

# **REPOSITORY CRITICALITY CONTROL WITH DEPLETED- URANIUM-DIOXIDE CERMET WASTE PACKAGES**

Charles W. Forsberg

Oak Ridge National Laboratory\*  
P.O. Box 2008  
Oak Ridge, Tennessee 37831-6179  
Tel: (865) 574-6783  
Fax: (865) 574-9512  
Email: forsbergcw@ornl.gov

File Name: Cermet: Cermet.criticality.Reno  
Page limit: 10 pages  
Abstract Number: 35835  
Session: Geological Disposal

Prepared for  
Session: Geological Disposal  
Embedded Topical Meeting on Practical Implementation of Nuclear Criticality Safety  
American Nuclear Society  
Reno, Nevada  
November 11–15, 2001

Manuscript Date: July 7, 2001

The submitted manuscript has been authored by a contractor of the U.S. Government under contract DE-AC05-00OR22725. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes.

---

\*Oak Ridge National Laboratory, managed by UT-Battelle, LLC, for the U.S. Dept. of Energy under contract DE-AC05-00OR22725.

# REPOSITORY CRITICALITY CONTROL WITH DEPLETED-URANIUM-DIOXIDE CERMET WASTE PACKAGES

Charles W. Forsberg  
Oak Ridge National Laboratory  
P.O. Box 2008; Oak Ridge, Tennessee 37831-6179  
Tel: (865) 574-6783; Fax: (865) 574-9512; Email: [forsbergcw@ornl.gov](mailto:forsbergcw@ornl.gov)

## SUMMARY

It is proposed that the structural components and internal basket structures of waste packages (WPs) be constructed of depleted uranium dioxide (DUO<sub>2</sub>)–steel cermets. The cermet contains DUO<sub>2</sub> imbedded in a steel matrix. The WPs are filled with spent nuclear fuel (SNF) and placed in a geological repository. The WP provides a handling container for placement of SNF in the repository and is an engineered barrier to delay SNF degradation and subsequent release of radionuclides.

SNF and other fissile wastes contain enriched uranium and transuranic fissile isotopes; thus, the potential for nuclear criticality exists. Most of the transuranic fissile isotopes, such as <sup>239</sup>Pu, will have decayed to <sup>233</sup>U or <sup>235</sup>U before significant fissile-isotope migration from the degraded SNF or other fissile waste forms has occurred. Consequently, post-closure repository criticality issues are primarily from the fissile isotopes of uranium. As the WP degrades, the <sup>238</sup>U in the DUO<sub>2</sub>-steel cermet would mix with the degrading SNF and isotopically dilute <sup>233</sup>U and <sup>235</sup>U to levels that would ensure that post-closure criticality would not occur.

## I. INTRODUCTION

The proposed Yucca Mountain (YM) repository will potentially contain many different types of waste with fissile isotopes: commercial light-water reactor (LWR) SNF, U.S. Department of Energy SNF, U.S. Navy SNF, aluminum waste forms from processing enriched-uranium aluminum-clad SNF, and potentially immobilized plutonium. The uranium enrichment levels will vary from <1 wt % <sup>235</sup>U to levels that may exceed 90 wt % <sup>235</sup>U in <sup>238</sup>U. Criticality control in the repository must be maintained under two very different conditions:

- *Operational criticality control.* Criticality control must be maintained during the operational phase of the repository. Traditional approaches to criticality control may be used that depend upon (1) limiting the fissile mass per WP or some subdivision of the WP, (2) controlling the geometry, (3) controlling the chemistry, and (4) adding neutron absorbers.
- *Post-closure criticality control.* Criticality control must be maintained after repository closure to (1) ensure repository performance, (2) meet licensing requirements, and (3) obtain public acceptance. Nuclear criticality generates (1) added radioactivity and (2) heat that can

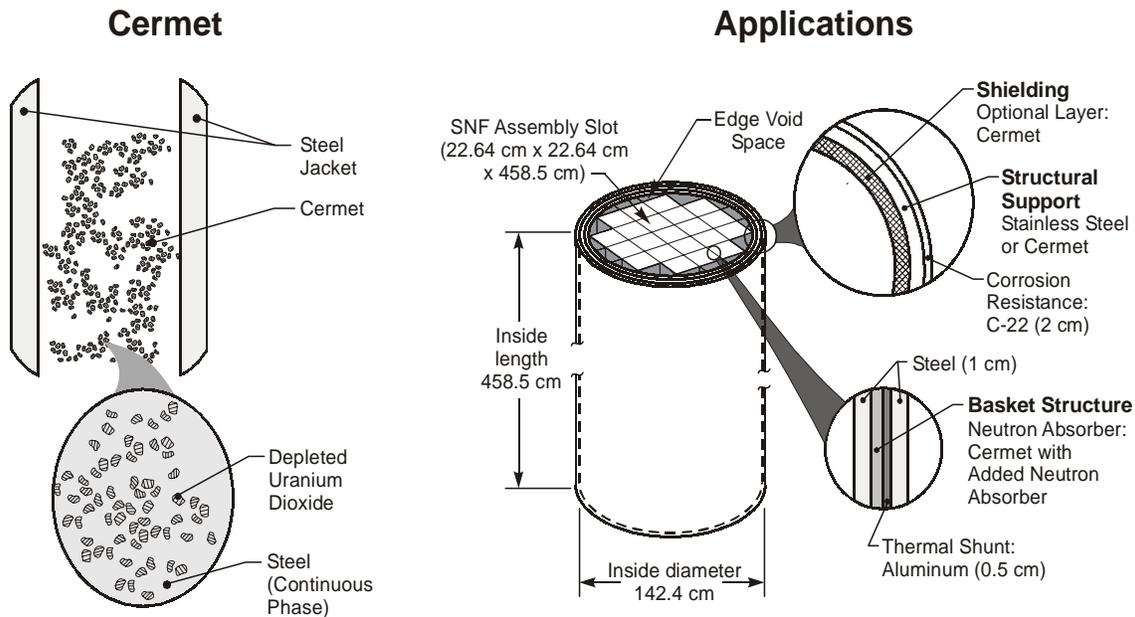
alter repository performance. Post-closure criticality control is more difficult to maintain because as the WPs degrade, the geometry and chemical composition change over time. The fissile materials may migrate with the groundwater. The close spacing of WPs may allow fissile materials from multiple WPs to migrate together in groundwater. Many geochemical mechanisms can concentrate and separate fissile materials from other elements (including added neutron absorbers) in the degraded WP and the geological environment. These phenomena have in fact created natural nuclear reactors (Oklo, etc.) earlier in the earth's history. Without appropriate controls, criticality may occur kilometers from the repository site in a geology that is very different from that of the repository. Such a criticality creates the potential of added radionuclide releases to the environment from newly generated fission products and actinides.

The primary risk of nuclear criticality in a geological repository [1] is from fissile isotopes of uranium ( $^{233}\text{U}$  and  $^{235}\text{U}$ ) because of (1) radioactive decay of other fissile actinides to these isotopes, (2) the design of repositories, and (3) the geochemistry of fissile materials. With enriched uranium and  $^{233}\text{U}$ , criticality control can be ensured by isotopic dilution with DU containing  $^{238}\text{U}$ . Because all uranium isotopes have the same chemistry, isotopic mixing of fissile uranium with DU ensures criticality control even as the geometry and chemical environment of the uranium change over time. The use of  $\text{DUO}_2$ -steel cermet to maintain operational and post-closure repository criticality control is discussed herein.

## II. CERMET CHARACTERISTICS

A  $\text{DUO}_2$ -steel cermet [2] consists of  $\text{DUO}_2$  particulates embedded in a continuous-steel phase (Fig. 1). Typical cermets use sandwich construction, with a clean uncontaminated steel layer on each side of the cermet. The  $\text{DUO}_2$  cermet can replace steel components within the WP (structural shell, shielding, and the basket). For the various applications, different grades of steel may be used and the ratio of  $\text{DUO}_2$  to metal will vary. If a cermet were used in the body of the WP, an outer layer of corrosion-resistant metal would be chosen to maximize corrosion resistance in the particular geological environment. Cermets [2,3] may meet near-term WP requirements (structural support, radiation shielding, criticality control) while (1) improving the repository post-closure WP performance, (2) making beneficial use of excess DU, and (3) addressing the post-closure repository criticality control issue. Cermets may be used as either a first or second generation WP.

A  $\text{DUO}_2$ -steel cermet provides a means for adding DU to a WP in a form such that (1) there is no  $\text{DUO}_2$  contamination during WP handling operations, (2) the DU is in a ductile form suitable for a WP, and (3) the DU is in a chemical form that is acceptable to the repository and the U.S. Nuclear Regulatory Commission (NRC). The chemical form of DU is the same as that of uranium in LWR SNF; thus, issues associated with chemical compatibility between different repository components are minimized. A recent NRC advisory letter [4] indicated that oxides would be an acceptable DU disposal form in a repository; however, the letter also noted that significant questions remained about the inclusion of DU metal in a disposal site because of its long-term chemical behavior.



**Fig. 1. Characteristics and uses of cermets in WPs for pressurized-water reactor (PWR) SNF**

The methods used to manufacture cermets [2] allow other neutron absorbers (gadolinium oxide, boron carbide, etc.) to be added. The use of high cross-section absorbers in the basket (with added DU in the shell), may reduce the thickness of basket walls and thus increase basket capacity for a given volume. The addition of such absorbers to the basket structure allows the same cermets to be used to ensure operational and post-closure criticality control. Cermets can be made with  $\text{DUO}_2$  contents approaching 90 vol %; however, the  $\text{DUO}_2$  content of most cermets will typically be closer to 50 vol %. Several factors are expected to lower the average  $\text{DUO}_2$  content: (1) the need for clean external layers over the cermet and (2) sufficient steel to meet strength, ductility, and thermal conductivity requirements.

Cermets [2,3] have been used as nuclear fuels and are currently being investigated in Europe for use as very-high-burnup PWR fuels; thus, the physical properties are understood. Non- $\text{DUO}_2$  cermets containing other neutron absorbers have been manufactured for use in construction of SNF baskets. Non- $\text{UO}_2$  cermets are produced in large quantities (>100,000 t/year) for a variety of non-nuclear applications.

Sufficient DU is available for this application. The United States has >500,000 tons of excess DU from the production of enriched uranium. Four to seven tons of DU are produced for every ton of LWR fuel. Using this DU as a component of cermet in the WP would avoid costs associated with the disposal costs of this material. If self-shielded WPs are used, half or more of this DU inventory would possibly be used. The avoided costs of DU disposal may be a significant factor in the overall economics of DUO<sub>2</sub>-cermet WPs.

### III. POST-CLOSURE REPOSITORY CRITICALITY CONTROL

In the repository post-closure environment, the potential for nuclear criticality is primarily from the fissile uranium isotopes <sup>233</sup>U and <sup>235</sup>U. Most other fissile isotopes (including <sup>239</sup>Pu) are expected to decay to fissile uranium isotopes before significant migration from the waste form and WP [1]. For fissile uranium in a disposal facility (unless DU is added), there are three possible fates when uranium dissolves and moves with groundwater.

- *Fission.* The earth's geochemistry [1] can concentrate uranium into ore bodies by selective precipitation from groundwater and other mechanisms. The normal geochemical processes separate uranium from all neutron absorbers—except <sup>238</sup>U. The geological record shows that natural nuclear reactors (such as at Oklo, Gabon, Africa) have occurred when the enrichments of the uranium were as low as 1.3 wt % <sup>235</sup>U in <sup>238</sup>U. However, nuclear criticality can no longer occur in natural uranium ore bodies because the decay of <sup>235</sup>U since these early reactors has lowered the assay of natural uranium to 0.71 wt % <sup>235</sup>U.
- *Isotopic dilution with natural uranium.* In time, dissolved uranium in groundwater from the repository with fissionable concentrations different from those of natural uranium will isotopically exchange with natural uranium in the rock and be isotopically diluted to ~ 0.7 wt % <sup>235</sup>U and trace quantities of <sup>233</sup>U if the fissile uranium contains <sup>233</sup>U. This interchange of natural uranium in rock with uranium in SNF from a repository ultimately eliminates the potential for nuclear criticality.
- *Radioactive decay.* Fissile uranium isotopes decay to nonfissile isotopes and this ultimately eliminates the potential for nuclear criticality. The half-life of <sup>235</sup>U is 700-million years. However, the geochemical processes that result in mountain building and destruction (by erosion) are an order of magnitude faster than this; thus, enriched uranium is preferentially destroyed by fission or isotopic dilution before significant radioactive decay occurs.

There are three post-closure criticality control strategies.

- *Predict future evolution of the disposal site.* The behavior of fissile materials in the WP can be modeled with time to determine if post-closure criticality may occur and the nature of its possible consequences. The model must include fissile waste form degradation, WP degradation, migration of all fissile materials, and migration of all significant neutron absorbers in the rock until (1) isotopic dilution with natural uranium in the rock occurs, (2) the risks of nuclear criticality are shown to be acceptable, or (3) the time is beyond that of regulatory interest. If the estimated risk from nuclear criticality is judged to be unacceptable, the repository design must be changed so that the risks are acceptable.

- *Add DU to the WP.* The DU can be incorporated into the WP. As the fissile waste forms and WP degrade and the uranium begins to migrate, DU intermixes with the fissile uranium and isotopically dilutes it to a level below that required for nuclear criticality to occur (option discussed herein).
- *Add DU to the waste.* DU can be isotopically mixed with the fissile uranium in the waste until nuclear criticality is not credible. This option requires that the waste be processed.

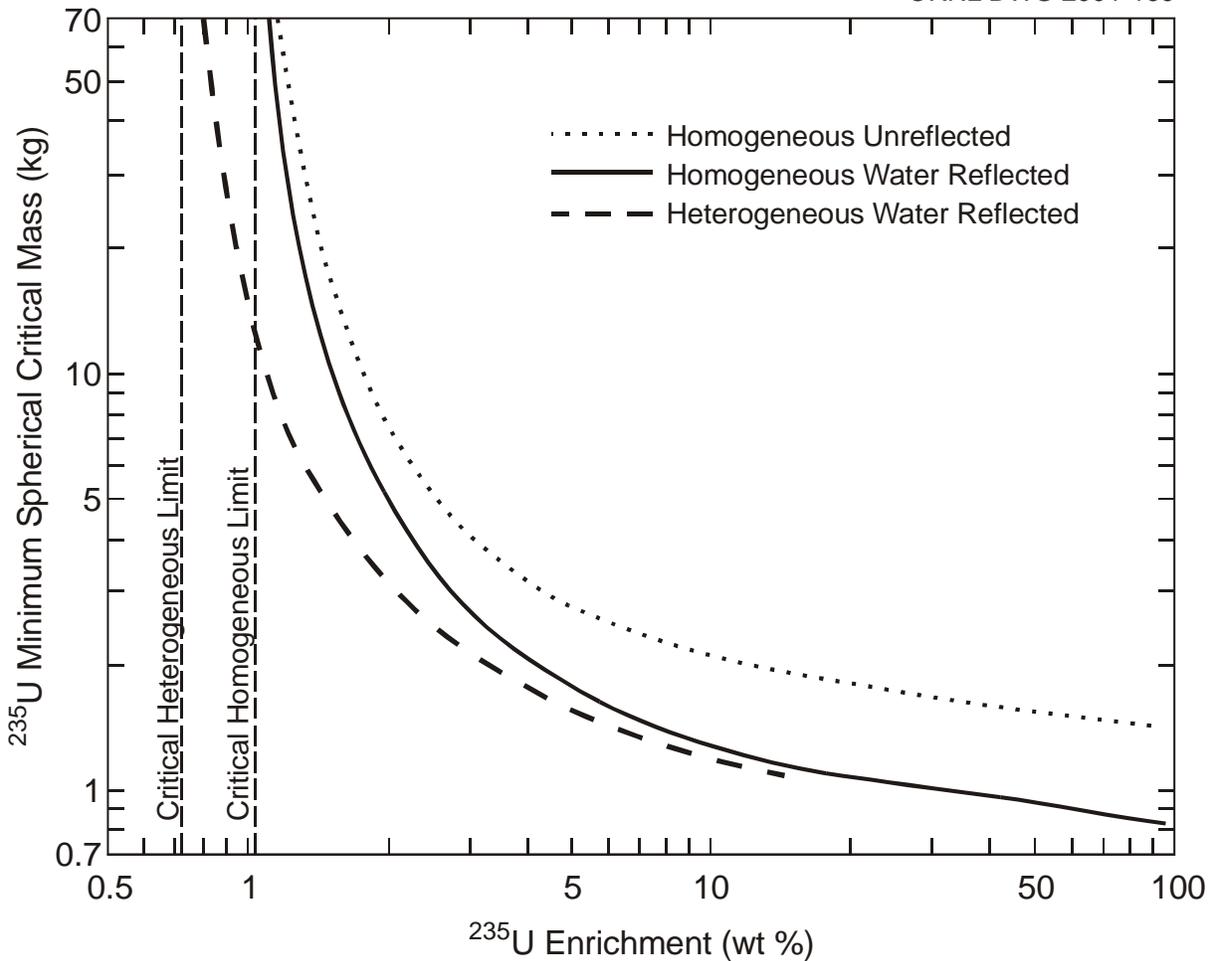
#### IV. DU OPTIONS AND CONSTRAINTS

If isotopic dilution with DU is used for repository criticality control, a key issue is how much DU must be added. There are several factors to consider.

- *Fissile content.* The DU is used for isotopic dilution of fissile uranium isotopes. However, most of the transuranic isotopes decay to uranium isotopes. For example,  $^{239}\text{Pu}$  decays to  $^{235}\text{U}$  with a half life of 24,000 years. Given the long time frames of repository operation, the decay of transuranic isotopes must be considered. For example, typical LWR SNF has a fissile uranium content  $<1$  wt %  $^{235}\text{U}$  in  $^{238}\text{U}$ . However, after accounting for the decay of higher isotopes, the ultimate uranium fissile content is - 1.5 wt % equivalent  $^{235}\text{U}$  in  $^{238}\text{U}$ .
- *Isotopic dilution limits.* Under most conditions, criticality control is ensured when  $^{235}\text{U}$  is diluted to  $<1$  wt %  $^{235}\text{U}$  in  $^{238}\text{U}$  [1] and  $^{233}\text{U}$  is diluted to  $<0.66$  wt %  $^{233}\text{U}$  in  $^{238}\text{U}$  [5]. Analysis [1] of natural reactors and theoretical geochemical studies indicates that nuclear criticality is unlikely when  $^{235}\text{U}$  is diluted to  $<1.3$  wt %  $^{235}\text{U}$  in  $^{238}\text{U}$ . The natural environment does not contain highly purified materials with very low absorption cross sections that are required for criticality to be achieved at very low uranium enrichments.

The risk of nuclear criticality drops rapidly with isotopic dilution. Figure 2 shows the minimum quantity of  $^{235}\text{U}$  needed in several idealized water-moderated systems for nuclear criticality to occur as a function of enrichment [6]. While the figure shows the  $^{235}\text{U}$  quantities for criticality to occur, at each enrichment there is a different quantity of associated  $^{238}\text{U}$ . For example, at 1 wt %  $^{235}\text{U}$ , the total mass of uranium is 100 times the quantity of  $^{235}\text{U}$ . Ten kilograms of  $^{235}\text{U}$  is mixed with a 1000 kg of uranium—primarily  $^{238}\text{U}$ . The chances that large uranium deposits with low impurities and the proper geometries will naturally form are much less than for small deposits. With higher enrichments, there is a greater incentive to add DU to the WP because it is harder to assure post-closure criticality will not occur.

- *Methods of isotopic dilution.* As the cermet and waste form degrade, the different uranium isotopes will intermix. Sufficient isotopic dilution for criticality control can be assured if sufficient DU is added to overwhelm mixing difficulties. The quantity of excess DU beyond that theoretically needed depends upon two factors: intermixing and chemistry. If the  $\text{DUO}_2$ -cermet is used for the WP basket, physical intermixing between the waste form and the DU will be greater than if the cermet is used only for the WP structural shell. The closer the chemical form of the fissile waste form and the  $\text{DUO}_2$ -cermet, the greater the likelihood of efficient isotopic mixing.



**Fig. 2. Critical mass of  $^{235}\text{U}$  as a function of enrichment.**

The  $\text{DUO}_2$  cermet by itself may be used for criticality control or may contain other neutron absorbers that are added to ensure (1) operational criticality control and (2) early post-closure criticality control between the time the fissile waste form begins to degrade but before full isotopic dilution occurs. The other neutron absorbers can be (1) added to the cermet metal as an alloying ingredient, (2) intermixed as an oxide with  $\text{DUO}_2$  particulates in the cermet, or (3) incorporated into the  $\text{UO}_2$  crystal structure. The last form is essentially identical to the use of rare earth neutron absorbers (gadolinium oxide, etc.) in  $\text{UO}_2$  fuels as burnable absorbers.

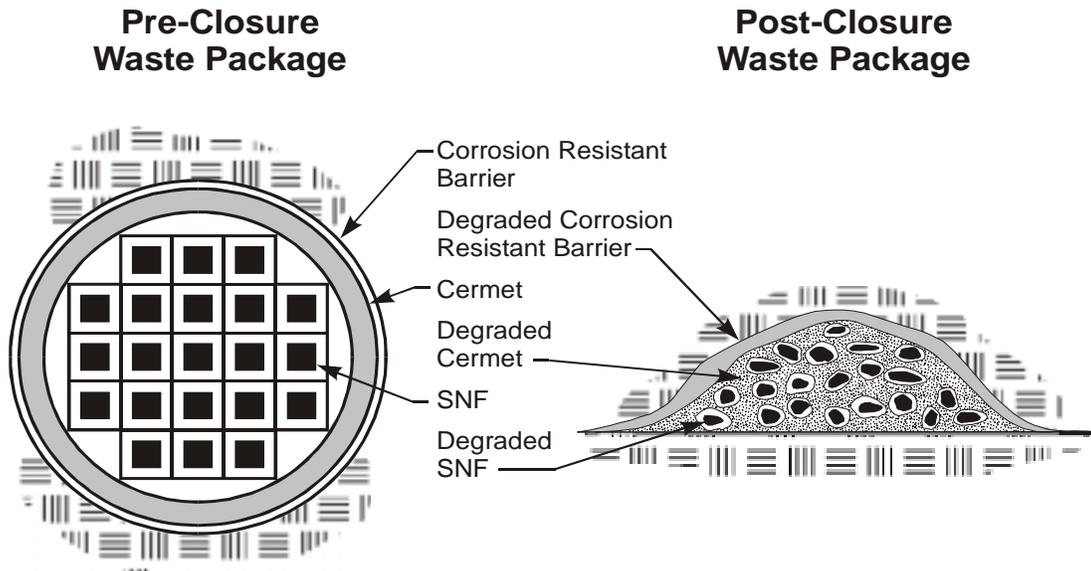
## V. WASTE FORM AND WP EVOLUTION

### *Evolution of WP*

A cermet WP starts with a well-defined geometry that changes over time. After WP failure, the cermet will begin to corrode. As the metal corrodes,  $\text{DUO}_2$  is exposed, released from the cermet, and reacts with the groundwater. In an oxidizing environment, it would be oxidized to hydrates of higher oxidation state uranium oxides [7], such as  $\text{U}_3\text{O}_8$  and  $\text{UO}_3 \cdot x\text{H}_2\text{O}$ . As this occurs, the waste form begins to degrade. The degrading cermet creates a particulate bed of metal oxides and hydrated DU oxides that surrounds and intermixes with the waste form. Figure 3 shows new WP and the fully degraded form where the WP components and SNF have been oxidized and void spaces collapsed.

If the basket material is more corrosion resistant than the SNF, there will be layers of hydrated DU oxides between the degrading fissile waste forms. SNF assembly debris will have consolidated before basket breakup. If the basket material is less corrosion resistant than the SNF (YM WP design with carbon steel basket), the basket will degrade first and the resultant particulates from the degradation of the cermet will tend to fill the coolant channels of the SNF. The isotopic dilution occurs when groundwater flows through the degraded WP and fissile waste forms. Several stages of operation can be identified.

- *Three-layer system.* Groundwater from above contacts the hydrated DU oxides that were associated with cermets in the top of the WP. The groundwater becomes partly or fully saturated in DU. As the groundwater flows downward into fissile waste form, DU exchanges with the SNF or other fissile-waste-form enriched uranium. If the WP was constructed using a cermet, there will be altering layers of DU and enriched uranium. The groundwater fissile-uranium isotopic assay increases. The groundwater continues to flow downward into the DU hydrated oxides associated with cermets on the bottom of the WP. Isotopic exchange continues with the uranium isotopic assay decreasing toward the bottom of the degraded WP.
- *Two-layer system.* After the DU above the fissile waste form has fully dissolved, the fresh groundwater will be in contact with the partly isotopically-diluted fissile waste form. This uranium will dissolve, flow downward, and be isotopically diluted by the DU below the waste form.
- *Single layer system.* When the fissile waste form is fully dissolved, the now partly-enriched DU that was originally from the cermet on the bottom of the WP will begin to dissolve and be transported.
- *Beyond the WP.* Uranium with groundwater will continuously exchange with absorbed uranium on the rock. Mixing is a continuous process beyond the WP.



**Fig. 3. Evolution of cermet WP.**

The cermet originally contained  $\text{DUO}_2$  particulates that were  $<1$  mm in size. As the cermet degrades, these particulates break free of the oxidizing steel to create a high  $\text{UO}_2$  surface area that enhances isotopic exchange with uranium in the groundwater. In an oxidizing groundwater environment, such as YM,  $\text{UO}_2$  from the cermet or the waste form is oxidized. The oxidation process [7] tends to form smaller particles than the initial  $\text{UO}_2$  particles which further increases the surface area for isotopic exchange. For criticality control,  $\text{DUO}_2$  in bulk form in the WP is less desirable.

Isotopic exchange begins as soon as the waste form and cermet begin to degrade in the presence of water. In many WP failure scenarios, the WP may partly fill with water. This environment accelerates isotopic exchange even though there may be no transport of uranium from the WP.

The uncertainties associated with nuclear criticality are drastically reduced as the quantities of DU in the WP increase. The potential effects of various barriers to isotopic dilution, such as cladding, become much less significant. The transport of uranium from the degraded WP is limited by the solubility of uranium in groundwater [1]. In the proposed YM repository for a self-shielded 21-PWR WP [1] using a DU cermet (42.26 t of uranium, groundwater solubility of  $7.6 \text{ g U/m}^3$ , and  $3.25 \text{ cm/year}$  of groundwater flow), it is estimated that it will take more than 20 million years for all the uranium to be transported from the location of the original WP. In contrast, SNF cladding (estimated to last a few thousand to several tens of thousands of years) and other potential barriers to isotopic dilution disappear quickly.

If other neutron absorbers are needed in the WP basket for operational criticality control, the absorbers should remain with the degraded basket and SNF for criticality control until isotopic dilution has occurred. It is usually more beneficial to incorporate insoluble absorbers (such as gadolinium oxides) within the DUO<sub>2</sub> crystal structure—rather than as a separate oxide in the cermet or alloyed as a component of the metal. All uranium (DU and SNF) in the WP evolves toward more thermodynamically-stable hydrated uranium oxides and silicates over time. These similar chemistries and densities reduce the potential for separation of the neutron absorber from the remains of the cermet until after isotopic dilution has occurred. In contrast, if the neutron absorber is associated with other metal oxides, its properties will be different from those of uranium oxides, which increases the potential that physical or chemical mechanisms will separate these neutron absorbers from the uranium.

Cermets may allow significant inventories of SNF with moderately to high enriched uranium in a single WP. The DU may avoid fissile mass limits per WP. The addition of DUO<sub>2</sub> to the WP is also expected to reduce the radionuclide release rate [1] from the WP by a variety of mechanisms; hence, there are multiple incentives for adding DUO<sub>2</sub> to WPs.

### ***Methodology***

The basic principles of cermet applications for criticality control have been described herein. Implementation of this approach requires a methodology that defines how much DUO<sub>2</sub> is required for a specific WP and compares design options. The methodology [8,9] developed to assess the potential for nuclear criticality in the proposed YM repository would also be used for DUO<sub>2</sub>-cermet WPs. The methodology, in its simplest form, consists of two steps.

- *Geochemistry.* Mechanical and geochemical models are used to predict the evolution of the WP over time. The output is a detailed description of the location, geometry, and chemical composition of the fissile materials and their surroundings.
- *Neutronic analysis.* The results of the geochemical and mechanical modeling are put into a nuclear criticality neutronics code to determine if nuclear criticality can occur. If nuclear criticality is predicted or if  $k_{\text{eff}}$  is too high, the design is modified.

The criticality analysis of a WP with DU is much simpler with fewer uncertainties than the equivalent criticality analysis of a WP without DU. Sufficient DU is added that a semi-deterministic approach is used to assure criticality control. The primary uncertainty is the rate of isotopic mixing. The criticality analysis would be used to provide confidence that sufficient isotopic mixing occurs; that is, the behavior is as expected. The cermet criticality analysis can stop as soon as significant isotopic dilution occurs and, in most cases, can stop by the time the uranium leaves the WP. There is no near-field (near the WP) and far-field criticality analysis. Without DU, assuring criticality control becomes complex and difficult because the repository may evolve in many different ways with time.

## **VI. CONCLUSIONS**

The use of DUO<sub>2</sub>-steel cermets provides a method in which the potential for nuclear criticality in the repository is minimized by isotopic dilution with <sup>238</sup>U. If the DU is in the form of a cermet

that is incorporated into the WP, there is little impact on operations. The benefits of isotopic dilution increase significantly with the fissile-enrichment level of the fissile waste form.

## ACKNOWLEDGMENT

This work on cermet is done under the auspices of the U.S. Department of Energy Depleted Uranium Uses Research and Development Program.

## REFERENCES

1. C. W. Forsberg, "Effect of Depleted-Uranium-Dioxide Particulate Fill on Spent-Nuclear-Fuel Waste Packages," *Nucl. Technol.*, **131**, p. 337–353 (September 2000).
2. C. W. Forsberg, "Cermet Waste Packages Using Depleted Uranium Dioxide and Steel," *2001 International High-Level Radioactive Waste Management Conference, Las Vegas, Nevada, April 29–May 3, 2001*, Am. Nucl. Soc., La Grange Park, Illinois.
3. C. W. Forsberg and L. B. Shappert, "Cermet Transport, Storage, and Waste Packages Using Depleted Uranium Dioxide (DUO<sub>2</sub>) and Steel (in preparation)," *13<sup>th</sup> International Sym. on the Packaging and Transportation of Radioactive Materials, U.S. Department of Energy, Chicago, Illinois., September 3–7, 2001*, Am. Nucl. Soc., La Grange Park, Illinois.
4. E. J. Leeds, "Comments on DUF<sub>6</sub> Materials Use Roadmap, Draft, Dated September 1, 2000," Letter to the U.S. Department of Energy from the U.S. Nuclear Regulatory Commission, Special Projects Branch, Division of Fuel Cycle Safety and Safeguards, Office of Nuclear Material Safety and Safeguards, ML003762080 (October 18, 2000).
5. K. R. Elam, C. W. Forsberg, C. M. Hopper, and R. Q. Wright, "Isotopic Dilution Requirements for <sup>233</sup>U Criticality Safety in Processing and Disposal Facilities," ORNL/TM-13524, Oak Ridge National Laboratory (November 1997).
6. H. C. Paxton and N. L. Pruvost, *Critical Dimensions of Systems Containing <sup>235</sup>U, <sup>239</sup>Pu, and <sup>233</sup>U: 1986 Revision*, LA-10860-MS, (July 1987).
7. C. W. Forsberg, "Depleted Uranium Dioxide as a Spent-Nuclear-Fuel Waste-Package Particulate Fill: Fill Behavior," *Waste Management 2001*, Tucson, Arizona, February 25–March 1, 2001.
8. H. R. Radulescu and J. W. Davis, "Disposal of Shippingport LWBR Spent Nuclear Fuel in Yucca Mountain: Waste Package Internal Criticality Analysis," *Spectrum 2000: International Conference on Nuclear and Hazardous Waste Management, Chattanooga, Tennessee, September 24–28, 2000*, Am. Nucl. Soc., La Grange, Illinois.
9. U.S. Department of Energy, *Disposal Criticality Analysis Methodology Topical Report*, YMP/TR-004Q, Las Vegas, Nevada (November 1998).