ICEM'05:
The 10th International Conference on Environmental Remediation and Radioactive Waste Management
September 4–8, 2005, Scottish Exhibition & Conference Centre, Glasgow, Scotland

ICEM05-1387

U.S.–RUSSIAN COLLABORATIVE RESEARCH AND DEVELOPMENT ON USES OF DEPLETED URANIUM

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ABSTRACT

Oak Ridge National Laboratory (ORNL) began investigating potential beneficial uses of surplus depleted uranium (DU) in 1998. This work followed U.S. Congressional legislation, Environmental Impact Statements, and subsequent roadmaps related to the DU Hexafluoride (DUF_6) Disposition Project. ORNL's work is now focused on (1) the use of DU in a geologic repository to enhance repository performance and (2) development, testing, licensing, and deployment of new DU composite materials for spent nuclear fuel and high-level-waste storage and transport casks.

In 2003 the United States and Russia began collaborating on developing beneficial uses of DU through the International Science and Technology Center. The U.S. Department of Energy funded additional development work in Russia in September 2004 and August 2005. The work is centered in the Russian Academy of Sciences Institutes (the Institute of Physical Chemistry, the Institute of Chemical Technology, and the Vernadsky Institute of Geochemistry and Analytical Chemistry) and Rosatom Institutes (Research Institute of Experimental Physics—VNIIEF). The Russian private company developing DU casks is MSZ Electrostal.

This paper describes the U.S.–Russian collaboration program to develop beneficial uses of DU that are capable of consuming the entire inventory of DU in both countries.

INTRODUCTION

In 1994 the United States Department of Energy (DOE) initiated a program to reconsider its approach to long-term management of surplus depleted uranium hexafluoride (DUF_6) in storage. DOE requested recommendations for alternative management strategies and uses for DU from a variety of organizations and from the general public. A Programmatic Environmental Impact Statement was written. Alternative management strategies were recommended, among them being the development of beneficial uses of DU. Hundreds of beneficial uses of DU were identified. Research was conducted on novel uses that take advantage of the unique electronic and chemical properties of uranium, such as the use of DU as catalysts, in semiconductors, in fuel cells, etc. [1–3]. In 2003, research became focused on uses that have the potential to consume a significant portion of the stored inventory of DU. These include the use of DU as a chemical barrier in spent nuclear fuel (SNF) geologic repositories and the use of DU to provide shielding for SNF and high-level-waste (HLW) storage and transport casks. Through the Oak Ridge National Laboratory (ORNL), DOE began to collaborate with the Russian Academy of Sciences and Rosatom in 2003.

The United States and Russia are presently developing a next-generation SNF HLW cask that will have smaller dimensions, weigh less, be capable of higher heat loadings, be more resistant to terrorist assault, and be more proliferation resistant. These characteristics are enabled, in large part, by new materials. These newly developed materials include DU oxides in concrete (DUCRETE™) and DUO_2–steel cermets. ORNL, the lead in this endeavor, conducts materials development and testing while Russia is developing the composition and laboratory technology for DU aggregates (DUAGG™) and DUO_2–steel cermet production; investigating operational and technological features; and manufacturing demonstration samples.

U.S. RESEARCH

The U.S. government has more than 500,000 metric tons of surplus DU stored at DOE sites across the country. This material is mostly DUF_6 that resulted from uranium enrichment operations. On August 2, 1999, DOE issued a *Record of Decision (ROD) for Long-Term Management and Uses of Depleted Uranium Hexafluoride* [4]. This ROD indicated that DOE has decided to promptly convert the DUF_6 inventory to a more stable compound. Construction began in July 2004 on two plants to convert the DUF_6 into uranium oxides. Developing beneficial reuses of DUF_6 in the United States is the responsibility of the DOE-sponsored DU Uses Research and Development Program [5, 6] located at ORNL.

Repository Applications

DU could be disposed of beneficially in a geologic repository via a variety of means, as shown in Fig. 1 [7]. In these applications the chemical, nuclear, and geochemical characteristics of DU oxide are used to improve repository performance and provide a beneficial disposal method. Unlike nuclear reactors, SNF and HLW repositories will be constructed progressively over the lifetime of the facility. Consequently, it is expected that the design of the repository will change as new technologies, such as the use of DU in various beneficial applications, become available. It is expected that repository licensing amendments will reflect those changes.

Waste Packages

There are several promising options for manufacture and use of storage, transport, and disposal casks made with DUO_2 as a major shielding component. One is as DUCRETE^{TM}, a DUO_2 aggregate that would replace stone in concrete and that would be placed in a steel annulus to form a cask. Advantages are high shielding efficiency, estimated low cost of fabrication, and ease of production. A second option is as DUO_2 cermet [8], which is a physical mixture of DUO_2 and a metal such as steel. Advantages are high shielding efficiency, high thermal conductivity, estimated low cost of fabrication, and ease of production.

Richards Barrier

A Richards Barrier is a special type of backfill that uses the difference in permeability and capillary properties of two backfill materials to divert groundwater. A Richards barrier made of DUO_2 has the advantages of (1) providing protection of the waste packages (WPs) from potential rockfall; (2) saturating the water coming into the waste repository with DUO_2, thus inhibiting dissolution of the UO_2 in the spent fuel; (3) providing a sorbent for possible radionuclides released from the spent fuel, especially neptunium and technetium; and (4) providing a barrier to lava flow in the drifts and subsequent expulsion of radionuclides into the atmosphere in the unlikely event of a volcanic incident.

Fill Between Spent Fuel Pins

By filling the space between spent fuel pins in the WPs with DUO_2 particulates, dissolution of the UO_2 in the spent fuel would be inhibited, because incident water would already be saturated [9–11]. In addition, the DUO_2 would act as a sorbent for possible radionuclides released from the spent fuel, especially Neptunium.

Invert

If made of DUO_2, the invert would act as a sorbent for possible radionuclide release from the drift.

Shielding Applications

Heavy Concrete for Spent Fuel and Waste Storage

Heavy concrete is standard concrete in which conventional aggregate (typically gravel) is replaced with aggregate composed of a dense material such as DUO_2. Although the manufacture of heavy concrete for a given application is expected to be more expensive than standard concrete, there are circumstances where the reduced volume of the heavy concrete is expected to yield compensating cost reductions or other advantages. This is so because the reduced volume of the cask walls makes available more space for spent fuel storage in the cask. The purpose of this task is to bring heavy concrete technology to the point where a demonstrated technical basis for deployment exists. In particular, there is a need to establish: (1) the ability to manufacture large, heavy DUAGG™ concrete shapes; (2) the cost of such shapes; and (3) the chemical/physical stability of the DU aggregate (DUAGG^{TM}) [12]. This information is needed to underpin regulatory and purchaser confidence. Figure 2 [13] shows the much smaller cross-sectional area required for a DUCRETE^{TM} cask with the same shielding characteristics as the conventional concrete cask.

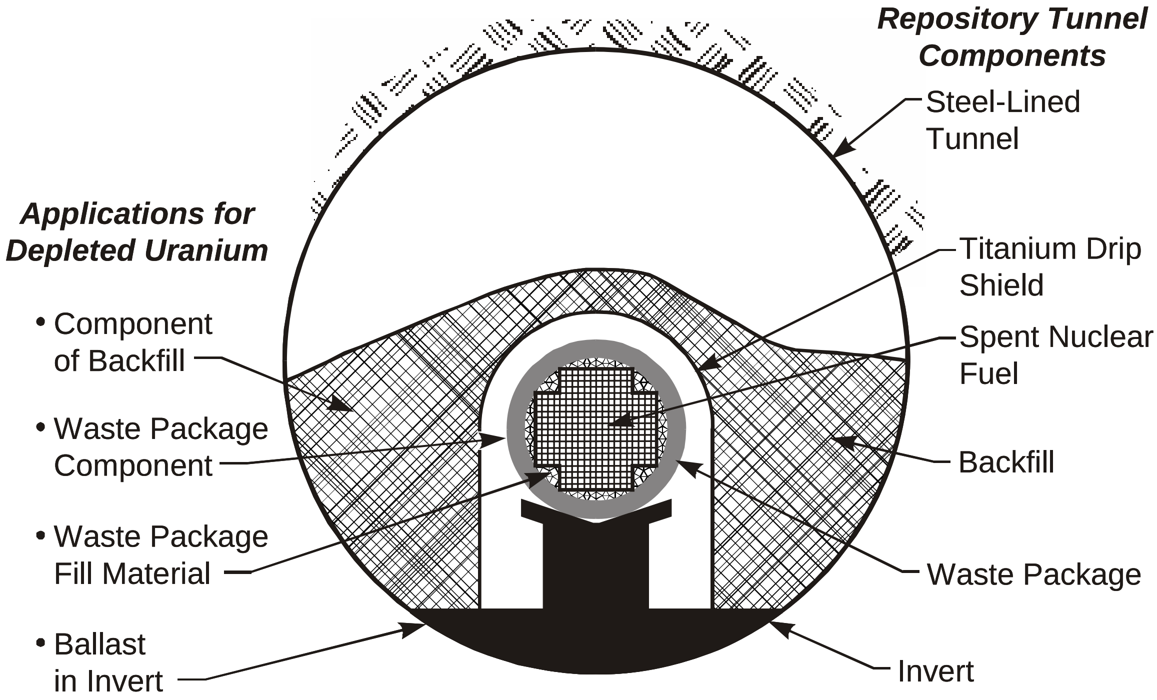

Fig. 1. Potential depleted uranium dioxide applications in a spent nuclear fuel geologic repository.

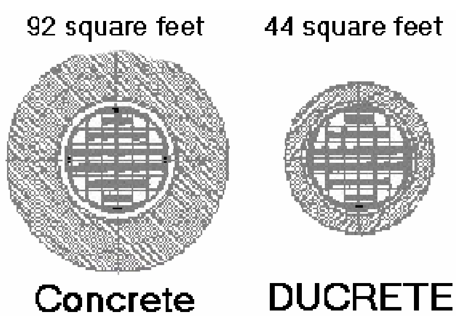

Fig. 2. Comparative diameters of concrete and DUCRETE™ dry-storage cask or silo. Using DUCRETE™ in an SNF cask or silo reduces the weight by 30%, the footprint by 50%, and the diameter from 132 in. (3.5 m) to 90 in. (2.3 m).

The corrosion of DU aggregate (DUAGG™) pellets to replace stone aggregate in concrete has been studied [14, 15] in 27-month-long experiments at ORNL at three temperatures (20, 67, and 150°C) in de-ionized water (DI) water; 1 *N* NaOH solution; and saturated cement pore solution made from (1) ordinary portland cement (OPC) and (2) a mix of OPC, blast furnace slag (BFS), and fly ash. The pellets were completely immersed in the solution with a 10 (cm^3 leachate) to 1 (cm^2 solid) ratio of the volume of leachant to the surface area of the pellet, with no change of the solution during the test (static cumulative test). No special handling of the pellet or the solutions was made to control the oxidation state. The analyses of the leachates were made, and the pellets were observed by scanning electron microscopy (SEM) to identify the nature of the products formed during curing. For comparison, the same conditions and testing were performed on pellets of high-fired DUO_2 for 9 months.

- The total release of uranium is minimal in the conditions of our tests: after 27 months at 150EC, maxima ranging from 0.40 to 0.0003% uranium (amount leached divided by the total amount of uranium present in the pellet before testing) were released from DUAGG™ and 0.90 to 0.0005% from high-fired DUO_2 after 9 months at 150EC.

- The release of uranium from DUAGG™ is lower than that from a DUO_2 pellet: under most conditions, a difference of at least one order of magnitude exists between DUAGG™ and DUO_2.

- For both DUAGG™ and DUO_2, the cement pore solutions have a beneficial effect for both DUAGG™ and DUO_2 on the uranium release. For DUO_2, the maximum release was as much as 260 times lower in BFS and 750 times lower for OPC than in DI water. For DUAGG™, the maximum release was as much as 600 times lower in BFS and 70 times lower in OPC than in DI water.

- The release rate of uranium has been compared with data found in the literature for release rates of uranium from UO_2 or simulated nuclear fuel and is lower for DUAGG™. It is comparable for DUO_2 in the presence of DI water, but the contact of pure uranium pellet with cement pore solutions decreased the release rate.

It was found that the combination of uranium and basalt in DUAGG™ results in a competition between the different species (uranium, aluminum, silicon, iron, titanium, and zirconium) for interaction with the solution species. The examination of the samples after more than 2 years of cure provides strong evidence that the basalt phase effectively protects the UO_2. A protective coating of recrystallization of basalt dissolution products covers the DU particles and forms a very dense layer that slows or stops the exchange of species between the pellet and the solution. The nature of the compounds formed after curing of the high-fired DUO_2 is similar to those reported in the literature for SNF or surrogate SIMFUEL. The SEM examination of DUO_2 showed the formation of schoepite or dehydrated schoepite in the samples kept in NaOH solution. In the cement pore solution, these recrystallization phases were not visible. For the DUAGG™ pellets, such products were not visible. Some DUAGG™ samples exhibited the presence of crystals of hydrotalcite, a mineral found in the alteration of basaltic and nuclear waste glass. A layer of products from the basalt dissolution was seen around the DUAGG™ pellet, providing a protective layer that slowed or stopped the exchange of species between the pellet and the surrounding solution. This finding indicates that DUAGG™ in DU concrete may have the same characteristic secondary mineral coating as a nuclear glass when the glass corrosion product precipitates. Such glasses are currently used for the long-term storage of HLW; they have been studied extensively during the years and are approved as being safe for storage of long-lived radionuclides. The results gathered in this project provide evidence that DUCRETE™ casks could have service lives sufficient to meet the projected needs of DOE and the commercial nuclear power industry.

DUO_2 CERMET FOR SNF WPs

Cermets are *cer*amic *met*al composites. For DU applications, the cermet consists of DUO_2 particulates embedded in steel with clean layers of steel on both sides of the cermet, as shown in Fig. 3. The clean layers of steel on the outside of the cermet avoid contamination issues and make the DU application “invisible” to operations.

High-temperature manufacturing techniques are used to make cermets; consequently, DUO_2 is the preferred chemical form because most other chemical forms of DU would decompose or react with the steel in the production process. Steel–UO_2 cermets have been manufactured in small quantities for use as nuclear fuels. Large quantities of non-uranium cermets are manufactured for other purposes. New methods may allow low-cost cermet production.

The cermet cask may be an enabling technology for a multipurpose cask system [16–18] where SNF is loaded at the reactor and remains in the cask through disposal in the repository. Such a system potentially (1) minimizes SNF handling; (2) improves resistance against thief, assault, and accidents; and (3) may offer superior long-term economics.

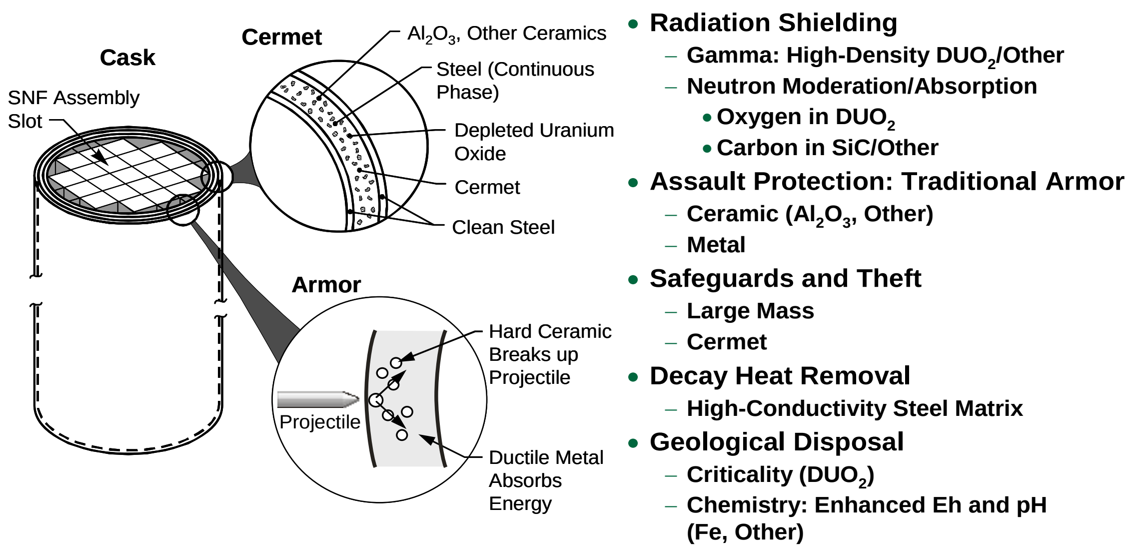

Fig. 3. SNF casks made of cermet (ceramics embedded in steel).

Uranium dioxide–steel cermet casks are being developed for the storage, transport, and disposal of SNF. Based on the properties of the cermets, there are three potential advantages: (1) higher-capacity casks with the same weight limits, (2) higher-capacity casks with the same dimensional limits, and (3) higher resistance to terrorist assault. Cermets are the traditional material used for armor. The research and development (R&D) activities have (1) identified and partly quantified the benefits of cermet casks and (2) partly developed new methods to fabricate the cermet casks. Because there are clear benefits for DUO_2 cermet casks, the primary issue is economics—how to minimize cask manufacturing costs. Two new methods were identified for manufacturing cermet casts [19, 20]. Each has two major advantages over historical methods of cermet manufacture: (1) lower costs and (2) avoidance of cermet welding—an exceedingly difficult technical challenge. One of the new fabrication methods has been patented (U.S. Patent 6,811,745). Preliminary laboratory experiments are under way to better understand the manufacturing requirements and thus allow reliable estimates of manufacturing costs. A joint program is under way with Russia to examine a third method of cask manufacture involving cermet casting.

U.S. AND RUSSIAN COLLABORATIVE R&D

The following paragraphs describe U.S. and Russian collaborative R&D to develop beneficial uses of DU in casks and its use as a chemical barrier to release of radionuclides in SNF geologic repositories.

Production and Testing of DUCRETE™, ISTC Project No. 2691

A joint U.S.–Russia research project is under way to produce DUAGG™ briquettes. DUAGG™ replaces stone in concrete, thereby enabling the synthesis of a heavy concrete termed DUCRETE™. Sensitivity studies have been made of the DUAGG™ reference recipe to obtain an optimum recipe and production process. Samples of DUAGG™ will be shipped to the United States for experiments. Russia is conducting tests of DUAGG™ and DUCRETE™. Analysis will be performed concerning the use of DUCRETE™ as a radiation shielding material in casks for SNF and HLW. The Rosatom Institutes VNIIEF, VNIINM, and VNIIKhT are participating in this project.

Funding for this project was received in February 2004 and will continue for 3 years. DUAGG™ samples have been made with the reference U.S. recipe. ISTC Project No. 2691 has also tested high-temperature DUO_2 ceramics of various other mineral mixtures. As a result of these tests, an advanced ceramic recipe and production technology have been developed. The new production technique eliminates the need to crush sintered briquettes, thereby avoiding DUO_2 dust formation. Because the new technique requires fewer components, it should result in lower manufacturing cost.

Production and Testing of DUO_2–Steel Cermets, ISTC Project No. 2693

A joint U.S.–Russia project is under way to produce and test a DUO_2–steel cermet material. The objective of this research is to produce and test the properties of cast metal–ceramic alloy (cermet) for use as a shielding material in the construction of casks designed for transportation, storage, and disposal of SNF and radioactive wastes [21]. The project will result in the development of a low-cost casting technology and will produce samples for testing. Comprehensive testing of cermet samples will be conducted in both Russia and the United States.

Russia is represented in this project by the All-Russian Research Institute of Experimental Physics, Sarov; the All-Russian Research Institute of Chemical Technology, Moscow; and the All-Russian Research Institute of Inorganic Materials, Moscow. The project is funded by the U.S. Department of State through the International Scientific Technical Center; funding for this project began in February 2004, and will continue for 3 years. The experiments on cast cermet production are being carried out using an existing laboratory cold-wall melter furnace, the IPKhT-100M. Two modes of cast cermet production have been tested.

- The first mode is based on direct mixing of high-dispersive DUO_2 powder in molten steel. The average DUO_2 particle dimension was about 0.5 micron; the average apparent density was equal to 1.65 g/cm^3.

- In the second mode, larger particulates of granulated DUO_2 (∃-phase of U_4O_9) are introduced into steel. The experiments were carried out using granules of UO_2 with various dimensions (1.0–2.0 mm) and specific density of not less than 10 g/cm^3.

Experiments using the first mode demonstrated that cast cermet produced with 50 volume percent of DUO_2 does not provide the requisite density and strength properties. However, experiments using the second mode resulted in cast cermet with a density of not less than 9.4 g/cm^3 and with satisfactory strength properties. It is possible to produce cermet with density near 10 g/cm^3 by optimizing the granulated particle size.

Sorption Capture of Long-Lived Radionuclides by DU Oxides and Hydroxides, ISTC Project No. 2694

The objective of this U.S. Department of State funded research is to obtain experimental data on the sorption of long-lived radionuclides (^{237}Np and ^{99}Tc) from underground waters onto DUO_2 and uranium hydroxides at various temperatures, pHs, and Ehs. A model is being developed of long-lived radionuclide transport through layers of DUO_2 surrounding SNF buried in geological repositories. This work is being conducted at the Institute of Physical Chemistry, Russian Academy of Sciences, Moscow, Russia. Funding for this project began in December 2003, and will continue for 2 years.

The sorption of long-lived radionuclides from J-13 groundwater (Yucca Mountain) onto DUO_2 has been measured for the first time. These data show that urania in a repository will significantly reduce the maximum site boundary dose by sorbing radionuclides (e.g., neptunium) that are the primary contributors to the site boundary dose. This discovery indicates the possible use of DU oxides as a geochemical shielding barrier in geological repositories of radioactive wastes. These experimental results are reported in another paper in this session, "Sorption of Long-Lived Radionuclides in Geological Repository Underground Waters by Depleted Uranium Oxides," by T. V. Kazakovskaya et al.

Production of Granulated DUO_2 Particles by Melting in an Induction Cold Crucible Melter, Civilian Research and Development Foundation (CRDF) Project No. RUC2-20203-MO-05

Both DUCRETE™ and DU–steel cermet materials use DUO_2 particulates. An inexpensive method is needed to manufacture these particulates. Some of the benefits of melting urania and then solidifying poured drops of UO_2 are as follows: (1) The converted DUF_6 from the conversion plants under construction will be primarily in the form of U_3O_8. However, its use requires that it be UO_2. Use of the cold crucible melter method forms UO_2 and eliminates a step to convert it from U_3O_8 to UO_2, thus saving substantial expense. (2) The converted DUF_6 contains impurities (e.g. fluorine) that are detrimental to DUCRETE™ and DU–steel cermet performance. Use of a cold crucible melter process removes these impurities. (3) Use of a cold crucible melter that produces DUO_2 particulates eliminates several process unit operations to produce DUAGG™, thus lowering production costs. (4) DUO_2 particulates and DUAGG™ produced at very high temperatures (~2900EC) are chemically less reactive than those produced by current methods. Less-reactive depleted urania will improve the performance of DU–steel cermets and DUCRETE™. This task will design, fabricate, and operate an induction cold crucible melter for the study of DUO_2 melting and granulation. Samples of melted DUO_2 and its granules will be produced for testing at the Russian Research Institute of Chemical Technology.

On September 3, 2004, the Acting Assistant Secretary for Environmental Management, Paul Golan, signed a letter announcing that DOE-EM would fund the project. Funding of this work is through the CRDF. This work began in July 2005 and will continue for 4 years.

The objective of the Beneficial Uses of DU R&D Program is to develop the technical basis for new commercial products. One such commercial product is a next generation storage/shipping cask for SNF. In order to market next-generation casks using new DU composite materials, the cask must be licensed. In order to obtain a cask license, prototype casks must be built and tested. The following are two projects that address these requirements.

The Fabrication of a ¼-Scale SNF Storage Cask Utilizing DUCRETE™ Material, Initiative for Proliferation Prevention (IPP) Project No. T2-206

The objective of this task is to fabricate and test a ¼-scale storage cask that utilizes the new DUCRETE™ shielding material. This task is funded by DOE's IPP Program and will be performed at VNIIEF. This work is scheduled to begin in October 2005 and will continue for 4 years.

The Fabrication of a ¼-Scale SNF Transport Cask Utilizing DU–Steel Cermet Material, ISTC Project No. 3258

The objective of this task is to fabricate and test a ¼-scale transport/storage cask that uses the new DU–steel cermet shielding material. This task will be performed at VNIIEF. A proposal will be submitted to DOE to conduct this work by May 2006.

SUMMARY

Laboratories, institutes, and private companies in the United States and Russia are working together to develop new composite materials for use in SNF and HLW storage and transport casks. The laboratories and institutes are responsible for developing materials to the point where new products are feasible. The basic scientific research for the utilization of existing inventories of DU in these products is almost completed. The ¼-scale prototype casks are being designed and fabricated and will be tested to obtain data for licensing. Private companies will design, license, and market casks made of new DU composite materials. The first deployment of these new casks is expected in about the year 2012.

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