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RADIATION SHIELDING USING DEPLETED URANIUM OXIDE IN NONMETALLIC MATRICES

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ABSTRACT

The depleted uranium (DU) inventory in the United States exceeds 500,000 metric tons (tonnes). This report reviews the status of U.S. Department of Energy (DOE) research concerning the use of its inventory of DU as neutron and gamma shielding as uranium oxide (UO_x) in nonmetallic matrices, (e.g., concrete). This program envisions that a large portion of the U.S. inventory of DU will be used in the fabrication of nuclear shielding for the storage, transport, and disposal of spent nuclear fuels.* Just in the storage of commercial U.S. spent nuclear fuel (SNF), the cumulative amount of DU oxide (DUO_2) that could be used in nonmetallic matrix dry-storage casks though 2020 is over 408,455 tonnes (360,058 tonnes of DU).

The purpose of this U.S. research effort is to develop a DU shielding technology to the point that a demonstrated technical basis exists for deployment. In particular, a need exists to (1) establish the ability to manufacture DU coarse aggregates for heavy concretes; (2) optimize the design and costs; and (3) ensure confidence in the reliability and safety of the chemical and physical stability of the DU aggregate. In addition, the neutron-shielding characteristics of these aggregates and their binders can be enhanced.

Aggregates made of UO_2 are combined with cementitious binders that enhance neutron shielding and result in high strengths. Uranium is a very effective gamma shield because of its high density and high atomic number (Z). Binders considered in this study include Portland cement, blast furnace slag, and pozzolanic cements. These DU-based shielding materials greatly reduce the size and weight of storage, transport, and disposal casks. The economic advantage gained through using smaller and lighter casks offsets the increased fabrication costs.

This report describes the production of this shielding material and documents measurements of DUO_2 -aggregates' physical properties and chemical durability. Current testing at the Oak Ridge National Laboratory (ORNL) measures the extent and rates of surface reactions of the aggregates under the expected service temperatures and the simulated chemical environments of cement pastes. Intact DUO_2 -aggregates were tested for chemical reactivity with the cement paste the using a modified American Society for Testing and Materials ASTM C289-94 method to measure reactions with concrete pore liquids.

* This work was performed under by the U.S. Department of Energy, Office of Environmental Management (EM-21) under their Depleted Uranium R&D Program.

BACKGROUND

DUCRETE Origins and Development

One of the most obvious uses for the large U.S. inventory of depleted uranium (DU)—which exceeds 500,000 metric tons (tonnes)—is as nuclear shielding. A research program being conducted by the U.S. Department of Energy (DOE) envisions that a large portion of this DU will be used in the fabrication of nuclear shielding for the storage, transport, and disposal of spent nuclear fuel (SNF). DU metal has been used in casks as shielding because its high density provides the needed gamma attenuation for the lowest-weight and smallest casks. Studies have assessed the use of uranium metal for shielding in both spent fuel¹ and high-level waste (HLW)² casks. A review of DU metal production and fabrication costs showed that depleted metal was more expensive than other common shielding materials such as steel, lead, and concrete.³ Therefore, the primary application for uranium metal shielding is for transportation casks, where the most stringent total-package size and weight limits exist and where high-cost, DU metal shielding can be justified. In addition, there is an added benefit to the nuclear community because this use as shielding will consume a large quantity of DU from the existing national inventory.

These findings led to the consideration of alternative uses for DU such as a DU ceramic, which is still very dense but has considerably lower production and fabrication costs than DU metal. The first alternative developed was a concrete called DUCRETE™. This material has characteristics of both an efficient gamma absorber (uranium) and a low-atomic number (low-Z) neutron-slowing material such as hydrogen or carbon in the cement matrices. Figure 1 shows the effectiveness of using DUO₂, such as DUCRETE, to reduce the size and weight of a dry-storage cask or silo for spent nuclear fuel (SNF).

DUCRETE consists of a DU ceramic that replaces the coarse aggregate used in standard concrete. The DUO₂ is a very dense, stable, low-cost, coarse aggregate that is combined with Portland cement, sand, and water in the same volumetric ratios used for ordinary concrete. If the ceramic can be produced at a low enough cost, it would be practical to consider using DUCRETE concrete as a shielding material.⁴ The cost of concrete cask fabrication is low when compared to fabricating steel, lead, and DU metal casks. DUCRETE concrete was conceived at the Idaho National Engineering and Environmental Laboratory (INEEL) by W. Quapp and P. Lessing, who jointly developed the process and were awarded both U.S. and foreign patents in 1998 and 2000, respectively.^{5,6} Emulating nuclear fuel technology, the sintered uranium oxide (UO_x) aggregate has a very high density (>95% theoretical density). Thus, a theoretical concrete density of 7.2 g/cm³ is possible.

Based on this conceptual work, J. Sterbentz of INEEL performed the first shielding calculations⁷ for DUCRETE in a spent fuel application. Figure 2 shows the nuclear shielding effectiveness of this conceptual DUCRETE shielding material. The figure shows comparisons of the relative effectiveness for gamma and neutron attenuation of DUCRETE and that of other common shielding materials in a proposed SNF storage silo or cask.

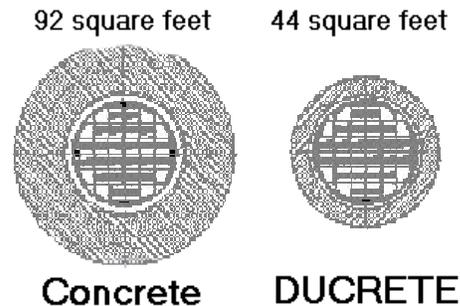
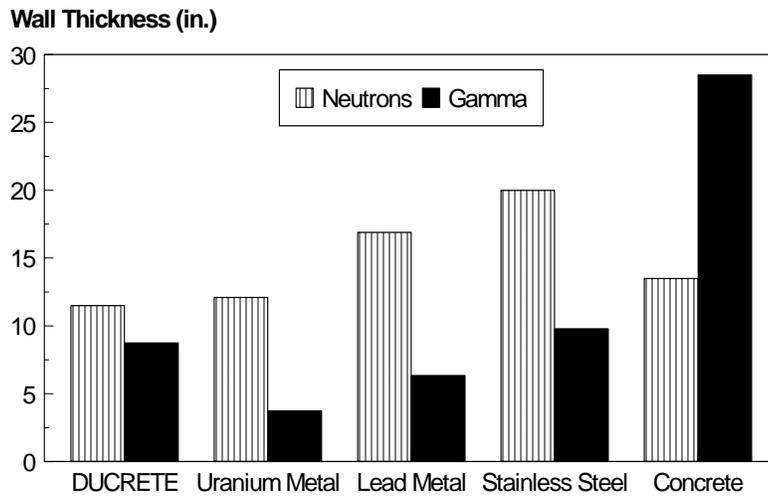


Fig. 1. Comparative diameters of concrete and DUCRETE dry-storage cask or silo. Using DUCRETE in a spent nuclear fuel cask or silo reduces the weight by 30%, the footprint by 50%, and the diameter from 132 in. (3.5 m) to 90 in. (2.3 m).

Another series of studies conducted by two firms in the U.S. private sector, Packaging Technology and Sierra Nuclear Company, provided a commercial perspective for the DUCRETE concept.^{8,9,10,11} Their results showed that if DUCRETE is found to be producible at relatively low cost, the concept appeared to be technically sound.



Dr. J. Sterbentz, INEEL

Fig. 2. Comparison of storage cask or silo wall thicknesses required to attenuate neutron and gamma doses from pressurized water reactor (PWR) spent nuclear fuel assemblies to 10 mR/h.

Sierra Nuclear Company found that spent fuel storage casks, with performance improvements and weight savings with their VSC-24 cask (Fig. 3), would nearly offset the greater fabrication costs.

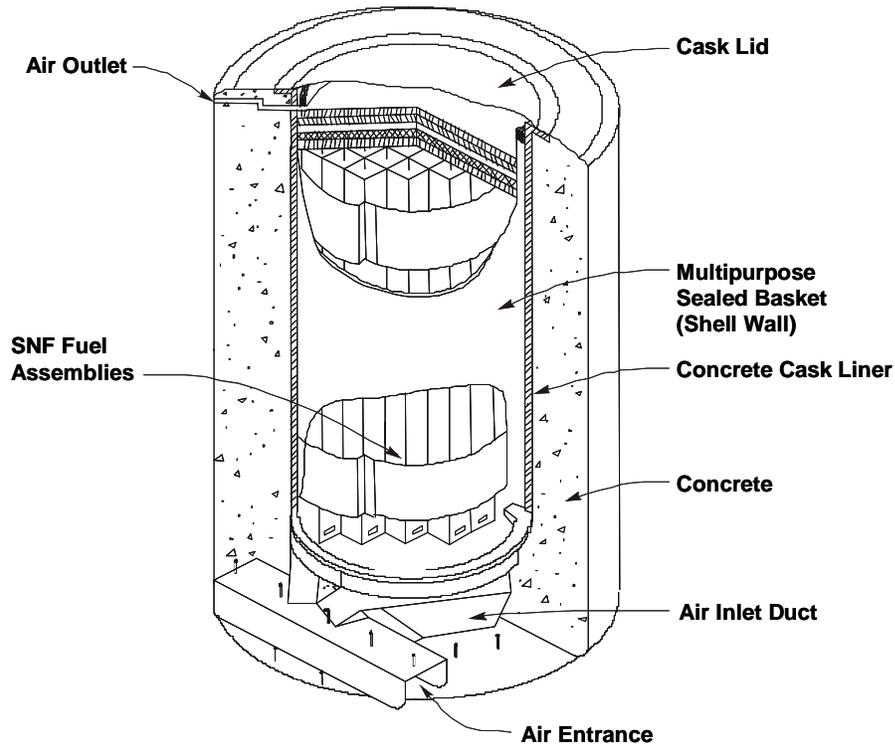


Fig. 3. Diagram of the Sierra Nuclear VSC-24 spent fuel cask. DUCRETE reduces the weight by ~20 tons (from >130 tons to <112 tons) and the footprint by a factor of 2x.

Reactor Spent Fuel Storage

Sierra Nuclear developed a conceptual model of its VSC-24 storage casks with DUCRETE. This dry-storage cask will store 24 pressurized water reactor (PWR) or 61 boiling water reactor (BWR) fuel assemblies. Through a series of calculations, Sierra Nuclear showed that such a storage cask is about 20% lighter than one made of ordinary concrete and has a much smaller footprint on the storage pad (see Fig. 1).

The DUCRETE storage cask had a diameter of 89 in. (226 cm), compared with 132 in. (335 cm) for standard heavy concretes, which use magnetite (iron) or barite (barium sulfate) for their dense aggregates.

The DOE Integrated Data Base Report for 1994¹² estimates the potential cumulative number of SNF assemblies expected from PWRs and BWRs through the year 2020. Shown in Fig. 4, these data indicate that low estimates conservatively predict as many as 268,000 total LWR SNF assemblies will be released. These assemblies require dry storage in some type of interim storage facility. These dry-storage facilities will be at the reactor sites and perhaps at the geological disposal site as well. Because of the events of September 11, 2001, it is anticipated that there will be a strong push to accelerate the removal of the current large backlog of LWR SNF assemblies from their current storage in at-the-reactor, wet-storage pools.

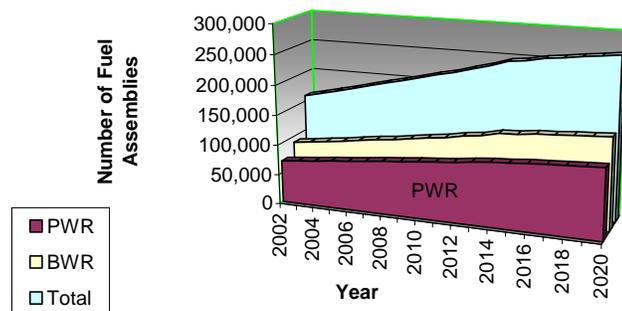


Fig. 4. Estimated cumulative number of SNF assemblies for PWR and BWR light-water reactors through 2020, low-case scenario.

Studies of the use of DUCRETE in these silos showed that as much as 55.68 tonnes of DUO_2 would be used in the DUAGG for each dry-storage cask or silo. This is the equivalent of 49.09 tonnes of DU, or 72.59 tonnes of DUF_6 , for each cask or silo. Given the conservative assumptions of 24 PWR and 61 BWR assemblies per cask or silo, the cumulative amount of DUO_2 that could be used in these dry-storage casks though 2020 is 408,455 tonnes (Fig. 5). This translates into 360,058 tonnes of DU or 532,473 tonnes of UF_6 to be used in the shielding of SNF assemblies in dry-storage casks or silos.

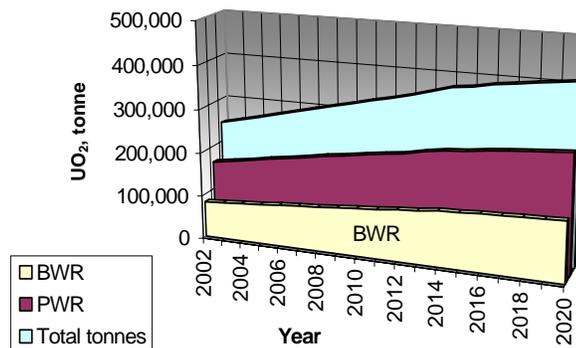


Fig. 5. Potential cumulative use of DUO_2 in SNF storage casks through 2020. Twenty-four PWR and 61 BWR assemblies per storage silo are considered.

Required Formation of Pelletized-DUO₂ Aggregates: DUAGG

The conversion and reduction of the stockpiled UF₆ produces DUO₂ powder or fine granules. Direct use of this fine DU-oxide has two limitations. First, concretes depend on their coarse aggregates to carry compressive stresses. If the shielding is required to have significant compressive strength (>4,500 psi), the powder must be sintered into dense aggregate-pellets with sufficient strength to be used in high-strength concretes. If the shielding is not required to provide compressive strength, as in some cask designs, this untreated DUO₂ powder still cannot be used to form even a low-strength, fine grout because of its chemical reactivity. The second limitation is that in an oxygenated environment (aerobic conditions), very dense DUO₂ inevitably oxidizes to form less dense UO₃ and even lighter U₃O₈ resulting in destabilizing expansions of the concrete/grout matrices. Therefore, the raw DUO₂ must be treated and formed for use in stable high-strength concretes.

Quapp and Lessing^{5,6} resolved these two issues by using a basaltic sintering agent that both reduced the pellets' sintering temperatures and made a protective coating of the DUO₂ that chemically stabilized the DUAGG in cement paste matrices. Therefore, they made it possible to make very stable, high-strength concretes with over three times the density of standard construction concretes.

This DUAGG process coupled with cask manufacturing is evaluated for its economic and technical viability in the subsequent sections.

ECONOMIC ISSUES

Preconceptual Plant Design and Cost Studies of DUAGG Production

The cost of producing DUAGG is an important consideration for any interested private firm in determining whether DUCRETE is economically viable as a material of construction in next-generation SNF casks. This study analyzed the project as if it were a stand-alone project; however, it was not intended to be a life-cycle cost analysis. The capital cost includes engineering design, equipment costs and installation, start up, and management.

This study¹³ (1) used previous DUAGG process developments to design a plant that will produce DUAGG at a baseline rate, (2) determined the size of the equipment required to meet the DUAGG production scale, (3) estimated the facility's capital and operating costs, and (4) performed a parametric sensitivity analysis on those elements of cost that most affect the total operating expenses. Because the study does not include preoperational, decontamination, decommissioning, and closure costs, it cannot be considered a complete life-cycle cost analysis. However, the purpose of this analysis is to establish the potential viability of the DUAGG process as a private commercial venture. To this end, we created a preconceptual baseline production plant equipment and layout described in Fig. 6.

Using DUO₂ as the feed material for this baseline DUAGG plant in Fig. 6, we assumed a production rate to support a commercial SNF market penetration of 30% in the domestic demand for casks. This would require sufficient DUAGG production to make 50 SNF casks a year, which requires 2,834 tonnes of DUO₂ to form 3,114 tonnes of DUAGG. Therefore, the equipment, site support facilities, and plant layout were also based on this production capacity. Capital and operating costs for the United States were determined based on the unit-operations of the equipment used in the flowsheet, the layout of the plant, and the labor requirements.

Table 1 indicates the total capital cost estimates, including engineering, piping, management, etc. The total estimated capital cost is \$11,601K. Most of the equipment can be readily obtained off-the-shelf from national vendors.

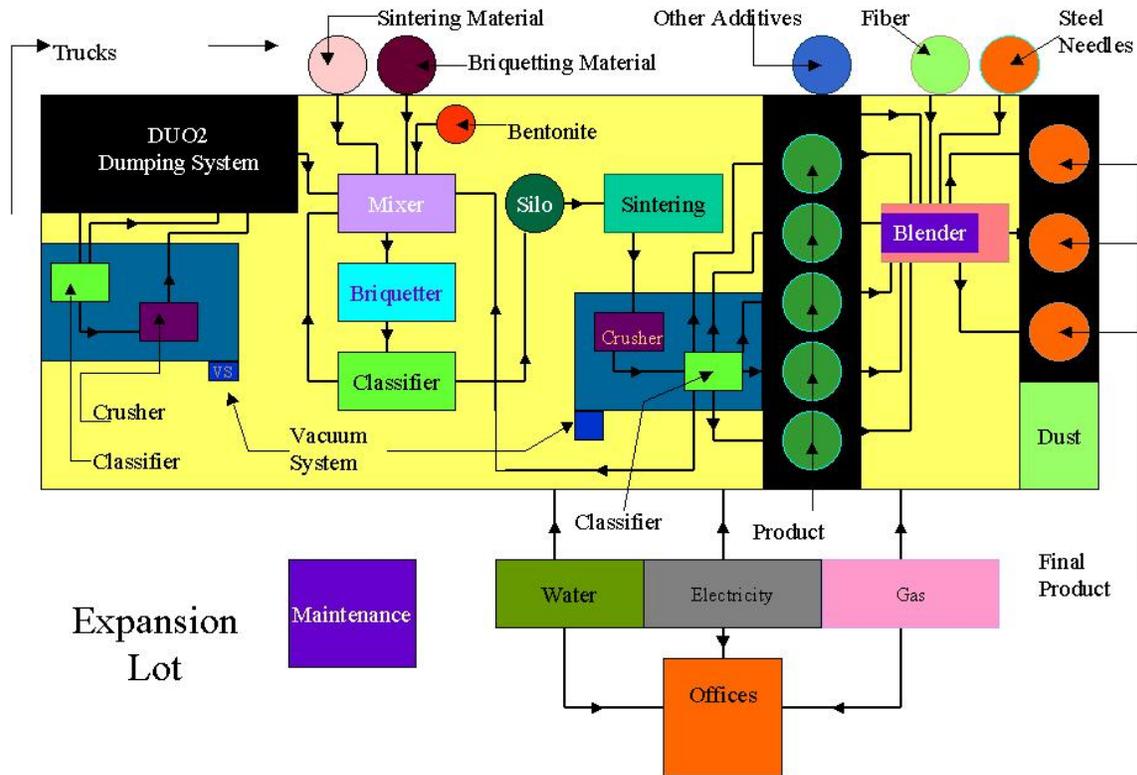


Fig. 6. Layout of the process equipment to produce DUAGG.

A summary of the results of this study shows:

- DUAGG cannot be produced at a cost that is competitive with conventional barium sulfate aggregate. The cost of DUAGG is ~\$840–\$2000/tonne whereas delivered graded barium aggregate is ~\$340/tonne. The cost for DUAGG in an advanced SNF cask is ~\$48,000–\$129,000; whereas, the goal for the total cost per cask is ~\$550,000.
- The commercial viability of DUAGG/DUCRETE depends on its enabling improved, unique cask performance characteristics. For example, DUCRETE may permit smaller, lighter-weight casks that can be transported by railcar. Conversely, DUCRETE may permit casks to contain more spent fuel assemblies at lower maximum temperatures within current volume and weight limits. DUCRETE may also enable the removal of the extensive matrix of rebar in current concrete cask designs.
- Operating costs dominate unit costs. Labor cost (at 62%) is the largest contributor to baseline operating costs. Capital cost recovery is ~36% of annual operating costs.
- Unit operating costs are sensitive to the credit of UO₂ feed materials. A credit of \$384/tonne reduces the unit cost by 17%.
- Operating costs (security, health physics, licensing) could be greatly reduced if the DUAGG fabrication plant were co-located with another uranium processing facility.

Table 1. Total capital cost estimate for the baseline case

Capital cost item	Cost estimate (\$K)
Civil/Site preparation	500
Utilities building services	56
Process equipment, land and buildings	4,844
Special process services	35
Engineering	1,591
Piping	1,204
Installation labor	1,205
Electrical	220
Spare parts	346
Management	1,000
Safety system	600
Total capital cost	11,601

Based on the sensitivity analysis, the estimated DUAGG costs are shown in Table 2.

Table 2. Estimated DUAGG costs

	\$384/tonne DUO₂ credit		Baseline: zero-cost DUO₂		\$384/tonne DUO₂ credit + savings for DUO₂ delivered as briquettes	
	Labor cost (\$/h)		Labor cost (\$/h)		Labor cost (\$/h)	
	80	40	80	40	80	40
Capital	\$11.6M	\$11.6M	\$11.6M	\$11.6M	\$8.9M	\$8.9M
Operating (year)	\$5.2M	\$3.8M	\$6.4M	\$4.7M	\$4.2M	\$2.4M
Unit (cask)	\$104K (\$1.67/kg)	\$76K (\$1.34/kg)	\$129K (\$2.27/kg)	\$94.9K (\$1.67/kg)	\$82K (\$1.32/kg)	\$48K (\$0.84/kg)

DUCRETE/Steel Cask Production Costs

A baseline flowsheet was developed with the series of stages involved in cask manufacturing. Figure 7 illustrates the cask manufacturing processes. The economic analysis estimates the production cost of SNF casks made with DUCRETE, assuming a commercial company would pursue the commercial venture of manufacturing these casks. This preliminary estimate was not intended to establish the life-cycle cost of a cask manufacturing project. Therefore, a complete study of the life duration of the plant has not been a part of this analysis. However, this study does include a baseline case and variations on the production cost due to changes in labor costs and other economic parameters of interest.

Due to limited funding, there was no attempt to optimize the flowsheet that had been developed in another case, and no credit was given for the avoided disposal costs or beneficial uses of DU_3O_8 .

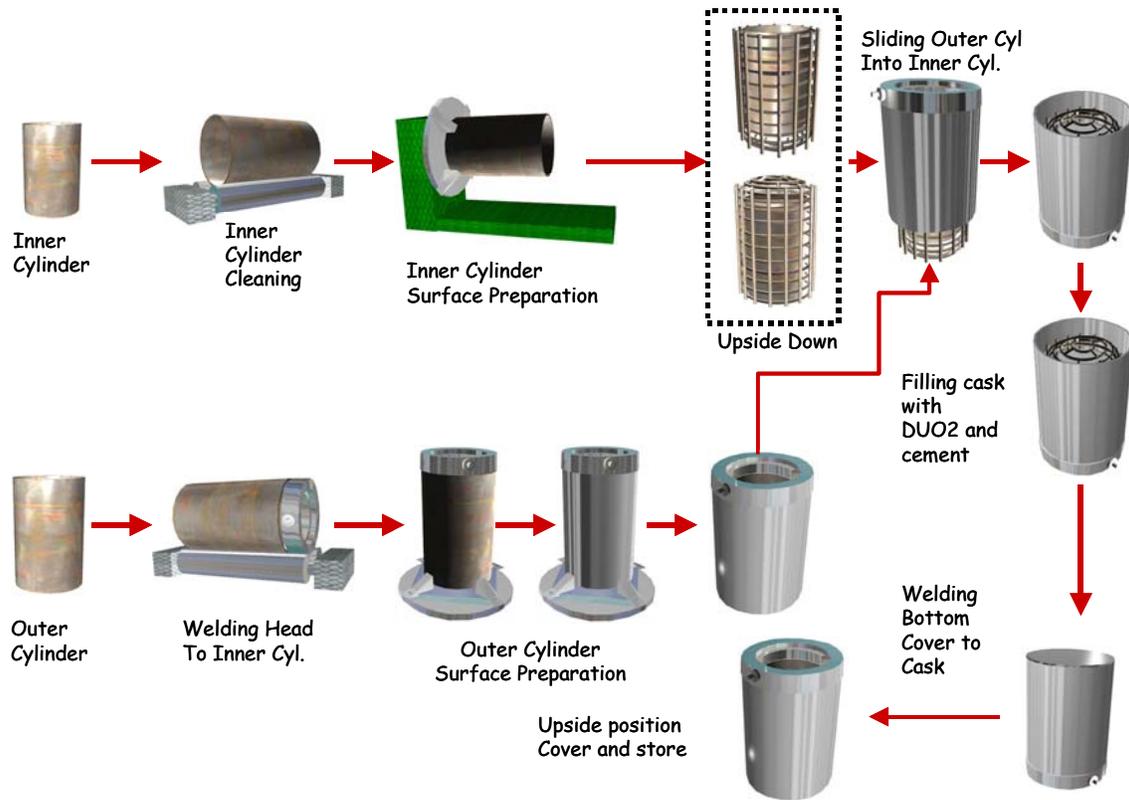


Fig. 8. Flowsheet showing the use of DUCRETE in the cask manufacturing, which encases the DUO_2 within a steel annulus.

This economic analysis focuses on (1) the design of a SNF cask plant that receives DUAGG for the required DUCRETE, (2) the DUAGG that will be used in high-strength DUCRETE for SNF casks, and (3) the potential cost elements that can vary when the project is in place. The process receives DUAGG from external source that most likely will be the DUF_6 conversion plant at Portsmouth or Paducah. The final product consists of SNF casks that are transportable and can be used for storage. In the future, a potential exists for the cask to be used for disposal purposes at the repository.

The production rate of this process will meet 30% penetration of the domestic market for SNF storage and transport casks (about 50 casks). The cask facility will receive prefabricated inner and outer cylinders, lids, and covers for the casks. It was assumed that 3 days would be necessary (1 shift per day) to completely produce one cask. The plant will work 5 days a week or 150 days per year (30 weeks per year or 1200 h/year). This production rate established the size of the equipment needed to implement the production schedule as well as the site support facilities and the plant layout. Based on the unit-operations equipment used in the flowsheet, the layout of the plant, the labor requirements, and the capital and operating costs were determined.

The operating cost estimates include (1) labor cost, (2) DUAGG cost, (3) cement cost, (4) capital recovery, (5) steel cost, (6) waste management cost, and (7) energy cost. Table 3 summarizes the operating cost for the baseline case.

Table 3. Baseline case estimates for the operating cost of DUCRETE cask manufacturing

Cost item	Cost estimate, \$/year
Labor	11,337,000
Steel (inner and outer cylinders, covers, lids, reinforcement @ \$33,000 per cask)	1,675,000
DUAGG (assumed the most conservative cost of the material @ \$128,000 per cask)	6,400,000
Cement	100,000
Waste management	500,000
Energy	100,000
Capital recovery (assumed 4 years of recovery or 25%)	4,620,000
Total operating cost, \$/year	24,732,000

Based on the total yearly operating cost of approximately \$24,750,000, the simple manufacturing cost per cask would be \$495,000. This simple manufacturing cost does not include licensing, marketing, transportation, or other significant costs, which bring the final costs of current, similar casks to \$1.0M to 1.2M in the United States. This result does shows that DUCRETE casks can be made at a cost that could be competitive in today’s market.

TECHNICAL ISSUES

Because of the possible reactions between the sintered DUO₂ particles and the cement pore solution, which is a very basic media (pH ~12.6) containing alkalies (Na and K), the stability of DUAGG pellets in concrete is a major concern. Since the potential reaction products of the uranium oxide and/or the constituents of the basalt-like binder could create expansive mineral growths, these expansive minerals could disrupt the concrete’s structure, generating cracks and causing spalling, similar to the alkali-aggregate (alkali-silica) reactions that are well known in standard concretes.

Testing conducted at the Oak Ridge National Laboratory¹⁴ (ORNL) has measured the extent and rates of surface reactions of the DUAGG under the expected service temperatures and the simulated chemical environments of cement pastes. Aggregates of DUAGG, obtained from the Starmet CMI (formerly Carolina Metals, Inc.), were tested for aggregate reactivity using a modified ASTM C289-94 method to measure interactions with the expected pore liquids that are anticipated in concrete pastes.

At a consistent surface to liquid ratio of 1:10, the sintered DUAGG pellets were exposed to (1) distilled/deionized water, (2) a 1N sodium hydroxide standard solution, and (3) a saturated water extract of high-alkali cement. Three exposure temperatures, 25°C, 66°C, and 150°C and six leaching time intervals were 30-, 60-, 90-, 180-, 240-, and 360-days were tested. At the end of each exposure period, the vessels in which the samples were leached were cooled to room temperature and opened. The liquid phase was separated and filtered prior to analysis for silica, uranium, alkalies, and other measurable elements by ion-coupled plasma atomic emission

spectroscopy (ICP-AES). The quantities leached were compared to the initial content of a DUAGG pellet to determine the degree of corrosion of the aggregate surfaces. Consistent with the guidelines of ASTM C295-98, the surfaces of the exposed aggregates were subsequently examined and compared by scanning electron microscopy (SEM) and energy dispersive X-ray fluorescence (EDX) analyses. The extent of surface corrosion, alteration, and formation of secondary minerals was used to indicate the potential for aggregate-paste interactions in DUCRETE concretes.

In order to determine whether any deleterious phases were formed, the surfaces of the samples at 2, 3, 6 and 13 months of exposure were examined by SEM equipped with EDX. The observations made indicate that except for the samples kept in deionized (DI) water (Fig. 8A), the surface of the DUAGG is covered by crystals that mask the initial morphology of the sample. The images at the surface of the sample made with back-scattered electrons (BSE) allow the average atomic number of each particle to be determined: the DU particles that have a high average Z will be seen as white on a BSE image. Using this feature, one can see that in the sample kept in DI water, the repartition of UO_2 particles is identical to that of the original unexposed DUAGG pellet. However, for the samples cured in NaOH or in cement pore solution, the BSE images do not show as many white areas, confirming that the surface is covered by crystal growth. The samples kept in DI water show some erosion of the surface, with a crystalline phase containing large amounts of titanium. This titanium phase, probably resembling rutile, is more resistant to leaching than the other basalt phases and appeared unaltered even after the other basalt phases were leached out.

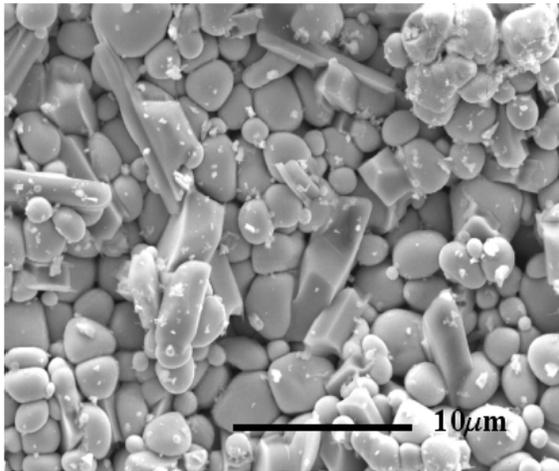
Of the chemicals that produce the basalt phase, aluminum and silicon are the elements that leach the most in the cement pore water, and this is even more striking in the 1 N NaOH solution. Both of these solutions are very basic, with pH above 12.6; at these high pHs, both alumina and silica are extremely soluble. The amount leached is higher for NaOH and increased with the temperature and time of exposure. This indicates that the dissolution of the basalt phase is progressive and does not produce a protective layer that would slow down the dissolution process. Observation using the SEM showed the presence of crystallized phases that contained Na, Al, Ti, Ca, and some Si and that resembled the crystals found in alkali-aggregate reaction (rosette type), as illustrated in Fig. 8B.

In the cement pore solution, the release of aluminum and silicon was elevated at the beginning of the test and at high temperature (about 10% for aluminum and silicon each). However, for all temperatures, the amount decreased with time, with less than 1% aluminum released at 6 months and less than 0.1% silicon released. This decrease can be explained by the formation of a protective layer of cement hydration products covering the surface of the DUAGG pellet. SEM examination of the pellets after 6 months of exposure confirmed that the grains of DUO_2 were almost invisible (Figs. 8C and 8D). This finding has been previously explained in studies of natural basalt rocks.^{15,16}

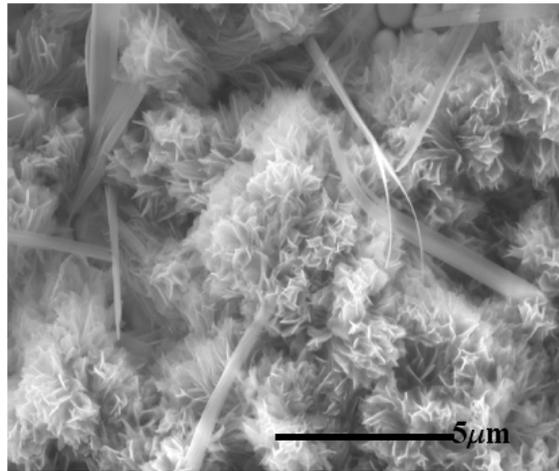
The leaching of other basalt constituents such as titanium and iron is not as pronounced. Of the two, iron is the element that is leached more (with ~2 % in NaOH at 150°C). At lower temperatures, the release is found to be less pronounced (between 0.25 and 0.7%). In the cement pore solution, the release of iron in solution is quasi-constant at 0.3%, regardless of the temperature or the time of exposure. Titanium shows the same pattern as iron, but its release is less abundant, with the maximum found at 0.15%.

Zirconium exhibits a very atypical behavior: the amount released is low (~0.04%) and is the same regardless of the temperature, the time of exposure, or even the leaching medium (as long as the

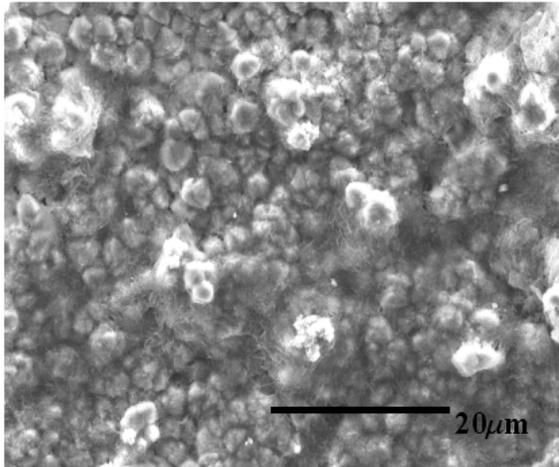
solution is alkaline). This finding indicates that the zirconium is probably not part of the glassy basalt phase but instead resides in separate crystals that are resisting the corrosiveness of the alkali solutions.



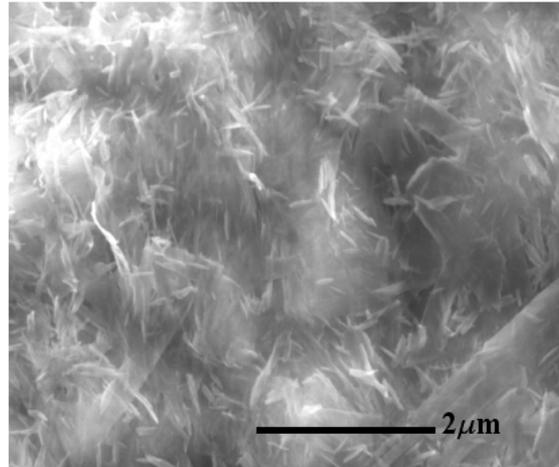
A DI Water - 150°C
secondary electrons
The oblong particles contain Ti and some Mg while the rounded ones are made of DU



B NaOH - 150°C
secondary electrons
Alkali-reaction products containing Na, Al, Si, Ca and Ti.



C Cement pore solution - 150°C
secondary electrons
Cement hydration products are covering the DU particles



D Cement pore solution - 150°C
secondary electrons
Cement hydration products contain Ca, Si and Al

Fig. 8. SEM images (secondary electrons) after 6 months at 150°C.

Uranium is not found to be leached in large amounts from the DUAGG pellet. Even though the pellet is composed of more than 90% uranium, a maximum of only 0.008% was leached. The NaOH solution is slightly more corrosive to the uranium than the cement pore solution is. As noted for almost all the other elements, the higher exposure temperatures are associated with greater releases of material. At 6 months, the results for the cement pore solution do not follow the trend developed in the previous periods of exposure. Because only one sample is obtained for

each time interval, the 1-year results will help determine whether a change in the reaction is actually occurring.

The corrosion of the DUAGG surface after 6 months at 150°C in saturated cement water appears to be minimal. A protective coating of cement hydration crystals covers the DU particles as well as the interstitial basalt. Thus far, no deleterious crystals have resulted from alkali-aggregate reactions. From the ICP-AES results, the amount of uranium leached from the DUAGG pellet appears to be very low. These results show that the stability of the DUAGG pellets is very good in cement pastes, at least after 6 months of exposure. Therefore, concretes containing DUAGG aggregates should be stable.¹⁷ However, more time is needed to verify these test results and extended-exposure tests are continuing as funding permits.

CONCLUSIONS

- Using depleted uranium dioxide aggregates in concrete for shielding is technically feasible using off-the-self processing and production technologies
- Using DUAGG/DUCRETE in casks used to store and dispose of SNF can reuse the nation's inventory of depleted uranium
- Using standard ASTM testing protocols for extended times and constant temperature show that the chemical and physical stability of DUAGG is adequate to ensure very long service lives for the composite casks
- Current storage, transport, and disposal schemes marginalize the potential savings from this reuse of depleted uranium in casks, which could result in much greater savings if they were to be integrated into an overall optimized SNF disposition scenario.
- A current incomplete cost analysis shows that the estimated simple manufacturing costs of DUCRETE storage casks, ~\$500K, are well within the current cask market cost, considering the very conservative assumptions used in this baseline case. (The simple manufacturing cost does not include licensing, marketing, transportation, or other significant costs, which bring the final costs of current, similar casks to \$1.0M to 1.2M in the United States.)
- These results show that DUCRETE casks can be made at a cost that could be competitive in today's market.

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