

ICEM05-1373

USES OF TAILS FROM NUCLEAR FUEL PRODUCTION IN AGGREGATES FOR USES IN SPENT NUCLEAR FUEL CONCRETE-STEEL TRANSPORT AND DISPOSAL CASKS

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ABSTRACT

The depleted uranium (DU) inventory in the United States exceeds 500,000 metric tons (tonnes). This paper reviews the status of the United States (U.S.) Department of Energy's (DOE's) 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 (SNFs).^{*} Just in the storage of commercial U.S. SNF, the cumulative amount of DU oxide (DUO₂) that could be used in nonmetallic matrix dry-storage casks through 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.

Uranium is a very effective gamma shield because of its high density and high atomic number (z). Aggregates made of UO₂ are combined with cementitious binders that enhance neutron shielding and result in high strengths. 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 paper describes the production of this shielding material and documents measurements of the physical properties and chemical durability of DUO₂ aggregates. Intact DUO₂ aggregates were tested for chemical reactivity with the cement paste, using a modified American Society for Testing and Materials (ASTM) C289-94 method to measure reactions with concrete pore liquids.

Using DUO₂ aggregates in concrete for shielding is technically feasible using off-the-self processing and production technologies. Results show that DUCRETE casks can be made at a cost that could be competitive in today's market.

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 United States (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

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

casks. Studies have assessed the use of uranium metal for shielding in both spent fuel [1] and high-level waste (HLW) [2] casks. A review of DU metal production and fabrication costs showed that DU 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.

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. DU oxides are used primarily for SNF storage casks. There is an additional benefit to the nuclear community because this use as shielding will consume a large quantity of DU from the existing national inventory.

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 SNF.

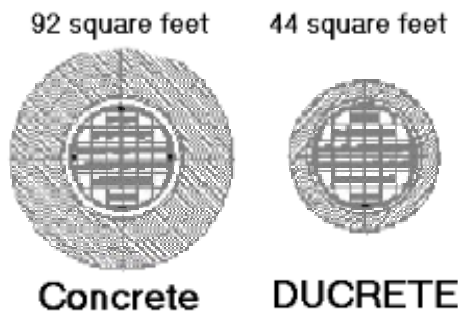


Fig. 1. Comparative diameters of concrete and DUCRETE dry-storage cask or silo. Using DUCRETE in an SNF cask or silo reduces the weight by ~15%, the footprint by 50%, and the diameter from 132 in. (3.35 m) to 90 in. (2.28 m).

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 with 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 calculations [7] for DUCRETE in a spent fuel application. Figure 2 shows the nuclear shielding effectiveness of this conceptual DUCRETE shielding material. The figure compares 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.

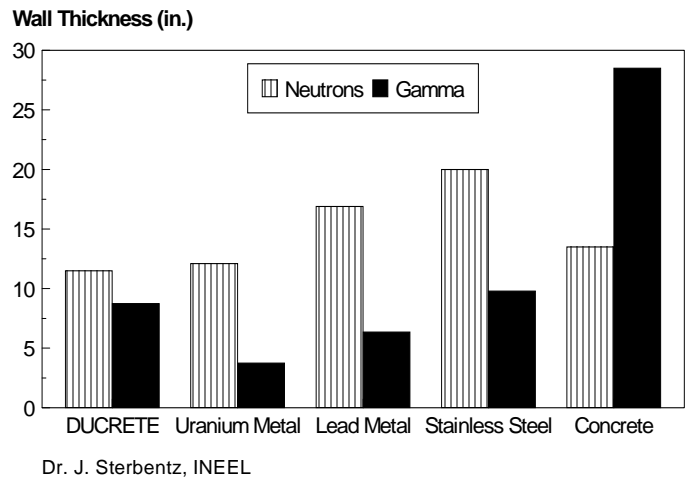


Fig. 2. Comparison of storage cask or silo wall thicknesses required to attenuate neutron and gamma doses from pressurized-water-reactor SNF assemblies to 10 mR/h.

SNF Cask Storage Studies

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.

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

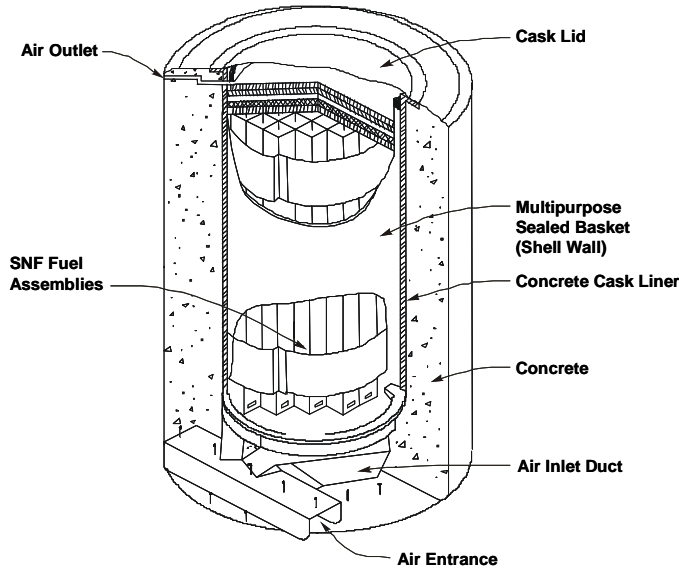


Fig. 3. Diagram of the Sierra Nuclear VSC-24 spent fuel cask.

SNF Storage Casks

Sierra Nuclear developed a conceptual model of its VSC-24 storage casks with DUCRETE. The use of DUCRETE reduces the weight of the VSC-24 cask by ~20 tons (from >130 tons to <112 tons) and the footprint by a factor of 2. This dry-storage cask is capable of storing 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 ~15% lighter than one made of ordinary concrete and has a much smaller footprint on the storage pad (see Fig. 1).

The Sierra Nuclear DUCRETE storage cask has a diameter of 90 in. (228 cm), compared with 132 in. (335 cm) for standard heavy concretes, which use magnetite (iron) or barite (barium sulfate) for their dense aggregates.

Potential Market

DOE's Integrated Data Base Report for 1994 [12] estimates the potential cumulative number of SNF assemblies expected from PWRs and BWRs through the year 2020. These data are shown in Fig. 4 and conservatively predict as many as 268,000 total light-water reactor (LWR) SNF assemblies will be discharged from reactors by 2020. These assemblies will 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 the removal of the current large backlog of LWR SNF assemblies from their current storage in at-the-reactor, wet-storage pools will accelerate.

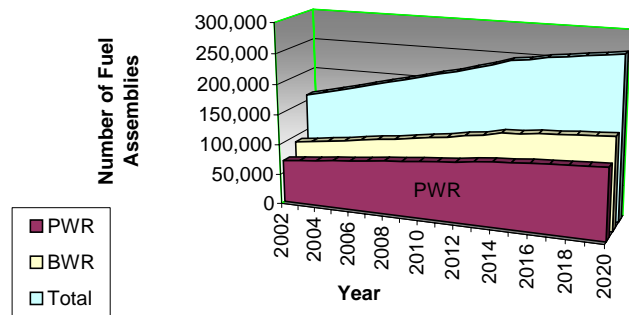


Fig. 4. Estimated cumulative number of SNF assemblies for PWR and BWR LWRs through 2020 (low-case scenario).

Studies of the use of DUCRETE in SNF storage silos showed that as much as 55.68 tonnes of DUO_2 would be used in each dry-storage cask or silo. This level of utilization is the equivalent to 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.

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 private firm interested 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, costs may be reduced if the DUAGG production facility is collocated at a nuclear facility. The capital cost in this study includes engineering design, equipment costs and installation, start-up, and management.

This study [13] (1) uses previous DUAGG process developments to develop a conceptual design of a plant that will produce DUAGG at a baseline rate, (2) determines the size of the equipment required to meet the DUAGG production scale, (3) estimates the facility's capital and operating costs, and (4) performs 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 ascertain the commercial viability of the project, an equipment layout for a preconceptual dry process baseline production plant is shown in Fig. 6.

Using DUO_2 as the feed material for the baseline DUAGG plant depicted in Fig. 6, a production rate is assumed to support an SNF market penetration of 30% in the domestic demand for casks. This would require sufficient DUAGG production to make 50 SNF casks per year, which requires 2834 tonnes of DUO_2 to form 3114 tonnes of DUAGG. Therefore, the equipment, site support facilities, and plant layout are based on this production capacity. Capital and operating costs are 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.

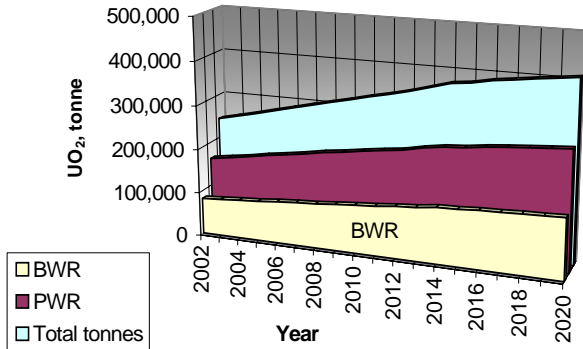


Fig. 5. Potential cumulative use of DUO_2 in SNF storage casks through 2020.

Required Formation of Pelletized- DUO_2 Aggregates: DUAGG

The process of conversion of the stockpiled UF_6 produces DU_3O_8 that can be reduced to DUO_2 powder or fine granules. DUO_2 is the basic material used in the formation of DUAGG, which is the concrete aggregate used to make the DUCRETE for the SNF casks. 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 (>4500 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 DUO_2 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 DUO_2 inevitably oxidizes to form less dense UO_3 and even lighter U_3O_8 , resulting in destabilizing expansions of the concrete/grout matrices. Therefore, the raw DUO_2 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 DUO_2 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.

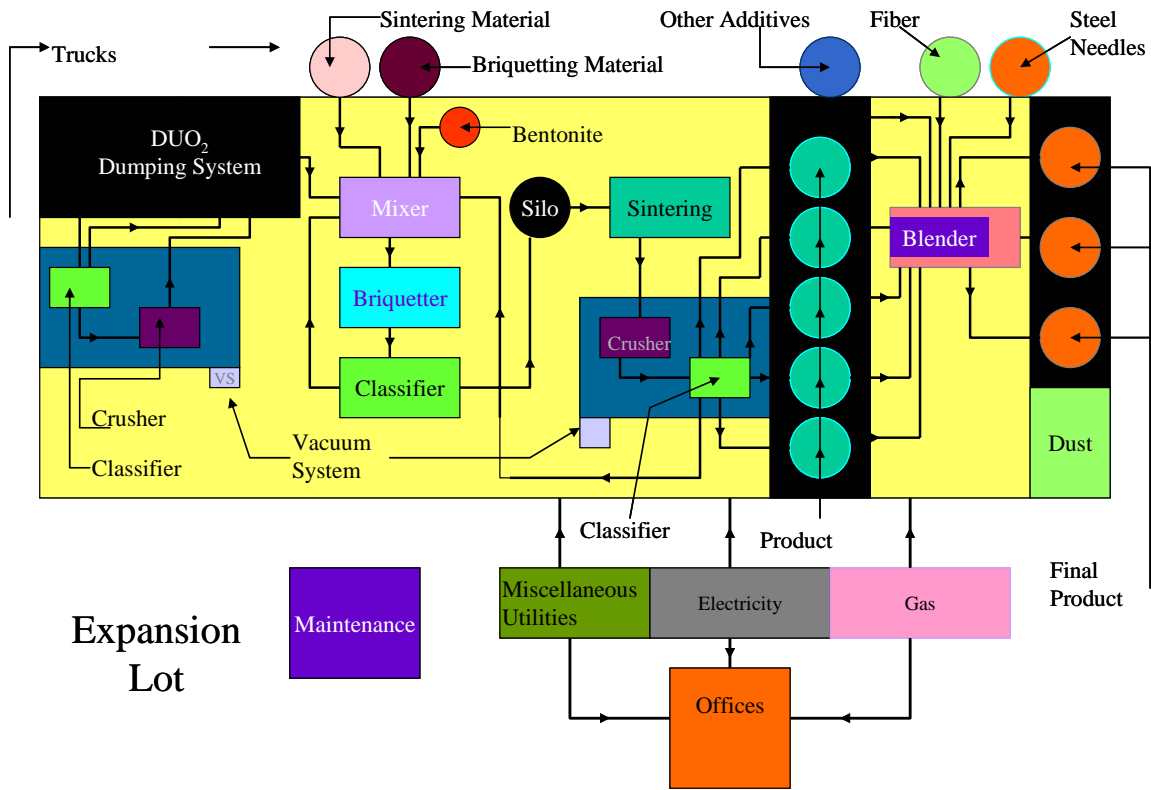


Fig. 6. Layout of the process equipment to produce DUAGG.

Table 1. Estimated capital cost for the baseline case

Capital cost item	Cost estimate (\$K) (2003 dollar value)
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

Conclusions of this study are as follows:

- 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, which leaves a margin for the completion of the production process that should result in a total cost per cask that meets the goal of ~\$550,000.
- The commercial viability of DUAGG/DUCRETE depends on its potential to enable 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. There are performance advantages, but it is fair to say at this point that it is sometimes difficult to estimate all the cost savings of the improved performance of the casks.
- 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, if any, of UO_2 feed materials. A credit of \$384/tonne reduces the unit cost by 17%. This credit is assumed for the cost avoidance of disposing of DU_3O_8 .
- Operating costs (security, health physics, licensing) could be greatly reduced if the DUAGG fabrication plant were collocated with another uranium processing facility.

A sensitivity analysis of the estimated DUAGG costs is shown in Table 2.

DUCRETE/Steel Cask Production Costs

A baseline flowsheet was developed with the unit operations involved in cask manufacturing. Figure 7 illustrates the cask-manufacturing process. The economic analysis estimates the production cost of SNF casks made with DUCRETE, assuming that a company would pursue the commercial venture of manufacturing these casks.

At this early stage of DUCRETE cask development and design, there was no attempt to optimize the

flowsheet and no credit was given for the avoided disposal costs or beneficial uses of DU_3O_8 .

The economic analysis focuses on (1) the design of a SNF cask plant that receives DUAGG for the production of DUCRETE, (2) the DUAGG that will be used in high-strength DUCRETE for SNF casks, and (3) potential cost elements that can vary when the project is in place. The process receives DUAGG from an external source that most likely will be the new DUF_6 conversion-to-oxide plant. 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.

It is assumed that the production rate of this process meets 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 is assumed that 3 days would be necessary (one 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 establishes the size of the equipment needed to implement the production schedule as well as the site support facilities and the plant layout. The capital and operating costs were determined based on the unit-operations equipment used in the flowsheet, the layout of the plant, and the labor requirements.

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.

Based on the total yearly operating cost of ~\$24,750,000, the simple manufacturing cost per cask would be \$495,000. This figure does not include licensing, marketing, transportation, or other significant costs. The ultimate goal is to sell storage casks for about half a million dollars. This result shows that DUCRETE casks can be manufactured and sold at a cost that could be competitive in today's market

DUAGG STABILITY

Testing conducted at the Oak Ridge National Laboratory [14] (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.

Table 2. Estimated DUAGG costs based on labor costs of \$80/h and \$40/h

	\$384/tonne DUO ₂ credit		Baseline: zero-cost DUO ₂		\$384/tonne DUO ₂ credit + savings for DUO ₂ 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)

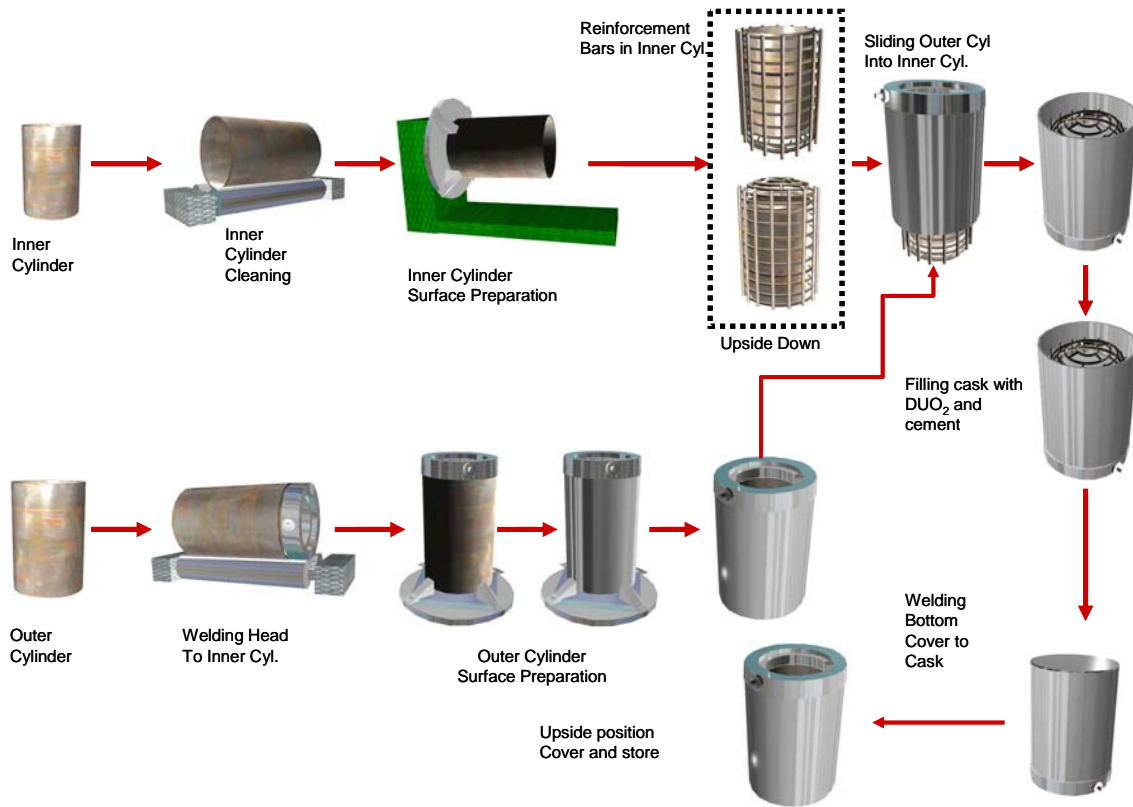


Fig. 7. Flowsheet showing the use of DUCRETE in cask manufacturing, which encases the DUO₂

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

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 in 1N NaOH. The NaOH leach solution is slightly more corrosive to the uranium than the cement pore solution. 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 [15].

CONCLUSIONS

- Using DUO₂ 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 represent a beneficial use of the nation's inventory of DU.

- Current storage, transport, and disposal schemes marginalize the potential savings from the beneficial use of DU in casks, which could result in much greater savings if they were to be integrated into an overall optimized SNF disposition scenario.
- The current incomplete cost analysis shows that the estimated simple manufacturing costs of DUCRETE storage casks are well within the current cask market cost, considering the very conservative assumptions used in this baseline case.

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