineers and the British Nuclear Energy Society. The ASME meetings are held every two years in countries outside the United States. This year there were over 700 registrants, 150 more than attended 2 years ago. Twenty-nine countries sent representatives to the 61 sessions, at which over 300 papers were presented. Over 40 companies had exhibits or sponsored events. This indicates a high international interest in the conference subject matter. The ICEM07 conference is scheduled for Bruges, Belgium, in September 2007.

Through the ISTC, the U.S. Department of State is funding three projects in Russia to develop beneficial uses of DU: ISTC Project 2691 ("Production and Testing of Heavy Concretes Including Depleted Uranium Dioxide Concerning Their Use as a Shielding Material in Construction of Casks for Spent Nuclear Fuel"), ISTC Project 2693 ("Production and Testing of Cast Cermet on the Base of Stainless Steel and Depleted Uranium as Applied to Its Use as Shielding Materials in Construction of Casks for Spent Nuclear Fuel and Radioactive Wastes"), and ISTC Project 2694 ("Interaction Between Depleted Uranium Dioxide and Underground Waters and Radionuclides Sorption by Oxides and Hydroxides of Depleted Uranium"). The travelers are the U.S. technical contacts for these ISTC projects. M. J. Haire presented a paper giving an overview of these ISTC projects, "Collaborative Research and Development on Uses of Depleted Uranium." The travelers presented five papers at this conference. Travel expenses were paid by the U.S. Department of Energy, Division of Radioactive Waste (RW), Office of Science and Technology (RW-40).

Sorption of Radionuclides by UO_2

Dr. Elena Zakharova (with M. J. Haire et al.) presented the paper "Sorption of Long-Living Radionuclides (^{237}Np and ^{99}Tc) from Yucca Mountain Repository Ground Waters by Depleted Uranium Dioxide." Important points from this paper are as follows:

1. The primary contributor to long-term (>30,000-year) dose at YM geologic repository is ^{237}Np. Russian experiments show that Np(V, IV) is strongly sorbed onto UO_2. In experiments, Np(IV) equilibrium is reached within 2 hours; Np(V) equilibrium is reached within 24 hours. The amount of Np(V) sorption is determined by the degree of deprotonation of the OH-group surface. Neptunium(V) sorption onto UO_2 is practically irreversible. Sorption of Np(V) is accompanied by partial reduction of Np(V) to Np(IV). The sorption of Np(V) onto shoepite (the primary product of UO_2 corrosion) is less than the sorption of Np(V) onto UO_2.

2. Technetium and iodine are the primary contributors to the YM site boundary dose at short (<30,000-year) times. Technetium-99 sorbs onto UO_2. Early Russian work on this subject is continuing. To our knowledge, these are the first data for sorption of Np and Tc onto UO_2. The sorption of Np onto uranium oxides potentially suggests that there will be a lower

release of Np and Tc from the repository and that repository performance is better than current models predict.

3. The solubility of UO_2 in deionized water and YM J-13 simulated water was determined by the U(VI) content in the surface layer and by the uranium-to-oxygen ratio. It was found that the solubility of UO_2 formed at lower temperatures (~700°C) is 10 times higher than that at higher temperatures (~800°C).

4. The filtration properties of radionuclides through UO_2 show that the liquid flows slowly and then stops almost completely.

ISTC Project 2694, as currently funded, ends in November 2005. Because of the very encouraging and significant results of the first two years, this work should be extended to further explore and understand the potential benefits for the repository.

Depleted Uranium Dioxide Concrete as a Radiation Shielding Material

ISTC Project 2691**, "**Production and Testing of Heavy Concretes including Depleted Uranium Dioxide Concerning Their Use as Shielding Materials in Construction of Casks for Spent Nuclear Fuel," was reviewed in three papers in Session 45 of the 10th International Conference on Environmental Remediation and Radioactive Waste Management, ICEM 2005. Two papers by Dole, Ferrada, and Mattus reviewed the economic incentives to use DU in steel and concrete storage and transport casks and the results of two years of durability testing at ORNL: "Uses of Tails from Nuclear Fuel Production in Aggregates for Uses in Spent Nuclear Fuel Concrete-Steel Transport and Disposal Casks" and "Durability of Depleted Uranium Aggregates in DUCRETE Shielding Applications."

Serge Ermichev (with L. R. Dole et al.) submitted a paper summarizing the Russian Federation (RF) work in developing improved DU aggregates and concretes: "Optimization of Composition and Production Technology of High-Density Concrete with Ceramic Aggregate Based on Depleted Uranium Dioxide." This paper showed significant improvements in the economics of the DUAGG (depleted uranium aggregate) process and superior performance characteristics of its products. The RF contribution has greatly increased the likelihood that this technology will be used by the RF and United States.

The Russians have greatly improved the U.S. DUAGG formulation process and have enhanced the understanding of the associated materials science, as reported in a paper written by S. G. Ermichev. Experiments have been conducted to measure DUAGG density, uranium-to-oxygen ratio, compressive strength, porosity, chemical stability via leaching testing, and structural analysis. DU concrete with high compressive strength (~670 kg force/cm^2) and high density (~6.4 gm/cm^3) has been produced. This heavy concrete conforms to the specifications used in manufacturing International Atomic Energy Agency concrete-steel casks for spent fuel transport and storage.

Depleted Uranium Dioxide–Steel Cermets as a Radiation Shielding Material

DU oxide–steel cermets for SNF casks and other applications are being investigated in the United States and Russia, with each group investigating alternative fabrication methods to determine which method or combination of methods minimizes costs.

The traveler (C. Forsberg) presented the paper “Powder-Metallurgy Experiments and Manufacturing Studies on DUO_2-Steel Cermets for Spent Nuclear Fuel Casks,” which he coauthored with T. N. Tiegs and V. K. Sikka. The paper describes a new method for cermet cask manufacturing that reduces the capital and research costs associated with the fabrication process.

The Russians (Vitaly T. Gotovchikov, “Production and Testing of Cast Cermet on the Base of Stainless/Carbon Steel and Depleted Uranium Dioxide as Applied to Its Use as Shielding Materials in the Construction of Casks for Spent Nuclear Fuel and Radioactive Wastes”) reported that samples of DU oxide/steel cermets have been produced by casting and subsequently examined. The initial casting process failed to produce high-quality cermet. However, the Russian researchers have developed a successful process with large (~1-mm-diam) UO_2 particulates of high density mixed with stainless steel (density of >7.8 g/cm^3) in an ~50% volume ratio and determined that they can produce cermets with a density of about 9 g/cm^3. Cermets of at least 9.2-g/cm^3 density are possible. Microanalysis of the UO_2-steel interface needs to be made to determine its composition. The modified process heats the steel significantly above its melting point to improve bonding between the UO_2 surfaces and the steel.

The Russians [Vitaly T. Gotovchikov, “Use of an Induction Cold Crucible Melter to Manufacture Depleted Uranium Dioxide (DUO_2) Particles for Use in Spent Nuclear Fuel Casks”] reported on their initial experiments with the goal to produce low-cost, high-density DUO_2 particulates by melting. If successful, the process will produce the low-cost, high-density DUO_2 required for both the powder metallurgy and casting cermet processes described above. Initial nonradioactive tests have melted ZrO_2 at ~2700ºC. These tests will be followed by DUO_2, with its melting point of 2780ºC.

General Comments

It will be several years before any country has an operational geologic repository for SNF. Many countries have plans for repositories becoming operational in 5–60 years. This will present an accountability and security problem.

Travel Schedule

September 2–3, 2005	Air travel from Oak Ridge, Tennessee, to Glasgow, Scotland, United Kingdom
September 4, 2005	ICEM conference begins
September 4–7, 2005	Attend conference
September 7, 2005	ICEM conference ends
September 8, 2005	Air travel from Glasgow, Scotland, to Oak Ridge, Tennessee

C. W. Forsberg (one of the three travelers) also attended a DOE/NE GenIV meeting in the Czech Republic. He flew from Glasgow to Prague on September 8, attended meetings on September 9, and returned to Oak Ridge, Tennessee, on September 10. The details of this travel are contained in trip report 229335.

Major Persons Contacted

Elena V. Zakharova	IPC, Head of Laboratory, an experimentalist in ISTC Project 2694
Victor A. Seredenko	VNIIKhT, Division Director, Institute of Chemical Technology, Russian Academy of Sciences
Vitaly T. Gotovchikov	VNIIKhT, Head of Laboratory [PI for DOE induction cold crucible melter (ICCM) project]

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