DUPoly: Depleted Uranium Polyethylene Encapsulation
Bench-Scale Processibility and Preliminary Characterization

Final Report
FY 1996

J.W. Adams, P.R. Lageraaen, P.D. Kalb, and B.R. Patel

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ABSTRACT

Depleted uranium (DU), in form of UO_3 powders, was encapsulated in low-density polyethylene using a single-screw extrusion process. DU was oven dried to remove residual moisture prior to processing. Waste and binder materials were fed by calibrated volumetric feeders to the extruder, where the materials were thoroughly mixed and heated to form a homogeneous molten stream of extrudate. The encapsulated DU, known as DUPoly, was then cooled in cylindrical molds for performance testing and round disks for attenuation studies. Waste loadings as high as 90 wt% DU were successfully achieved. A maximum product density of 4.2 g/cm^3 was achieved using UO_3, but increased product density using UO_2 is estimated at 6.1 g/cm^3. Additional product density improvements up to about 7.2 g/cm^3 are estimated using a hybrid technique known as micro/macroencapsulation. Waste form performance testing included compressive strength, water immersion and leach testing. Compression test results are in keeping with measurements made with other waste materials encapsulated in polyethylene. Leach rates were relatively low (0.07- 1.1%) and increased as a function of waste loading. However, considering the insolubility of uranium trioxide, these leach data indicate the probable presence of other, more soluble uranium compounds. Ninety day water immersion tests showed sensitivity to one type of UO_3 ("batch" processed) for samples containing >85 wt% of the oxide. Samples containing UO_3 produced by a newer "continuous" process showed no deterioration at up to 90 wt% waste loadings.

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EXECUTIVE SUMMARY

Depleted uranium (DU) is a residual material that results from the enrichment of uranium ore in the making of nuclear fuel. The U.S. Department of Energy (DOE) maintains large inventories of depleted uranium at several sites. Approximately 560,000 metric tons of DU in the form of UF_6, containing an equivalent mass of 379,000 metric tons of uranium DU are stored at the DOE Paducah, Portsmouth and Oak Ridge Gaseous Diffusion plants. Some of the UF_6 has been converted to uranium oxide (e.g., UO_3) and about 20,000 metric tons of DU are currently stored at the Savannah River Site. Novel applications are currently being sought to convert these materials to stable, useful secondary products. Uses which provide a positive benefit to society while allowing potential recovery or extraction of the uranium are desirable, but techniques for stabilization of DU for long-term storage or disposal are also being evaluated. Potential beneficial end-uses (e.g., shielding, ballast, industrial counterweights, energy storage flywheels) will likely exploit the high density, shielding effectiveness and nuclear applicability of these materials. This study, in particular, was initiated to investigate the feasibility of processing depleted uranium (UO_3 powder) by polyethylene microencapsulation, to mitigate potential health effects and produce useful radiation shielding and other products.

Brookhaven National Laboratory (BNL) has successfully developed polyethylene encapsulation extrusion processing for many types of low-level radioactive, hazardous, and mixed wastes. During processing, waste materials are mechanically mixed into the molten polyethylene binder, producing a workable homogeneous product. The process has evolved from proof-of-principle, through bench-scale development and testing, to full-scale technology demonstration and technology transfer. This project examines the feasibility of encapsulating DU in polyethylene for secondary end-use applications and/or disposal.

DU in form of UO_3 powders, was encapsulated in low-density polyethylene using BNL's single-screw extrusion process. The UO_3, obtained from the Westinghouse Savannah River Site, was oven dried to remove residual moisture prior to processing. Waste and binder materials were fed by calibrated volumetric feeders to the extruder, where the materials were thoroughly mixed and heated to form a homogeneous molten stream of extrudate. The encapsulated DU, known as DUPoly, was then cooled in cylindrical molds for performance testing and round disks for attenuation studies. Waste loadings as high as 90 wt% DU were successfully achieved. A maximum product density of 4.2 g/cm^3 was achieved using UO_3, but increased product density using UO_2 is estimated at 6.1 g/cm^3. Additional product density improvements up to about 7.2 g/cm^3 are estimated using a hybrid technique known as micro/macroencapsulation. Waste form performance testing included compressive strength, water immersion and leach testing. Compression test results are in keeping with measurements made with other waste materials encapsulated in polyethylene. Leach rates were relatively low (0.07- 1.1%) and increased as a function of waste loading. However, considering the insolubility of uranium trioxide, these leach data indicate the probable presence of other, more soluble uranium compounds. Ninety day water immersion tests concluded that water absorption was inconsequential except for "batch process" UO_3 samples at >85 wt% waste loadings. "Continuous process" UO_3 samples were relatively benign to water immersion with no indication of deterioration at even the highest (90 wt%) waste loading.

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1.   INTRODUCTION

Department of Energy (DOE) facilities maintain large inventories of depleted uranium (DU). Novel applications are currently being sought to convert these materials to stable, useful secondary products. Uses which provide a positive benefit to society while allowing potential recovery or extraction of the uranium are desirable, but techniques for stabilization of DU for long-term storage or disposal are also being evaluated. Potential beneficial end-uses will likely exploit the high density, shielding effectiveness and nuclear applicability of these materials. This study, in particular, was initiated to investigate the feasibility of processing depleted uranium (e.g., UO_3 powder) by polyethylene microencapsulation, to mitigate potential health effects and produce useful radiation shielding and other products.

Natural uranium ore in the form of U_3O_8 contains about 0.7 percent of the isotope ^{235}U, with the remainder of uranium present as ^{238}U. Reactor fuel is produced from the ore by converting it to uranium hexafluoride gas and enriching the proportion of ^{235}U to around 3.5 percent, leaving the remaining portion depleted in ^{235}U. This residual material, with ^{235}U concentrations at around 0.25 percent, is known as depleted uranium (DU). Approximately 560,000 metric tons of DU in the form of UF_6, containing an equivalent mass of 379,000 metric tons of uranium DU, are stored at the DOE Paducah, Portsmouth and Oak Ridge Gaseous Diffusion plants. Some of the UF_6 has been converted to uranium trioxide (UO_3); about 20,000 metric tons of DU are currently stored at the Savannah River Site. UO_3 from Savannah River was used in this preliminary investigation.

Alternatives for management of this inventory under consideration by the U.S. DOE include 1) continue current management plan (no action); 2) revise current practices for long-term storage as either UF_6 or in an oxide form; 3) use of DU in shielding or high-density applications; 4) disposal of DU.[1] Since uranium and uranium oxides are considered valuable resources, use of the material (option 3) is most attractive. DU is an excellent radiation shielding material for nuclear and medical applications. High density applications proposed include use as energy storage flywheels, armor, projectiles, and ballast/counterweights in airplanes, helicopters, and missiles.

Treatment of DU materials by polyethylene encapsulation is a desirable option because of the immediate availability of the technology and proven record to effectively and efficiently process similar powder and granular materials. In addition, the process is very flexible. Polyethylene products can be heated and reworked if future needs change. DU can also be retrieved from DUPoly by thermal processing if needed as a resource in the future. Over the last twelve years, Brookhaven National Laboratory (BNL) has extensively developed the polyethylene encapsulation extrusion process for low-level radioactive, hazardous, and mixed wastes.[2,3,4,5,6,7,8,9] During processing, waste materials are mechanically mixed into the molten polyethylene binder, producing a workable homogeneous product. The process is not susceptible to chemical interactions between the waste and binder, enabling a wide range of acceptable waste types, high waste loadings, and technically simple processing under heterogeneous waste conditions. The process has evolved from proof-of-principle, through bench-scale development and testing, to full-scale technology demonstration and technology transfer.

2.   DUPOLY PROCESSING

2.1   DU Sample

Representative samples of depleted uranium materials from Westinghouse Savannah River Company were used for treatability testing. The inventory at Savannah River Site (SRS) alone consists of about 20 million kg (20,000 metric tons) of depleted uranium trioxide (UO_3) stored in some 35,000 (55 gallon) drums. This inventory consists of material corresponding to two different evaporation processes (batch and continuous) used to prepare the oxide. Approximately 99% of the SRS inventory is comprised of batch process material.

Two drums of batch processed UO_3, labeled drums C1066 and C1070, were received at BNL on January 29, 1996. The net weight of each drum was approximately 650 kg (1430 lb). Four subsamples, approximately 35 kg (75 lb) each, were taken from each drum and transferred into 20 liter (5 gal.) plastic buckets. In each lot, the bright yellow powders were free-flowing with little to no lumps. A sample of the continuous process UO_3 was received at BNL on March 5, 1996. A smaller 90 kg (200 lb) sample of this material was requested to simplify handling at BNL and to reduce shipping costs. The continuous process powder was also yellow but with a slight gray tint, and was somewhat inhomogeneous, containing clumps or hardened regions of noticeably brighter yellow colored material. This material was received in two 20 liter (5 gal.) shipping pails, net weight approximately 45 kg (100 lb) each.

The UO_3 inventory at SRS was characterized in a recent report by Carolina Metals, Inc.[10] The drummed material was generically described as a 200 mesh (74 μm average particle size), 96.5% uranium trioxide with trace impurities of aluminum, iron, phosphorous, sodium, silicon, chromium and nickel. The material has a bulk density range of about 2.5 g/cm^3 (158 lb/ft^3), uncompacted, to 3.5 g/cm^3 (223 lb/ft^3), compacted. The ^{235}U content was assayed at approximately 0.2% and the plutonium content at 3 ppb. Gross gamma was 53,100 dpm per gram of uranium. The two sample lots differ only in their particle size distribution, the continuous process material having a slightly larger mean particle size. No quantification of the particle size distribution was performed at BNL as specific particle size data was already published by Carolina Metals.

Moisture contents of the as-received powders were determined prior to extrusion processing as past experience has indicated excessive water volatilization if the moisture content of the bulk powder exceeds 2 wt% (for vented extrusion processing). Both batch process and continuous process samples were oven dried at 160ºC for 24 h to determine their respective dry weights. Moisture content of batch process material was measured to be 0.4 wt% while continuous process material was 1.6 wt%. Low temperature differential scanning calorimetry was also done on samples of the two lots, heating at 2ºC/min from 20-160ºC. As-received batch process material showed no peaks in the 20-160ºC temperature range while as-received continuous process material showed characteristic endotherms at about 40, 50, 85, 95, 105, and 145ºC (Figure 1), evidence of low temperature reactions or phase changes occurring in the material.
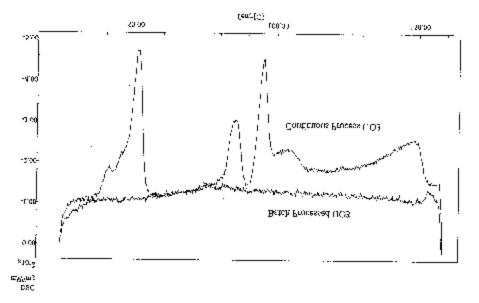
Figure 1.   Differential Scanning Calorimeter Output (mW/mg vs. ºC) for As-Received Batch and Continuous Process DU.

In contrast, samples of the dried materials (i.e., material heated at 160ºC for 24 h) showed no peaks in the 20-160ºC temperature range. While determination of the endothermic peaks is outside the scope of this effort, drying pretreatment was shown to produce a thermally stable product within the desired processing temperature range.

2.2   Equipment

Processing of depleted uranium was conducted by extrusion to assess the potential loading that can be incorporated in polyethylene. Extrusion is a robust thermoplastic processing technique that has been used extensively throughout the plastics industry in many applications. For this application, extrusion processing results provide an indication of the potential DU loading that can be achieved. Other processing techniques (e.g., thermokinetic mixing) may provide additional DU loading improvements. A 32 mm (1.25 in.) diameter single-screw, non-vented, Killion extruder, shown in Figure 2, was used for processibility testing. The extruder is equipped with a basic metering screw, three heating/cooling barrel zones and an individually heated die. DU and polyethylene are homogeneously mixed during processing in the extruder following simultaneous controlled feed metering using AccuRate, 300 Series, volumetric feeders. These feeders are designed to provide a constant volume output at a given operating setting that varies as a percentage from zero to 100% output. Feeder calibration is required for each material due to differing material densities and is conducted by recording the feeder output in grams over a one minute interval at five different feeder speed settings. Ten replicates are taken at each speed setting. The resulting data provides a plot of feeder output in grams per minute (g/min) versus feeder speed setting. During this study,

feeder calibrations were performed for the polyethylene and for each type of DU, i.e., batch process DU and continuous process DU.

Figure 2. Bench-Scale Killion Plastics Extruder.

2.3   Processibility Testing Procedure

Processibility testing included identifying key extrusion parameters such as temperature profiles (zone temperatures), feed and process rates, as well as monitoring product appearance, consistency and throughput. Current draw, melt temperature, melt pressure and extrudate product appearance were recorded at a constant extruder screw speed to gauge whether the material was amenable to extrusion processing. Note that 'extrudate' refers to the stream of molten product that exits the extruder through the output die. Monitoring these processing parameters along with visual observations of feeding, noise and output provided valuable information regarding the processibility of the DU. A number of different samples taken in replicate (typically ten) for statistical assurance were fabricated to quantitatively measure processing results. These samples are abbreviated as: *rate, grab, 2x4, and ALT*. Replicates of each

sample were taken sequentially and periodically throughout the processibility trials at given DU loadings. *Rate* and *grab* samples are used to monitor material processibility whereas *2x4* and *ALT* samples are used primarily to measure product performance. In addition to these processing and product samples, *disk* samples were also fabricated for possible future shielding and attenuation studies.

Rate samples were taken at one minute intervals to determine extruder output (g/min) and consistency over an extrusion trial. Low variation between replicate rate samples indicates that the output is continuous and that the material can be successfully processed at that DU loading. Typically, as the loading for a given waste is increased above its processibility limit, the output becomes discontinuous with noticeable surges.

Grab samples were taken periodically over an extrusion trial as small (approximately 3-10 g) representative specimens of the extrudate. The density of each grab sample was determined by weighing and using a Quantachrome Multipycnometer to measure their volume. Monitoring the product density is useful for quality control and to ensure homogeneity of the product. Low variation between replicate grab samples indicates that the DU material is feeding well and is consistently becoming well mixed with the polyethylene as it is processed in the extruder.

2x4 samples were fabricated as right cylindrical specimens for compressive strength and water immersion testing. The sample name refers to the nominal dimensions, 2 in. diameter by 4 in. height (5 cm x 10 cm) required for compliance with ASTM D695, "Compressive Properties of Rigid Plastics." For this project, these specimens were cast in pre-heated brass molds. Teflon plugs were inserted into the top of the mold after filling, then a slight compressive force was applied (max. 0.17 MPa (25 psi)). This technique produced smooth, uniform specimens.

ALT samples were fabricated for product leach testing in individual Teflon molds periodically throughout an extrusion trial. Samples were nominal 1 in. diameter by 1 in. high right cylinders (2.5 cm x 2.5 cm), as specified by the Accelerated Leach Test (ALT), ASTM C1308. These samples were molded under moderate compression of up to 1.72 MPa (250 psi). These samples were also used to determine DUPoly densities achievable when using a compression molding technique.

Disk samples were formed in circular glass petri dishes and molded under slight compression (max. 0.17 MPa (25 psi)). Disk samples were fabricated at varying thicknesses for possible future attenuation studies to determine the effectiveness of the product as a shielding material.

2.4   Processibility Testing Results

Processibility testing was conducted with samples representing both types of uranium trioxide in storage at Savannah River Site corresponding to the two different evaporation processes (batch and continuous) used in generating the oxide. Since the batch process depleted uranium represents over 99% of the SRS inventory and this was the first sample received at BNL, processibility testing initially focused on this material. Processibility testing concluded with extrusion trials of the continuous process DU.

2.4.1   Batch Process DU

Processibility testing with batch processed DU (batch DU) was initiated at a loading of 50 weight percent (wt%). This loading was selected based on previous experience with other materials and was expected to be readily achievable. Starting at this DU loading also enabled key process variables to be tuned for future attempts at higher DU loadings. If a maximum waste loading is attained or if a material is not readily processible, a number of conditions are observed such as an increase in die pressure, increased load or current draw on the drive motor, inconsistent output flow coupled with surging that can be observed on the ammeter and pressure transducer. Processing results at 50 wt% were not immediately successful because the extrudate appeared to contain entrained gas. Difficulties with void formation were observed in ALT and 2x4 product samples. It was believed that the off-gassing may have been caused by excess moisture in the DU that evolved during processing or due to air entrainment caused by agglomeration of the small DU particles during feeding. The moisture content of the DU was originally deemed not to be the cause of the off-gassing since the moisture content was measured to be 0.4 percent using a Sartorius Moisture Analyzer. Agglomeration effects were minimized during processing by starve feeding the DU and polyethylene to the extruder screw. This also ensured a more homogeneous feed and minimized segregation of the feed materials caused by their significant density differences. Utilizing this feeding technique permitted successful processing of the DU at this loading. These results prompted continued extrusion trials at increased waste loadings.

DU was also determined to be amenable to extrusion processing at a loading of 60 weight percent following some adjustments to the processing technique. At 60 wt%, surging was observed along with screw squealing and periodic gaseous discharges from the die indicating continued air entrainment or material off-gassing. In response, the DU feeder was recalibrated to ensure that the desired quantity of DU was being fed and processing conditions were modified by adjusting the feed rates and extruder screw speed to ensure the feed throat was maintained in a starved feed condition. The second feeder calibration plot was nearly identical to the initial calibration, confirming proper feeding conditions. The barrel zone temperatures were also lowered to attain a lower extruder temperature profile and cooling water was circulated through the feed throat. Each of these actions can potentially result in more consistent feeding and processing. However, altering these extruder operating parameters did not have a noticeable impact on the erratic processing results so, despite the low moisture content (0.4 wt%), the DU was oven-dried overnight at 160°C to remove any excess moisture.

Processing at 60 wt% with oven-dried DU produced excellent results. Some high pitched screw squealing that occurred prior to drying the DU persisted, but processing and product samples were not affected. Utilizing these processing parameters and dried DU, successful processing results were obtained at increasing waste loadings of 70, 75, 80, 85 and 90 weight percent. It was noted that the extrudate or product appearance did gradually change with the increases in DU loading. As the loading was increased, the extrudate's glossy appearance waned. Since the glossy appearance is caused by the polyethylene, the results are expected as the actual quantity of polyethylene is reduced with increased DU loading. At 85 wt% and especially 90 wt% the extrudate had a rough texture with a discontinuous surface whereas at 80 wt% and below the surface appearance of the extrudate was relatively smooth. However, even at 85 and 90 wt% the DU was readily processible and could be successfully cast into process and product specimens.

Attempts to extrude 95 wt% DU were not successful due to plugging in the output die, causing output to cease and die pressure readings to rise above their alarm set point (3570 psi). The extruder is equipped with a pressure safety relief valve rated at 7500 psi. At this loading there was insufficient polyethylene to mix, wet and convey the DU through the extruder barrel. DU flow was stopped immediately after noting the plugged condition. The clog was voided within several minutes by introducing pure polyethylene to the screw. Current draw by the screw rose slightly during this episode, but remained within acceptable limits. Therefore, a loading of 90 wt% represents the upper limit for microencapsulating batch DU into a polyethylene matrix utilizing a continuous extrusion process. Alternative processing techniques that may potentially improve maximum loadings will be investigated in future activities.

2.4.2   Continuous Process DU

The UO_3 produced by a new continuous evaporation process at SRS is reportedly chemically identical to the batch UO_3 but characterized by a slightly larger particle size. Since larger particles can be more easily compounded/mixed during extrusion processing and based on the processing successes with the batch DU it was expected that the continuous process DU (continuous DU) would have equivalent or improved processibility compared with the batch DU. For the continuous DU, loadings of 70, 80 and 90 wt% were selected to document its processibility. Results were successful and replicate processing and product samples were fabricated at each waste loading using dried DU. From a visual perspective, the product output was darker in color than with the batch DU but product observations were similar. The glossy appearance of the product waned with increasing DU loading and at 90 wt% the extrudate retained the rough texture with a discontinuous surface as initially observed with the batch DU.

Throughout processing with either sample of DU, squealing of the screw persisted without a deleterious impact on processibility. The squeaks were not heard while purging the extruder with polyethylene prior to and between each run. It is believed that the squeaks were caused by the shearing of the molten DUPoly between the screw flights and the barrel wall.

The overall success encountered during processing both the batch and continuous DU can be seen in evaluating the rate and grab sample data. The results from the process rate samples taken during each processability trial are shown in Table 1. The actual extruder output rate in grams per minute is not significant in gauging processability of the DU since different screw speeds and feed rates were used but rather the low deviation and small errors between replicate samples at each loading should be noted. The low variation between replicate samples taken at each DU loading indicate that the DU processed continuously and consistently, and is therefore amenable to extrusion processing even at a loading of 90 wt%. The extrusion trials were conducted at screw speeds of either 60 or 65 rpm and at combined (waste + binder) feed rates between 100 and 120 g/min. The feed rates were increased with each extrusion trial and DU loading increase, since extruders operate on a volume basis and the actual volume of feed material decreases with an increase in DU loading.

The grab samples which were taken during each processability trial are used to determine the density of the extrudate and to monitor extrudate homogeneity throughout an extrusion run. The data for the grab samples for all extrusion trials is shown in Table 2. For each DU sample at each waste loading, low deviation and errors were obtained between replicate samples indicating that the DU product is homogeneous and that the DU consistently becomes well mixed with the polyethylene as it is processed in the extruder. Despite the rough texture and discontinuous surface of the extrudate observed at 90 wt%, grab sample values indicate that the extrudate is still homogeneous. The actual density values increased with increasing DU loading, as expected.

Table 1. Process Rate Samples for Batch and Continuous Process DUPoly.

Waste Loading	Rate (g/min)	Std. Dev.	2σ Error	% Error
Batch DU (10 replicates per waste loading)				
50	114.23	3.45	2.47	2.16
60	109.93	2.71	1.94	1.76
70	111.69	3.37	2.41	2.16
75	117.78	1.48	1.06	0.90
80	125.63	2.27	1.62	1.29
85	124.13	2.87	2.05	1.65
90	120.60	2.36	1.69	1.40
Continuous DU (10 replicates per waste loading)				
70	110.41	2.10	1.50	1.36
80	113.45	1.97	1.41	1.25
90	117.87	3.85	2.75	2.34

Table 2. Grab Sample Densities for Batch and Continuous Process DUPoly.

Waste Loading	Density (g/cm³)	Std. Dev.	2σ Error	% Error
Batch DU (10 replicates per waste loading)				
50	1.50	0.04	0.03	1.89
60	1.73	0.02	0.01	0.80
70	2.13	0.04	0.03	1.42
75	2.50	0.03	0.02	0.88
80	2.70	0.09	0.07	2.46
85	2.98	0.04	0.03	1.05
90	4.21	0.05	0.04	0.84
Continuous DU (10 replicates per waste loading)				
70	2.34	0.03	0.02	1.03
80	2.86	0.03	0.02	0.84
90	4.03	0.07	0.05	1.16

3.   PRELIMINARY PRODUCT CHARACTERIZATION

This project proposes to characterize DUPoly for comparison with other high density/radiation shielding materials and to assess its potential commercial applicability. Tests conducted during FY96 were intended as a preliminary indicator of product strength, durability and leachability. Characterization included density measurement, compressive strength testing, accelerated leach testing and 90 day water immersion testing.

3.1   Density Measurement

Densities of all DUPoly samples prepared were measured. For all but the "grab" samples, density was calculated as sample mass divided by geometric volume. Test samples measured include nominal 2x4 right cylinders, both uncompressed samples formed in polyethylene containers and compressed samples formed in heated brass molds; 1x1 inch right cylinders (ALT samples), formed either uncompressed using 2.5 cm (1 in) diameter copper tubing as a mold, or under pressure using Teflon molds; and nominal 11.7 cm (4.6 in) diameter disk samples, as described above. The data shown in Table 3 represent the mean and 2σ values for each sample type and DU loading. At least 10 each of the 2x4 and 1x1 samples were measured for a given DU loading. Typically 6-8 disk samples, representing three different sample thicknesses, were measured for each DU loading.

Table 3. DUPoly Sample Densities (g/cm³).

DU type/wt%	disk compressed[1]	2x4 uncompressed	1x1 uncompressed	2x4 compressed[1]	1x1 compressed[2]
batch/50wt%	1.38 ± 0.06	1.38 ± 0.02	1.43 ± 0.02	1.62 ± 0.02	NA[3]
batch/60wt%	1.62 ± 0.05	1.66 ± 0.06	1.61 ± 0.04	1.83 ± 0.02	1.85 ± 0.04
batch/70wt%	1.87 ± 0.10	2.08 ± 0.10	NA	2.05 ± 0.04	2.18 ± 0.03
continuous/70wt%	2.19 ± 0.05	NA	NA	2.26 ± 0.02	2.34 ± 0.01
batch/75wt%	2.26 ± 0.11	2.28 ± 0.12	2.34 ± 0.11	2.39 ± 0.04	2.59 ± 0.07
batch/80wt%	2.45 ± 0.21	2.76 ± 0.16	2.68 ± 0.03	2.71 ± 0.03	2.99 ± 0.04
continuous/80wt%	2.80 ± 0.06	NA	NA	2.79 ± 0.03	3.01 ± 0.03
batch/85wt%	2.97 ± 0.06	2.94 ± 0.28	NA	3.03 ± 0.06	3.44 ± 0.03
batch/90wt%	3.93 ± 0.08	NA	NA	3.94 ± 0.06	4.25 ± 0.04
continuous/90wt%	3.67 ± 0.17	NA	NA	3.86 ± 0.07	4.14 ± 0.04

1. Formed at ≤ 0.17 MPa (25 psi) pressure.

2. Formed at ≤ 1.72 MPa (250 psi) pressure.

3. Sample not available.

3.2   Compressive Strength Testing

Compressive strength testing is a means of quantifying the mechanical integrity of a material. Force is exerted uniaxially on an unconstrained cylindrical sample until the sample fails. For this application, the maximum strength of the material as well as the mechanism by which it fails (plastic deformation vs brittle fracture) are of interest. Further, compressive strength can be useful to assess waste form performance following environmental testing. The Nuclear Regulatory Commission has recommended that licensable solidification processes must demonstrate a minimum waste form compressive strength of 0.41 MPa (60 psi). Hydraulic cement waste forms must exceed 3.45 MPa (500 psi) to be considered for licensing.[11]

Eight to eleven DUPoly 2x4 waste forms at each DU loading were compression tested in accordance with ASTM D-695, "Standard Test Method for Compressive Properties of Rigid Plastics." Compressive testing was done using a Soiltest hydraulic compression tester at an unloaded crosshead deflection rate of 1.3 ± 0.3 mm (0.05 ± 0.01 in.)/min. Crosshead speed and total deflection were monitored using a dial gauge and lab timer. Load and deformation were recorded at 60 second intervals. Mean compressive yield strengths and % deformation at yield are given in Table 4 for each of the DU types and waste loadings prepared. All samples failed by ductile fracture; these values represent maximum compressive yield strength for the forms.

Table 4. DUPoly Compression Test Results.

DU type/wt%	Compressive Yield Strength (psi)	Compressive Yield Strength (MPa)	% Deformation at Yield
batch/50wt%[1]	2500 ± 222	17.2 ± 1.53	25.8 ± 4.16
batch/60wt%[2]	2280 ± 119	15.7 ± 0.82	20.2 ± 1.78
batch/70wt%[1] continuous/70wt%[4]	1940 ± 136 2420 ± 174	13.4 ± 0.94 16.7 ± 1.20	NA[3] 19.2 ± 3.64
batch/75wt%[1]	2190 ± 140	15.1 ± 0.97	16.1 ± 1.89
batch/80wt%[1] continuous/80wt%[4]	2290 ± 31.8 2420 ± 101	15.8 ± 0.22 16.7 ± 0.70	13.6 ± 0.76 14.1 ± 1.22
batch/85wt%[4]	2290 ± 122	15.8 ± 0.84	NA[3]
batch/90wt%[4] continuous/90wt%[5]	2940 ± 131 2850 ± 127	20.3 ± 0.90 19.7 ± 0.88	6.6 ± 0.40 7.1 ± 0.57

1. Mean ± 2 sigma error for eight replicate samples.
2. Mean ± 2 sigma error for eleven replicate samples.
3. Data not available.
4. Mean ± 2 sigma error for ten replicate samples.
5. Mean ± 2 sigma error for nine replicate samples.

3.3   Leachability Testing

DUPoly forms containing 50, 70 and 90 wt% batch process UO_3 were tested in accordance with the Accelerated Leach Test (ALT). This ASTM standard method (ASTM C1308), developed at BNL[12], was devised to enable prediction of a sample leach rate providing data fits a diffusion-controlled model. In this model, leaching may be accelerated by preheating the leachants above ambient temperature (i.e., 35-65ºC). In theory, the effective diffusion coefficient (D_e) is temperature dependent according to the Arrhenius equation: $D_e = A\exp(-E_a/RT)$, where E_a is the activation energy. Due to limited scope, however, all leach testing was done at ambient temperature.

Samples tested were nominal 2.5 cm x 2.5 cm (1x1) right cylinders. The test procedure specifies 13 leachant changes (nominally 3000 ml of distilled water) over an 11 day period. Specimens are suspended using monofilament line approximately into the center of each solution. Each series tested includes 3 replicates of each sample. A pretest was done on a representative sample of each DU loading to determine probable leachate metal concentrations. Following this, it was decided that the 50 and 70 wt% DUPoly samples would be leached in 300 ml of leachate (a 10x concentration of the leached metals)

while the 90 wt% DUPoly sample would be leached in the full 3000 ml volume to eliminate solubility effects. A source term sample was leached concurrently to determine whether leachate saturation conditions were being reached. This sample was comprised of 50.0 g of depleted UO_3 powder agitated in 3000 ml of water. A 20.0 ml aliquot was removed at each leach sampling interval. In this case, the leachate was not refreshed at every interval.

Leachates were analyzed by inductively coupled plasma (ICP) spectroscopy for their total uranium metal concentration. Results of the metals analyses were evaluated using the ALT computer program which calculated the Incremental Fraction Leached (IFL), Cumulative Fraction Leached (CFL), and the diffusion coefficient that best fits the leaching data. Both incremental and cumulative leach fractions from the replicate samples are given in Table 5. Below each set of data is the calculated diffusion coefficient.

Table 5. Accelerated Leach Test Results for 50, 70, and 90 Wt% Batch Process DUPoly.

50WT%DUPOLY;25C								
Time	Incremental Fraction Leached				Cumulative Fraction Leached			
(days)	sample 4	sample 7	sample 11	mean IFL	sample 4	sample 7	sample 11	mean CFL
0.083	1.23e-05	1.60e-05	1.25e-05	1.36e-05	1.23e-05	1.60e-05	1.25e-05	1.36e-05
0.292	3.96e-05	4.61e-05	5.78e-05	4.78e-05	5.19e-05	6.22e-05	7.03e-05	6.14e-05
1.00	8.90e-05	8.82e-05	9.67e-05	9.13e-05	1.41e-04	1.50e-04	1.67e-04	1.53e-04
2.00	4.94e-05	5.81e-05	6.44e-05	5.73e-05	1.90e-04	2.08e-04	2.31e-04	2.10e-04
3.00	4.44e-05	4.29e-05	5.16e-05	4.63e-05	2.35e-04	2.51e-04	2.83e-04	2.56e-04
4.00	5.78e-05	6.09e-05	5.86e-05	5.91e-05	2.92e-04	3.12e-04	3.42e-04	3.15e-04
5.00	5.31e-05	5.73e-05	5.90e-05	5.64e-05	3.46e-04	3.70e-04	4.01e-04	3.72e-04
6.00	4.92e-05	4.66e-05	4.90e-05	4.83e-05	3.95e-04	4.16e-04	4.50e-04	4.20e-04
7.00	7.05e-05	6.90e-05	6.93e-05	6.96e-05	4.65e-04	4.85e-04	5.19e-04	4.90e-04
8.00	6.13e-05	6.29e-05	6.89e-05	6.44e-05	5.27e-04	5.48e-04	5.88e-04	5.54e-04
9.00	5.39e-05	5.83e-05	5.87e-05	5.70e-05	5.80e-04	6.06e-04	6.47e-04	6.11e-04
10.0	5.32e-05	5.25e-05	5.41e-05	5.33e-05	6.34e-04	6.59e-04	7.01e-04	6.64e-04
11.0	4.55e-05	5.25e-05	4.82e-05	4.87e-05	6.79e-04	7.11e-04	7.49e-04	7.13e-04
	Diffusion Model							
	D (cm/sec)	Error (%)						
sample 4	7.49e-14	3.77						
sample 7	8.27e-14	3.36						
sample 11	9.06e-14	2.62						

Table 5. Accelerated Leach Test Results (Continued).

70WT%DUPOLY:25C							
Time	Incremental Fraction Leacdhed				Cumulative Fraction Leached		
(days)	sample 13	sample 16	sample 17	mean IFL	sample 13	sample 16	sample 17	mean CFL
0.083	4.43e-05	3.80e-05	3.92e-05	4.05e-05	4.43e-05	3.80e-05	3.92e-05	4.05e-05
0.292	5.18e-05	3.72e-05	4.12e-05	4.34e-05	9.60e-05	7.53e-05	8.04e-05	8.39e-05
1.00	1.15e-04	8.13e-05	8.48e-05	9.38e-05	2.11e-04	1.57e-04	1.65e-04	1.78e-04
2.00	7.15e-05	6.73e-05	7.51e-05	7.13e-05	2.83e-04	2.24e-04	2.40e-04	2.49e-04
3.00	5.62e-05	5.36e-05	5.43e-05	5.47e-05	3.39e-04	2.77e-04	2.95e-04	3.04e-04
4.00	4.45e-05	5.92e-05	6.49e-05	5.62e-05	3.84e-04	3.37e-04	3.59e-04	3.60e-04
5.00	5.72e-05	5.34e-05	5.83e-05	5.63e-05	4.41e-04	3.90e-04	4.18e-04	4.16e-04
6.00	5.37e-05	4.80e-05	5.07e-05	5.08e-05	4.94e-04	4.38e-04	4.69e-04	4.67e-04
7.00	6.17e-05	5.59e-05	6.02e-05	5.93e-05	5.56e-04	4.94e-04	5.29e-04	5.26e-04
8.00	6.24e-05	5.90e-05	5.86e-05	6.00e-05	6.19e-04	5.53e-04	5.87e-04	5.86e-04
9.00	5.16e-05	4.99e-05	5.25e-05	5.13e-05	6.70e-04	6.03e-04	6.40e-04	6.38e-04
10.0	5.26e-05	5.34e-05	5.32e-05	5.31e-05	7.23e-04	6.56e-04	6.93e-04	6.91e-04
11.0	5.06e-05	4.56e-05	4.70e-05	4.77e-05	7.73e-04	7.02e-04	7.40e-04	7.38e-04
	Diffusion Model							
	D (cm/sec)	Error (%)						
sample 13	8.90e-14	1.47						
sample 16	7.77e-14	2.10						
sample 17	8.56e-14	1.84						

90WT%DUPOLY;25C							
Time	Incremental Fraction Leacdhed				Cumulative Fraction Leached		
(days)	sample 2	sample 3	sample 4	mean IFL	sample 2	sample 3	sample 4	mean CFL
0.083	1.69e-04	1.69e-04	1.63e-04	1.67e-04	1.69e-04	1.69e-04	1.63e-04	1.67e-04
0.292	2.35e-04	3.10e-04	2.54e-04	2.66e-04	4.04e-04	4.79e-04	4.17e-04	4.33e-04
1.00	9.92e-04	1.07e-03	1.02e-03	1.03e-03	1.40e-03	1.55e-03	1.43e-03	1.46e-03
2.00	1.15e-03	1.27e-03	1.25e-03	1.22e-03	2.54e-03	2.82e-03	2.68e-03	2.68e-03
3.00	9.01e-04	1.09e-03	1.09e-03	1.03e-03	3.44e-03	3.92e-03	3.77e-03	3.71e-03
4.00	7.43e-04	8.47e-04	8.28e-04	8.06e-04	4.18e-03	4.76e-03	4.59e-03	4.51e-03
5.00	9.66e-04	1.06e-03	1.06e-03	1.03e-03	5.15e-03	5.82e-03	5.66e-03	5.54e-03
6.00	9.52e-04	1.11e-03	1.03e-03	1.03e-03	6.10e-03	6.93e-03	6.69e-03	6.57e-03
7.00	8.34e-04	9.49e-04	9.01e-04	8.95e-04	6.94e-03	7.88e-03	7.59e-03	7.47e-03
8.00	8.83e-04	1.03e-03	9.05e-04	9.39e-04	7.82e-03	8.91e-03	8.49e-03	8.41e-03
9.00	9.35e-04	1.08e-03	9.86e-04	1.00e-03	8.75e-03	9.99e-03	9.48e-03	9.41e-03
10.0	9.59e-04	1.05e-03	8.90e-04	9.67e-04	9.71e-03	1.10e-02	1.04e-02	1.04e-02
11.0	8.72e-04	9.45e-04	8.48e-04	8.88e-04	1.06e-02	1.20e-02	1.12e-02	1.13e-02
	Diffusion Model							
	D (cm/sec)	Error (%)						

sample 2	2.15e-11	4.49						
sample 3	2.46e-11	4.56						
sample 4	2.26e-11	3.86						

3.4 Immersion Testing

Water immersion testing was performed using one 2x4 and one 1x1 form of each DU type and waste loading. Samples were immersed in distilled water to determine possible deleterious effects of a water saturated environment. Three or four similar samples were grouped together in a single polyethylene container, with a water/sample ratio of 1000 ml per sample for 2x4 samples and 200 ml per sample for 1x1's. The test, done at ambient temperature, was a 90 day static immersion after which time the sample weights and volumes were re-measured. Samples remaining intact on completion of the test were compression tested to determine potential for non-visible degradation.

After 90 days, visible degradation was only evident on samples containing 85 and 90 wt% batch process DU (BPDU). Samples containing 80 wt% or less batch process DU were visibly unchanged, as were all samples containing continuous process DU (CPDU), up to 90 wt%. The 90 wt% BPDU samples began showing signs of cracking around the top and bottom perimeter within the first week of immersion. Cracks in the 85 wt% BPDU samples were not noticed until the third month of the test. After 90d, 90 wt% BPDU samples were severely deteriorated, with cracks running lengthwise penetrating nearly to the center of both samples (Figure 3). Cracking at both top and bottom surfaces resulted in creation of a solid cone at either end of the samples. After 90d, 85 wt% BPDU samples contained only three or four minor cracks (<1 cm) along the sample sides. Larger cracks were noted on the bottom surface of the 2x4 sample (Figure 4). Immersion solutions for batch process DUPoly samples were bright yellow in color, in contrast to continuous process DUPoly immersion solutions which were much more pale with a slight brownish tint.

Post-immersion compressive strengths of 50, 60, 70, 75, 80 and 85 wt% BPDU samples were 2450, 2460, 1390, 2390, 1980, and 1340 psi (16.9, 17.0, 9.6, 16.5, 13.6, and 9.2 MPa), respectively. Post-immersion compressive strengths of 70, 80 and 90 wt% CPDU samples were 2680, 2440, and 2640 psi (18.5, 16.8, and 18.2 MPa), respectively. Percent changes in sample mass, volume and compressive strength due to 90 day water immersion are shown in Table 6.
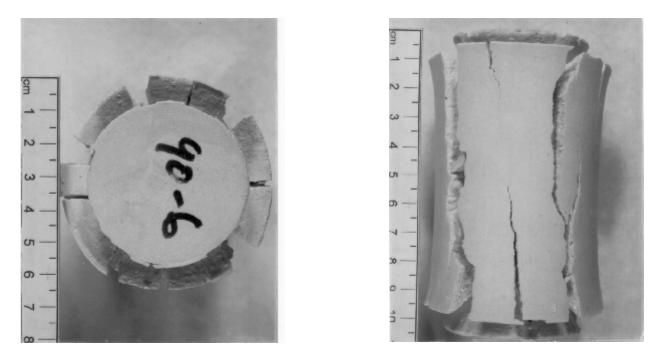
Figure 3. 2x4 Sample Containing 90 Weight Percent Batch Process UO₃, After 90 Day Water Immersion Test.
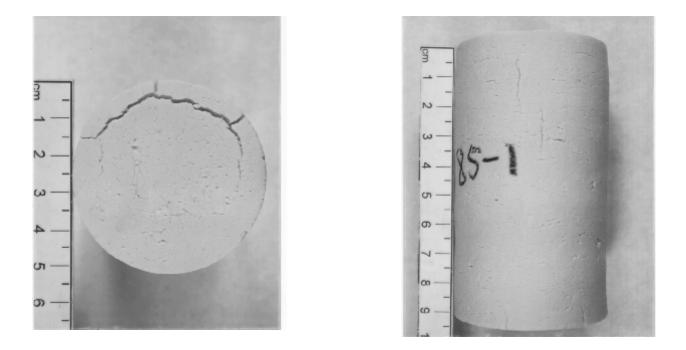

Figure 4. 2x4 Sample Containing 85 Weight Percent Batch Process UO₃, After 90 Day Water Immersion Test.

Table 6. DUPoly Immersion Test Results.

DU type/wt%	Percent Change in Sample Mass[1]	Percent Change in Sample Volume[1]	Percent Change in Compressive Yield Strength[2]
batch/50wt%	+0.6, +0.2	-1.2, +0.3	-1.9
batch/60wt%	+0.5, +0.2	+0.5, +0.0	+7.8
batch/70wt%	+0.6, +0.3	+1.2, -0.4	-28.5
batch/75wt%	+1.0, +0.5	+3.8, +1.5	+9.1
batch/80wt%	+1.9, +1.8	+5.6, +3.9	-13.6
batch/85wt%	+4.6, +5.4	+14.7, +10.8	-41.6
batch/90wt%	ND[3], +11.0	ND, ND	ND
continuous/50wt%	ND, +0.1	ND, -1.8	ND
continuous/60wt%	ND, +0.1	ND, -3.2	ND
continuous/70wt%	+0.2, +0.1	-0.9, -0.2	+10.8
continuous/80wt%	+0.3, +0.2	-0.0, +0.4	+0.8
continuous/90wt%	+1.1, +0.5	+1.2, +0.2	-7.2

1. First value is for 1x1 sample; second value is for 2x4 sample.

2. Compressive strengths measured for 2x4 samples only.

3. ND = No Data (sample not measured).

4.   DISCUSSION

Unlike DUPoly extrusion processing, which seems relatively insensitive to DU loading even up to the maximum limits, the physical characteristics and performance of the DUPoly product show obvious effects of increased DU loading. Product density is the most characteristic difference between samples of different DU loadings. DUPoly densities ranged from 1.38 to 3.93 g/cm^3 for uncompressed samples (disk, 2x4, and uncompressed 1x1 forms) for the range of 50 to 90 wt% DU. Disk samples and 2x4 samples, although formed under compression, have relatively large surface areas and thus were formed under low pressure (<0.17 MPa (25 psi)), so that density values were very similar to uncompressed samples. Compressed 1x1 (ALT) forms, on the other hand, had densities which were consistently and significantly higher than other samples. Because of their relatively small size, these samples were compressed with up to 1.72 MPa (250 psi) pressure. The density increase observed by compressing these forms was

approximately 10-15%, with mean values ranging from 1.62 to 4.25 g/cm^3 for compressed forms at 50 to 90 wt% DU. DU density as a function of wt% DU loading is depicted in Figure 3 for both compressed and uncompressed samples. This data is obtained by multiplying the mean DUPoly density times the weight percent DU in the DUPoly for a given DU loading.

Initial process runs using batch process DU produced non-linear density curves, depicted as solid lines in Figure 5. DUPoly process runs using continuous process DU produced nearly identical values for compressed forms, whereas uncompressed sample densities differed somewhat from the corresponding batch process samples. This is probably an artifact of sample formation, allowing fewer/more voids while filling the molds, or using slightly more/less pressure during cooling. For both batch and continuous process DUPoly, DU densities for 90 wt% samples were higher than the reported density of a vibration compacted sample of the dry powder (3.5 g/cm^3). Uncompacted DU powder, which has a density of about 2.5 g/cm^3, is surpassed at about 80 wt% DUPoly for compressed samples and about 85 wt% for uncompressed DUPoly. In other words, at these waste loadings, the DUPoly process represents a volume reduction compared with disposal of untreated DU.
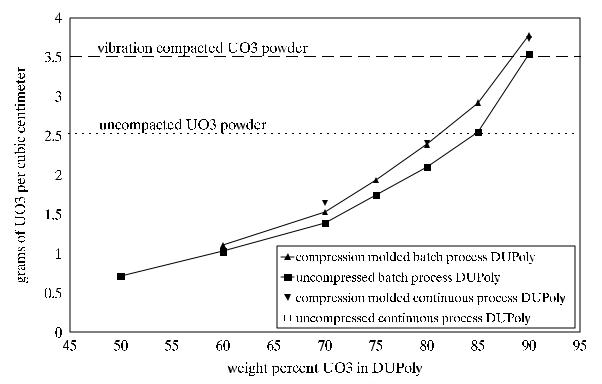

Figure 5. DUPoly Density versus DU Loading for Samples Prepared from UO$_3$.

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To quantify how much DU is in a drum of DUPoly compared to a drum of treated or untreated DU, simply divide the grams DU per cubic centimeter DUPoly by the grams DU per cubic centimeter in the form or container for the material to which it is being compared. Thus, for the highest density DUPoly forms achieved in these tests (90 wt% DU, compression molded forms), DU loadings are 1.08 times greater than vibration compacted DU powder, and 1.49 times greater than uncompacted DU powder. Ratios of >1 indicate that there is more DU in a DUPoly form than in the referenced material (DU powder) of an equivalent volume. To illustrate this point on a constant weight basis, the estimated volume for 1000 kg of DU stabilized in 90 wt% DUPoly would be 0.26 m^3, compared to a volume of 0.40 m^3 for uncompacted DU powder or 0.29 m^3 for vibration compacted DU powder. Such high product densities are achieved because of an increased volume packing efficiency for the DU particles during DUPoly processing. This effect may be attributed to one or more of the following factors: reduced particle agglomeration due to drying of the particles during thermal treatment; comminution of the particles due to mechanical abrasion during processing; or increased packing efficiency due to compressive forces exerted during forming.

Compressive yield strength is similarly plotted against DU loading in Figure 6. With batch and continuous process DUPoly data averaged together (filled squares), maximum yield strength is relatively constant between 50 and 85 wt% DU considering the range of measurement error. At 90 wt%, a statistically significant increase was noted, probably due to particle-to-particle contact of the DU in the matrix, with barely enough polyethylene present to fill void spaces. This fact is reflected in the percent deformation at yield, reduced from approximately 26% for 50 wt% DUPoly samples to only 7% for 90 wt% DUPoly samples.
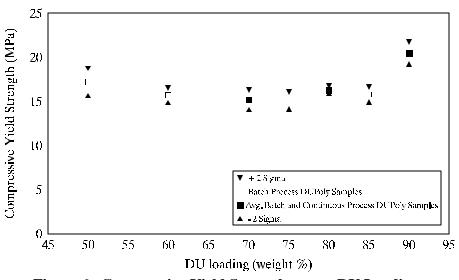

Figure 6. Compressive Yield Strength versus DU Loading.

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Accelerated Leach Testing of batch process DUPoly forms produced cumulative uranium releases of approximately 1.1% for 90 wt% DU and approximately 0.07% for both 50 and 70 wt% DU samples, after 12 days (Figure 7). These results are typical for waste materials microencapsulated in polyethylene. However, considering the insolubility of uranium trioxide in water[13], these data indicate the probable presence of other, more soluble uranium compounds. While the UO_3 is reportedly 96.5% pure (82.25-78.47% total U), it is likely that other soluble uranium salts are present and unaccounted for in the DU. Identification of these salts was beyond the scope of this effort. The high solubility of the as-received batch DU was further evidenced in that the source term leach sample (50 g batch process DU in 3000 ml water) saturated within the first (2 hr.) leach interval. Continuous process DUPoly samples were not tested. Additional ALT testing, e.g., of 75, 80 and 85 wt% batch process DU and corresponding continuous process DU samples, is recommended to optimize waste loadings with respect to sample leachability.

Ninety day water immersion tests concluded that water absorption was inconsequential except for batch process DU samples at very high (>85 wt%) waste loadings. Swelling and cracking in batch process DU are probably related to the same phenomenon observed in leach testing, i.e., presence of soluble compounds. In contrast, continuous process DU showed little evidence of leaching or swelling/cracking during 90 day immersion testing even at the highest waste loading of 90 wt%. Therefore, continuous process DU provides a significantly more stable and durable product.
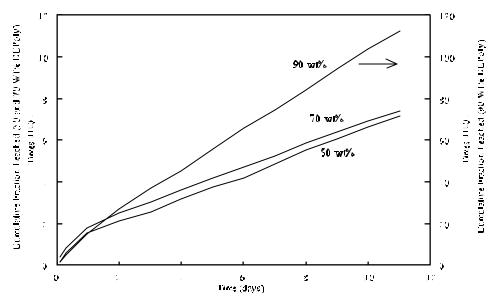

Figure 7. ALT Leach Results for Batch Process DUPoly Samples.

5.   POTENTIAL PROCESS IMPROVEMENTS

Alternative processing techniques can be used to improve the final polyethylene encapsulated DU product. Options for treated DU include re-use (as radiation shielding, counterweights in aviation and nautical applications, etc.) or disposal as low level radioactive waste. In either case it is desirable to maximize the amount of depleted uranium that can be loaded into the final product while maintaining the physical and performance characteristics required of the product. Greater depleted uranium loading is indicated by higher DUPoly product densities which also translates into enhanced shielding properties, smaller counterweights and lower disposal costs due to volume reduction.

Preliminary process optimization in order to maximize the DU loading for the polyethylene extrusion technology was conducted for this study. However, uranium packing efficiency may be further enhanced resulting in improved depleted uranium loading and/or DUPoly density through the use of several processing options, applied individually or combined. These include:

- compression molding techniques
- kinetic mixing to enhance extrusion processing
- use of uranium oxide powders (e.g., UO_2, U_3O_8) with higher densities than UO_3
- pelletization of uranium oxide powders for use as an aggregate additive to supplement the microencapsulated DU
- pelletization and sintering of uranium oxide powders prior to use as an aggregate additive

One approach successfully demonstrated in this study involved applying pressure to compress the DUPoly extrudate prior to solidification. Results at compression pressures up to 1.72 MPa (250 psi) showed higher densities for the compressed DUPoly product compared to the non-compressed, for the same weight percent DU loading. This translates into a greater quantity of depleted uranium within the same volume of product as discussed earlier. Further investigation of the effects and limitations of compression molding is recommended.

In previous work, BNL identified thermokinetic mixing as a potential alternative or supplement to extrusion processing for microencapsulation in polyethylene.[14] This process relies on high shear mixing and kinetic energy to homogenize and melt the mixture. Investigation of the use of thermokinetic mixing technology is recommended for removal of residual moisture, potential improvement of DU loadings, and improved mixing.

This investigation used depleted uranium in the form of UO_3 powders currently stored at SRS. Alternatively, conversion of UF_6 to the oxide form can be controlled to form oxides of higher density.[15] For example, the theoretical densities of UO_2 and U_3O_8 are 10.9 g/cm^3 and 8.3 g/cm^3, respectively, compared with a theoretical density of 7.3 g/cm^3 for UO_3. Potential improvement in product densities and volumetric loading of DU using UO_2 are projected in Figure 8.

The processing of DU performed for this project involved microencapsulation, in which individual DU particles are encapsulated within a polyethylene binder to form a homogeneous product.

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Macroencapsulation involves the encapsulation of larger particles within a plastic coating. Another technique to improve DU loading and resultant product densities is to supplement the microencapsulation treatment with pelletized DU aggregate. In other words, solid DU aggregate in the form of pellets or briquettes could be macroencapsulated with DUPoly in a hybrid micro/macroencapsulation process. By choosing to use the DUPoly extrudate (microencapsulated DU) as the binder material for macroencapsulation, a greater overall DU packing efficiency can be achieved for the final product compared to that of compressed DUPoly alone. Additional testing would be required to determine the total volume of depleted uranium that can be effectively incorporated into a micro/macro product. Several factors must be determined including achievable density of compacted DU pellets or briquettes, percent volume of DU pellets or briquettes that can be successfully encapsulated, and loading of the DU within the DUPoly binder. As seen in Figure 9, preliminary calculations indicate that improved DU loadings can be achieved for a micro/macro DU product (density of 4.6 g/cm^3) assuming 90 wt% DU in the DUPoly, 50 volume% DU briquettes and a briquette density of 5 g/cm^3 (twice the bulk density of DU). Similarly, as shown in Figure 10, even greater DU loadings can be attained if UO_2 is used to formulate the micro/macro product yielding an estimated product density of 6.8 g/cm^3.

A variation of the micro/macroencapsulation approach discussed above involves sintering uranium oxide powders at high temperature and pressure to achieve aggregate densities within 90% of the theoretical crystal densities. This approach was pioneered by INEL to form a dense aggregate for use with their DUCrete process.[15] Applying this technique in conjunction with micro/macroencapsulation of UO_2 would potentially yield the highest DUPoly waste loadings and densities. This is shown in Figure 11, which assumes a sintered aggregate density of 8.40 g/cm^3 (for ground UO_3 powder sintered at 1,250°C in a dry H_2 atmosphere), resulting in a predicted DU product density of 7.24 g/cm^3.

Each of the options discussed in this section are compared on an equivalent basis (using the bulk density of UO_3) in Figure 12. Assuming a disposal scenario, this plot shows potential for reductions in volume using the various alternatives, compared with the baseline of simply storing DU in a 55 gallon drum. The micro/macro DU processing alternative has the potential for incorporating the greatest volume of DU compared to all other alternatives, especially if sintered DU aggregate is used. Further investigation is suggested to confirm these engineering projections and quantify potential loadings and densities of optimized DUPoly products.
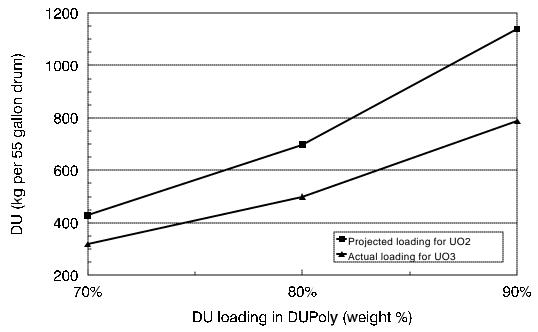
Figure 8. Projected Comparison of Loading Efficiency for UO₂ Based on Microencapsulation of UO₃.
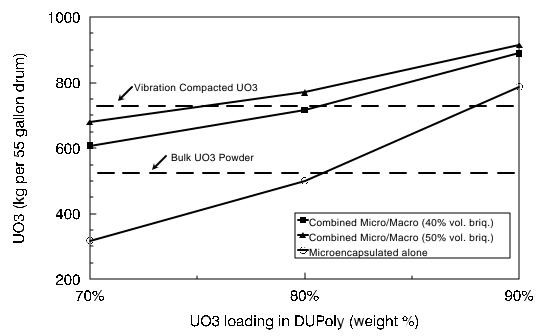
Figure 9. Comparison of DUPoly Microencapsulation with the Projected Loading for a Hybrid DUPoly Micro/Macroencapsulation Technique as a Function of UO₃.
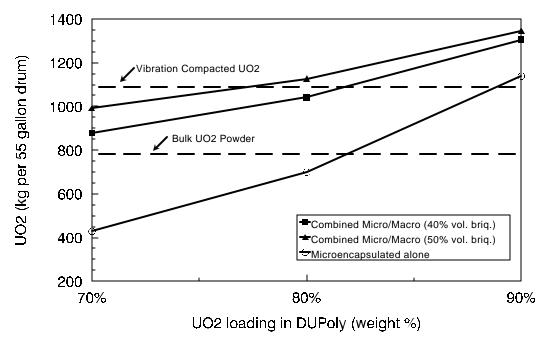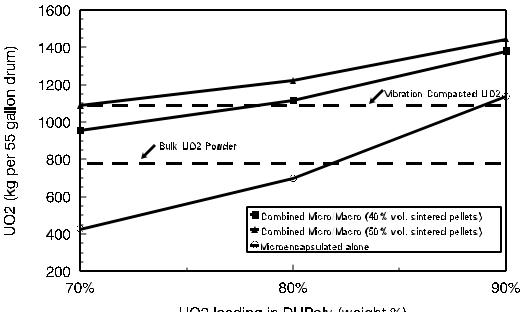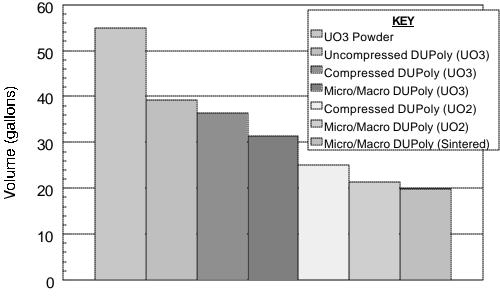
Figure 10. Comparison of DUPoly Microencapsulation with the Projected Loading for a Hybrid DUPoly Micro/Macroencapsulation Technique as a Function of UO$_2$.

Figure 11. Projected Comparison of DUPoly Microencapsulation (UO$_2$) with a Hybrid Micro/Macroencapsulation Technique using Sintered UO$_2$.

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Figure 12. Projected Volumes of Equivalent Quantities of UO$_3$ for Various Processing Alternatives.

6. CONCLUSIONS

This report summarizes bench-scale extrusion and preliminary characterization of polyethylene encapsulated depleted uranium (DU). Extrusion process runs were conducted over the range of 50 to 95 wt% DU using both batch process and continuous process depleted UO$_3$ obtained from the Savannah River Site. Processing using a non-vented extruder required pretreatment drying to guarantee uniform and reproducible process results, despite the relatively low as-received moisture contents of the powders (0.4-1.6 wt%). In these tests, DU was oven dried at 160ºC, equivalent to the maximum process temperature, for a period of at least 18 hours. Moisture problems can typically be circumvented using a vented extrusion process or a thermokinetic mixer, whereby small amounts of entrained gases are removed before the molten material reaches the output die of the extruder.

Process runs at 50 to 75 wt% DU produced extrudate which appeared dense and relatively fluid, with an obvious plastic appearance and characteristic (i.e., flows in a continuous stream). Runs at 80 wt% and higher produced increasingly rough extrudate surfaces, an observable indication that the plastic to DU ratio is lessening. Despite this appearance, even at 90 wt%, the material processed continuously and the process continued to successfully encapsulate the DU powder particles. Extrusion runs at 95 wt% DU were unsuccessful when the DUPoly mixture clogged the screw, causing output to cease and die pressure to rise excessively.

DUPoly product density increased significantly as a function of DU loading and also sample compression during molding. Mean densities ranged from 1.38 g/cm^3 at 50 wt% DU to 4.25 g/cm^3 at 90

wt% DU. Density was increased approximately 10-15% by cooling the molds under compression. Further process testing should ensure product densification through one of several pressure molding techniques. Commercial processing equipment/methods should be pursued to this end. Potential improvements in product density were identified by using UO_2 or U_3O_8 powders and/or sintered uranium oxide as an aggregate addition to the microencapsulated powder.

Mean compressive strength was consistently high for all samples: approximately 13.8 MPa (2000 psi) or greater for all samples. Within statistical error, the trend was flat with exception of 90 wt% DUPoly samples which were slightly higher, probably due to particle-to-particle contact of the DU in the matrix. Percent deformation at yield was noticeably different between waste loadings, with 90 wt% DU samples reaching their maximum strength at about 7% deformation, compared to approximately 26% deformation for 50 wt% DUPoly samples. All forms easily surpass the minimum 0.41 MPa (60 psi) compressive strength recommended by NRC for waste form burial.

Leachability and water immersion testing appear to indicate similar trends in that results are sensitive to both waste loading and type of UO_3 processed. 90 wt% batch process DUPoly leaches and degrades significantly faster than comparably loaded continuous process DUPoly or batch process DUPoly with lower waste loadings. In ALT tests, the leach rate for 90 wt% batch process DUPoly samples was approximately 15 times higher than for 50 or 70 wt% samples. Similarly, swelling and cracking of immersion samples was observed for batch process DU samples only at very high (>85 wt%) waste loadings. In contrast, continuous process DU showed little evidence of leaching or swelling/cracking during 90 day immersion testing even at the highest waste loading of 90 wt%. Leaching and swelling/cracking in batch process DU are probably related to the same phenomenon, i.e., presence of soluble compounds, although no effort was made to investigate the chemical differences in the two sources. While it is clear that continuous process DU provides a significantly more stable and durable product, further testing should be done to understand the UO_3 chemistries and leach mechanisms.

Product density improvements are achievable using alternative DU materials and/or processes enhancements. Uranium oxide crystal and bulk powder densities are the limiting parameters in achieving maximum product density and shielding performance. For example, a maximum product density of 6.1 g/cm^3 was estimated using UO_2 powder as opposed to UO_3 powder. Additional product density improvements up to about 7.2 g/cm^3 were estimated using UO_2 in a hybrid technique known as micro/macroencapsulation. The micro/macro DU processing alternative has the potential for incorporating the greatest volume of DU compared to all other alternatives.

Potential commercial applications for DUPoly products were identified including radiation shielding, counterweights/ballast (for use in airplanes, helicopters, ships and missiles), flywheels, armor, and projectiles. Since DUPoly is an effective shielding material for both gamma and neutron radiation it has application for shielding high activity waste (e.g., ion exchange resins, glass gems) spent fuel dry storage casks, and high energy experimental facilities (e.g., accelerator targets) to reduce radiation exposures to workers and the public. Continued development efforts for DUPoly are needed in order to:

- optimize formulations to achieve maximum loadings and product densities projected in feasibility studies without sacrificing performance objectives (e.g., strength, durability, leachability);
- explore processing options for production of secondary use DUPoly products (e.g., engineering design and production techniques for spent fuel storage casks;
- confirm long-term durability of DUPoly by investigating potential degradation mechanisms (e.g., oxidation, radiation stability);
- identify industrial partner(s) and complete technology transfer for the production of commercially viable DUPoly products.

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