Prepare a Conceptual Design of the Process and Equipment that Would Be Required to Solidify UNF Powder in a Hot Cell

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SUMMARY

The 2018 goals of the Volatilization project are to provide a path forward for the disposal of the powders that will be generated during the future hot cell demonstration of the pretreatment process. For this purpose, cermet and grout waste forms were made using surrogate samples.

The surrogate used nuclear fuel (UNF) powder was prepared by ammonia precipitation of a solution containing 300 g of uranium as uranyl nitrate, 1.249 g of $CsNO_3$, 0.162 g of CsI, 0.721 g of $Sr(NO_3)_2$, 1.213 g of $Ce(NO_3)_3.6H_2O$, and 2.047 g of $La(NO_3)_3.6H_2O$. The ammonium diuranate precipitate was dried and calcined to produce a solid solution of simulated fission products in a UO_3 matrix.

Five green pellets were prepared at a mixing ratio of 40 vol % UO_3 surrogate and 60 vol % Cu where the volumes are calculated based on the crystal densities. The mixture was pressed at 20,000 PSI and sintered for 6 h at a temperature of 900°C, which resulted in very good cermet waste forms. The density of the five pellets ranged from 96.6% to 98.9% of the theoretical, and microscopic analysis showed a very good microstructure with a continuous copper phase encapsulating the UNF surrogate waste particles.

To prepare the grout waste forms, a formulation with the following ingredients was used: 20 wt % Type II portland cement, 40 wt % blast furnace slag, 35 wt % class F fly ash, and 5 wt % bentonite. To make the grout, we mixed 400 g of the cementitious blend with 120 g of water and 3.9 g of plasticizer (Glenium) plus 0, 5, or 10 wt % (0, 27.6, or 55.1 g) of UNF surrogate. After 28 days of curing, three grout specimens for each loading (0, 5, and 10 wt %) were leached using the American National Standards Institute/American Nuclear Society 16.1 methodology. The leachability test showed satisfactory retention of all of the simulant species.

Prior experience associated with the disposal of nuclear fuel cycle research and development wastes at the Waste Isolation Pilot Plant (WIPP) indicates that a drum filled with four vertically stacked quart-size cans, each containing approximately 250 g of solidified UNF, will be acceptable.

The exact loading limits will have to be evaluated in detail for the composition of the UNF powder that will be generated, but for planning purposes, a conservative limit of 200 g of UNF per can (800 g per drum) solidified as a grout or cermet waste form was used. Both waste forms appear satisfactory. However, since the equipment needed to make grout is simpler, grout is the main option at this time.

A disposal path has been identified for the samples to be generated during kilogram-scale tests of the advanced pretreatment process, and disposal at WIPP appears achievable and straightforward.

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ACRONYMS

ANS	/American Nuclear Society
ANSI	American National Standards Institute
CANDU	Canada Deuterium Uranium
CERMET	composite material made up of ceramic and metallic materials
LI	leachability index
PSI	pounds per square inch
UNF	used nuclear fuel
WIPP	Waste Isolation Pilot Plant

PREPARE A CONCEPTUAL DESIGN OF THE PROCESS AND EQUIPMENT THAT WOULD BE REQUIRED TO SOLIDIFY UNF POWDER IN A HOT CELL

1. INTRODUCTION

A dry pretreatment process based on the oxidation of used nuclear fuel (UNF) is being studied for the upfront removal and capture of tritium and iodine. Treatment with nitrogen dioxide/oxygen (NO_2/O_2) mixtures converts the fuel into a fine uranium oxide powder, and with further reaction time and temperature adjustment, the oxide can be converted to a uranium nitrate. By adjusting the processing conditions, a U_3O_8, UO_3, or nitrate powder product can be selected. In the nitrate form, uranium may be extracted from the dry powder directly into an organic tri-*n*-butyl phosphate liquid phase. This would eliminate the nitric acid (HNO_3) leaching step and could eliminate the high-acidity raffinate that is normally generated in aqueous fuel-processing systems. All the process tenets and steps were corroborated and tested at the kilogram scale using surrogate materials and at the gram scale using authentic commercial UNF. The next planned development step is to test the entire process at the kilogram scale using UNF.

From the initial review of the planning documents it was determined that the powders to be generated during the testing of the process had no disposal path unless they were converted to a monolithic waste form. Consequently, to provide a path forward for the disposal of the powders that will be generated during the future hot cell demonstration, two potential waste forms, a ceramic-metal (cermet) composite and a grout, were tested using surrogate UNF powders.

2. CERMET WASTE FORMS

In the so-called “cermet” waste form, individual waste particles are homogeneously dispersed and fully encapsulated by a metal matrix material to produce the cermet composite waste form, except at the surface. The metal matrix provides containment while maintaining a very high thermal and electrical conductivity typical of metals.[1]

The production of cermet waste “pucks” to immobilize a waste stream containing uranium or thorium oxide as the primary constituent is described in previous patents.[2–3] Cermet waste forms display several good attributes, including high thermal conductivity (such that the centerline temperature of the waste form is decreased), increased waste-form density, and high overall waste loadings, typically 40 to 60 vol %.

Several metal alloys can be used to produce cermets. We selected copper because it displays some distinct advantages for this application. Copper is an excellent material for long-term storage or geologic disposition because it is highly durable under reducing conditions.[4] Copper has been used internationally in radioactive waste storage as an exterior canister liner because of its corrosion resistance and chemical stability. For example, it is used at the Onkalo geological repository in Finland[5] (see Figure 1) and for coating steel containers encasing CANDU (Canada Deuterium Uranium) reactor fuel for disposal in the Canadian Precambrian Shield.[6]

Figure 1. Copper exterior canister liner (*left*) and canister (*right*) for the Onkalo geological repository in Finland.

Copper is highly ductile and malleable, allowing for a very easy and homogeneous compression and sintering into a solid form of near theoretical density at temperatures in the 600°C to 900°C range, well below its melting point (1,085°C). Waste forms can be made using very simple processing techniques, such as cold pressing and sintering or hot isostatic pressing. A Cu liner can be added to create a fully encapsulated and contamination-free waste form.

As previously reported,[7] green pellets were prepared at a mixing ratio of 40 vol % surrogate waste and 60 vol % Cu. The mixture was pressed at 20,000 PSI and sintered for 6 h at temperature of 900°C under argon, which resulted in very good cermet waste forms. A more detailed optimization process may indicate that higher waste loadings, lower pressure and temperatures, or a combination of these conditions will also produce acceptable waste forms. However, because of the limited scope of the work and the fact that a good product could be made under easily achievable parameters, no further optimization was attempted.

3. EQUIPMENT REQUIRED TO PRODUCE CERMET WASTE-FORMS

Three major components are required to produce cermet waste forms. All three are commercial, off-the shelf items, and, depending on the facility, may already be available for use.

1) A hydraulic press (Figure 2).

   A typical benchtop laboratory-scale hydraulic press can be readily purchased and can be used without modifications. It has a minimum footprint of approximately 12 × 16 in. and a height of approximately 25 in. The press is manually operated and exerts a clamping force between two plates. The maximum clamping force ranges from12 to 30 tonnes, and the maximum working distance ranges between 6 and 12 in.

2) A set of dies (Figure 3).

   Dies are available in many diameters and heights. The principle of operation to form UNF pellets is rather simple; a mixture of waste powder and copper is poured into the cavity of the die and is compacted by the piston of the hydraulic press. A 1 in. diameter die set was used for our previously reported surrogate tests[7] because they are easy to handle and not too heavy.

3) A tube furnace (Figure 4).

   Tube furnaces can be purchased in many sizes and with maximum temperatures typically ranging from 1,100°C to 1,700°C. A small, basic model (e.g., 1,100°C, 1 ft of heated zone) fitted with a 1.5 in. diameter alumina tube is all that is needed.

Figure 2. Laboratory-scale hydraulic press.

Figure 3. Pellet pressing dies.

Figure 4. Laboratory-scale tube furnace.

4. GROUT WASTE FORMS

As previously reported, a surrogate UNF powder was prepared by ammonia precipitation of a solution containing 300 g of U as uranyl nitrate, 1.249 g of $CsNO_3$, 0.162 g of CsI, 0.721 g of $Sr(NO_3)_2$, 1.213 g of $Ce(NO_3)_3.6H_2O$, and 2.047 g of $La(NO_3)_3.6H_2O$. The ammonium diuranate precipitate was dried and calcined to produce a solid solution of simulated fission products in a UO_3 matrix.

Grout waste forms were prepared using a formulation with the following ingredients: 20 wt % Type II portland cement, 40 wt % blast furnace slag, 35 wt % class F fly ash, and 5 wt % bentonite. To make the grout, we mixed 400 g of the cementitious blend with 120 g of water and 3.9 g of plasticizer (Glenium 7920) plus 0, 5, or 10 wt % (0, 27.6, or 55.1 g) of UNF surrogate.

We cast five samples with 5 wt % UNF, five samples with 10 wt % UNF, and five blanks with only grout in plastic containers with an internal diameter of 2.3 cm, a height of 5 cm, and a volume of 21 mL. After 28 days of curing, three specimens for each loading (0, 5 and 10 wt %) were leached with 400 mL of distilled water using the ANSI/ANS 16.1 methodology.[8] The concentrations of Cs, Sr, Ce, La, and U in the leachate solution were measured using inductively coupled plasma mass spectrometry. The concentration levels for Ce, La, and Sr in the leachates from the surrogate grout samples were indistinguishable from the levels in the leachate from the pure grout control samples. Cesium and uranium concentrations were above background in most of the leachate samples, allowing for the determination of a "leachability index." The general criterion is that leachability indices above 6 are considered acceptable for waste disposal.[9] Table 1 displays the Cs and U leachability indices for the six grout specimens (three for 5% loading and three for 10% loading). All the grouted samples displayed a very good retention of leachable species with leachability indices well above the minimum requirement.

As expected, the samples with higher waste loadings display slightly lower leachability indices, and Cs was more leachable than uranium.

Because all specimens displayed very good retention, higher waste loadings may be possible. However, no further optimization was attempted because of the limited scope of the work and because a good product could be easily made.

The details of the leaching tests, analytical results, and calculation of the leachability indices are shown in Appendix 1.

Table 1. Leachability indices from grouted waste-form samples

Specimen	5% loading		10% loading	
	U	Cs	U	Cs
1	19.1	9.6	15.9	8.2
2	16.9	9.5	15.8	8.1
3	16.6	8.8	15.5	8.2
Average	17.5	9.3	15.7	8.1

5. EQUIPMENT REQUIRED TO PRODUCE GROUT WASTE FORMS

Grout waste forms were prepared using a formulation that was specially designed to minimize the amount of water. The resulting paste is relatively hard to stir and mix. Significant torque, which may be beyond the capabilities for manual stirring, particularly using hot cell manipulators, is required. A commercial stand mixer will suffice for blending grout on a laboratory scale (Figure 5).

Figure 5. Commercial mixer used as a laboratory-scale grout blender.

6. CONCLUSIONS

Both the cermet and grout waste forms are relatively easy to prepare and result in acceptable waste forms. Accordingly, a dual disposal path has been identified for the samples to be generated during kilogram-scale tests of the advanced pretreatment process, and disposal at WIPP appears easily achievable based on recent shipping experience.

The waste loadings for the cermet waste forms are typically higher than the loadings for grout. However, grout has the advantage of requiring less equipment and a smaller footprint. The grouted waste form is more susceptible to the generation of gaseous products because of the radiolysis of the water entrained in the grout. Nevertheless, precedents indicate that grouted samples are acceptable for disposal at WIPP.

Experience with the disposal at WIPP of wastes generated by nuclear fuel cycle research and development activities indicates that a drum filled with four vertically stacked quart-size cans containing approximately 250 g of solidified UNF fuel in each can will be acceptable.

The exact loading limits will have to be evaluated in detail for the composition of the UNF powder that will be generated, but for planning purposes, we are using a limit a conservative of 200 g of UNF per can (800 g per drum) solidified as a grout or cermet waste form. Both waste forms appear satisfactory.

7. REFERENCES

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APPENDIX A

Table A1 shows the initial amounts of U and Cs for the three specimens laden with 5 wt % surrogate waste and the three specimens containing 10w% surrogate waste.

Tables A2, A3, and A4, shows the leachate concentrations for Sr, Cs, La, Ce, and U as determined by inductively coupled plasma mass spectrometry.

Table A2 show the results for the three specimens loaded with 5 wt % of surrogate waste. Table A3 relates to the 10 wt % loading, and Table A4 elates to the three 100% grout specimens and the calculated average concentration for the three control specimens.

Tables A5 and A6 show the concentrations of Sr, Cs, La, Ce, and U after subtraction of the concentrations for the control samples.

The concentration levels for Ce, La, and Sr in the leachates from the surrogate grout samples were indistinguishable from the pure grout control samples, meaning that the leaching of those species is too low to be measured against the background.

Cesium was above background in all of the leachate samples; uranium was above background for all samples except for two. The concentration of uranium for the below-background samples was replaced by the detection limit, which was estimated to be 7.9 10^{-5} µg/L.

The "leachability indices" (LIs) shown in Tables A8 and A9 were calculated using the ANSI/ANS 16.1 methodology according to the equation

LI = log (β/D), where

D is the diffusivity (cm^2/s)

β = 1.0 cm^2/s

The diffusivity D is calculated by the equation

$$D = \pi.[\frac{an}{Ao}/\Delta tn]^2 .[V/S]^2.T$$

where

V is the volume of the grout specimens (20.8 cm^3)

S is the surface area (44.4 cm^2)

*A*o is the initial amount or U or Cs in the grout specimens shown in Table A1 minus the respective amounts of U and Cs leached after 30 s.

Δtn is the time interval

T is the leaching time (i.e., the "mean time" of the leaching interval in seconds), calculated as follows:

$T = \{½[(SQRT(t_n) + SQRT(t_{n-1})]\}^2$

Table A1. Initial amounts of uranium and cesium (g)

Waste loading	Initial uranium mass (g)			Initial cesium mass (g)		
	Specimen 1	Specimen 2	Specimen3	Specimen 1	Specimen 2	Specimen 3
5 wt %	1.50E+00	1.45E+00	1.50E+00	4.27E-03	4.12E-03	4.27E-03
10 wt %	3.07E+00	3.14E+00	3.12E+00	8.72E-03	8.91E-03	8.85E-03

Table A2. Analytical results for the three grout specimens containing 5 wt % surrogate waste

5w%	30 s	2 h	7 h	1 d	2 d	7 d	14 d	28 d
Element	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL
Specimen 1								
Sr	2.18E-03	6.21E-02	1.71E-03	1.56E-01	2.30E-03	7.31E-03	1.89E-02	2.75E-02
Cs	5.39E-03	1.04E-01	1.16E-02	2.10E-01	1.47E-02	4.19E-02	7.41E-02	1.01E-01
La	1.88E-04	1.60E-04	3.80E-05	1.07E-04	3.80E-05	3.80E-05	3.80E-05	3.80E-05
Ce	3.37E-04	4.16E-03	3.52E-05	6.72E-03	3.52E-05	3.52E-05	3.52E-05	3.52E-05
U	5.45E-02	3.62E-02	4.46E-04	1.80E-02	1.95E-04	3.30E-04	1.31E-03	1.39E-03
Specimen 2								
Sr	7.55E-04	7.16E-02	1.15E-03	1.70E-01	8.77E-03	2.53E-01	1.83E-01	1.16E-03
Cs	6.69E-04	1.06E-01	9.81E-03	1.96E-01	3.20E-02	3.46E-01	1.99E-01	3.59E-03
La	3.80E-05	8.50E-05	3.80E-05	1.27E-04	3.80E-05	8.70E-05	3.80E-05	1.37E-04
Ce	3.52E-05	2.53E-04	3.52E-05	5.13E-04	3.52E-05	2.61E-04	1.55E-04	1.81E-04
U	3.16E-04	6.32E-03	1.16E-04	2.20E-02	6.32E-04	1.45E-02	1.54E-02	3.52E-02
Specimen 3								
Sr	1.89E-01	4.57E-02	9.41E-02	1.29E-01	5.71E-03	1.62E-01	2.55E-01	3.17E-01
Cs	1.98E-01	6.70E-02	1.00E-01	1.56E-01	1.59E-02	2.54E-01	2.86E-01	3.10E-01
La	3.90E-05	5.80E-05	1.09E-04	9.60E-05	3.80E-05	6.70E-05	8.10E-05	5.80E-05
Ce	1.18E-04	1.64E-04	2.32E-04	3.30E-04	3.52E-05	1.06E-04	8.11E-04	1.71E-04
U	2.70E-02	9.00E-03	7.65E-03	1.11E-02	4.46E-04	8.64E-03	1.51E-02	2.44E-02

Table A3. Analytical results for the three grout specimens containing 10 wt % surrogate waste

10w%	30 s	2 h	7 h	1 d	2 d	7 d	14 d	28 d
Element	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL
Specimen 1								
Sr	1.39E-03	6.04E-02	9.13E-02	1.85E-01	1.50E-01	3.11E-01	3.32E-01	3.58E-01
Cs	1.35E-02	2.44E-01	2.73E-01	6.03E-01	4.98E-01	1.16E+00	8.13E-01	7.23E-01
La	1.21E-04	1.09E-04	1.03E-04	1.52E-04	8.70E-05	1.09E-04	7.40E-05	7.30E-05
Ce	3.13E-04	2.90E-03	2.78E-03	6.30E-04	2.08E-04	5.07E-04	2.39E-04	2.73E-04
U	1.31E-02	1.32E-02	1.29E-02	4.58E-02	2.62E-02	3.51E-02	4.21E-02	5.06E-02
Specimen 2								
Sr	1.70E-03	6.46E-02	7.10E-02	1.63E-01	1.75E-01	3.85E-01	3.19E-01	3.29E-01
Cs	1.23E-02	2.89E-01	2.43E-01	6.09E-01	6.31E-01	1.44E+00	9.61E-01	9.25E-01
La	2.26E-04	1.81E-04	9.00E-05	1.16E-04	9.50E-05	1.07E-04	9.00E-05	7.40E-05
Ce	1.10E-03	4.69E-04	3.45E-03	3.26E-04	2.38E-03	1.71E-03	2.57E-04	1.33E-04
U	4.80E-02	3.28E-02	1.33E-02	2.86E-02	3.10E-02	4.53E-02	4.88E-02	4.01E-02
Specimen 3								
Sr	1.49E-03	4.94E-02	8.20E-02	1.59E-01	1.43E-01	3.78E-01	3.16E-01	3.53E-01
Cs	1.16E-02	2.08E-01	2.59E-01	5.45E-01	5.13E-01	1.45E+00	8.71E-01	9.33E-01
La	1.07E-04	1.03E-04	1.19E-04	1.50E-04	9.90E-05	2.57E-04	9.50E-05	8.50E-05
Ce	5.64E-04	2.65E-04	2.16E-04	2.57E-04	2.37E-04	1.94E-03	6.41E-04	4.69E-04
U	2.94E-02	5.11E-02	1.51E-02	5.90E-02	3.56E-02	5.62E-02	4.63E-02	6.52E-02

Table A4. Analytical results for the three blank grout specimens containing no surrogate waste

Blank	30 s	2 h	7 h	1 d	2 d	7 d	14 d	28 d
Element	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL	µ/mL
Control 1								
Sr	1.76E-03	7.04E-02	8.01E-02	1.43E-01	1.19E-01	2.58E-01	2.31E-01	2.47E-01
Cs	3.68E-05	3.68E-05	3.68E-05	5.43E-04	8.60E-05	2.43E-04	2.75E-04	3.89E-04
La	1.03E-04	1.18E-04	1.13E-04	1.29E-04	9.10E-05	9.50E-05	1.04E-04	6.20E-05
Ce	1.19E-03	7.98E-04	3.72E-03	3.75E-04	3.53E-04	5.01E-04	8.18E-04	2.08E-04
U	1.16E-03	1.75E-03	5.21E-04	7.85E-04	1.35E-04	2.72E-04	8.21E-04	8.30E-05
Control 2								
Sr	2.17E-03	8.14E-02	9.97E-02	1.71E-01	1.62E-01	3.23E-01	2.62E-01	2.78E-01
Cs	3.68E-05	3.68E-05	3.90E-05	8.30E-05	1.22E-04	3.16E-04	3.91E-04	5.30E-04
La	1.08E-04	9.90E-05	1.01E-04	9.30E-05	8.40E-05	1.04E-04	7.90E-05	7.60E-05
Ce	8.09E-04	3.54E-03	1.08E-03	4.05E-04	2.52E-04	1.64E-03	6.67E-04	8.88E-04
U	1.81E-03	1.40E-03	8.38E-04	7.61E-04	2.02E-04	4.15E-04	1.88E-03	1.62E-02
Control 3								
Sr	1.49E-03	4.94E-02	8.20E-02	1.59E-01	1.43E-01	3.78E-01	3.16E-01	3.53E-01
Cs	1.16E-02	2.08E-01	2.59E-01	5.45E-01	5.13E-01	1.45E+00	8.71E-01	9.33E-01
La	1.07E-04	1.03E-04	1.19E-04	1.50E-04	9.90E-05	2.57E-04	9.50E-05	8.50E-05
Ce	5.64E-04	2.65E-04	2.16E-04	2.57E-04	2.37E-04	1.94E-03	6.41E-04	4.69E-04
U	2.94E-02	5.11E-02	1.51E-02	5.90E-02	3.56E-02	5.62E-02	4.63E-02	6.52E-02
Average concentrations from the 3 control samples								
Sr	2.17E-03	7.50E-02	9.07E-02	1.46E-01	1.35E-01	2.80E-01	2.45E-01	2.60E-01
Cs	3.68E-05	3.68E-05	3.75E-05	2.27E-04	9.77E-05	2.55E-04	3.15E-04	4.61E-04
La	1.07E-04	1.12E-04	1.01E-04	9.73E-05	9.13E-05	1.39E-04	9.73E-05	7.33E-05
Ce	3.14E-03	1.60E-03	1.75E-03	5.36E-04	3.22E-04	8.45E-04	9.02E-04	4.61E-04
U	1.21E-03	1.24E-03	5.65E-04	1.33E-03	1.82E-04	2.29E-03	2.07E-03	5.71E-03

Table A5. Analytical results for the three grout specimens containing 5 wt % surrogate waste
Average control subtracted

5we%	30 s	2 h	7 h	1 d	2 d	7 d	14 d	28 d
Element	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL
Specimen 1 minus average control								
Sr	1.33E-05	-1.29E-02	-8.90E-02	9.67E-03	-1.33E-01	-2.73E-01	-2.26E-01	-2.33E-01
Cs	5.35E-03	1.04E-01	1.16E-02	2.10E-01	1.46E-02	4.16E-02	7.38E-02	1.01E-01
La	8.13E-05	4.80E-05	-6.30E-05	9.67E-06	-5.33E-05	-1.01E-04	-5.93E-05	-3.53E-05
Ce	-2.80E-03	2.56E-03	-1.71E-03	6.18E-03	-2.86E-04	-8.09E-04	-8.66E-04	-4.26E-04
U	5.33E-02	3.50E-02	-1.19E-04	1.67E-02	1.33E-05	-1.96E-03	-7.60E-04	-4.32E-03
Specimen 2 minus average control								
Sr	-1.41E-03	-3.37E-03	-8.96E-02	2.37E-02	-1.27E-01	-2.73E-02	-6.23E-02	-2.59E-01
Cs	6.32E-04	1.06E-01	9.77E-03	1.96E-01	3.19E-02	3.46E-01	1.99E-01	3.13E-03
La	-6.87E-05	-2.70E-05	-6.30E-05	2.97E-05	-5.33E-05	-5.17E-05	-5.93E-05	6.37E-05
Ce	-3.10E-03	-1.34E-03	-1.71E-03	-2.33E-05	-2.86E-04	-5.84E-04	-7.47E-04	-2.80E-04
U	-8.96E-04	5.09E-03	-4.49E-04	2.07E-02	4.50E-04	1.22E-02	1.33E-02	2.95E-02
Specimen 3 minus average control								
Sr	1.87E-01	-2.93E-02	3.40E-03	-1.73E-02	-1.30E-01	-1.18E-01	9.67E-03	5.70E-02
Cs	1.98E-01	6.70E-02	1.00E-01	1.56E-01	1.58E-02	2.54E-01	2.86E-01	3.10E-01
La	-6.77E-05	-5.40E-05	8.00E-06	-1.33E-06	-5.33E-05	-7.17E-05	-1.63E-05	-1.53E-05
Ce	-3.02E-03	-1.43E-03	-1.52E-03	-2.06E-04	-2.86E-04	-7.39E-04	-9.07E-05	-2.90E-04
U	2.58E-02	7.77E-03	7.08E-03	9.77E-03	2.64E-04	6.35E-03	1.30E-02	1.87E-02

Table A6. Analytical results for the three grout specimens containing 10 wt % surrogate waste
Average control subtracted

10w%	30 s	2 h	7 h	1 d	2 d	7 d	14 d	28 d
Element	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL	µg/mL
Specimen 1 minus average control								
Sr	-7.77E-04	-1.46E-02	6.00E-04	3.87E-02	1.47E-02	3.07E-02	8.67E-02	9.80E-02
Cs	1.35E-02	2.44E-01	2.73E-01	6.03E-01	4.98E-01	1.16E+00	8.13E-01	7.23E-01
La	1.43E-05	-3.00E-06	2.00E-06	5.47E-05	-4.33E-06	-2.97E-05	-2.33E-05	-3.33E-07
Ce	-2.83E-03	1.30E-03	1.03E-03	9.37E-05	-1.14E-04	-3.38E-04	-6.63E-04	-1.88E-04
U	1.19E-02	1.20E-02	1.23E-02	4.45E-02	2.60E-02	3.28E-02	4.00E-02	4.49E-02
Specimen 2 minus average control								
Sr	-4.67E-04	-1.04E-02	-1.97E-02	1.67E-02	3.97E-02	1.05E-01	7.37E-02	6.90E-02
Cs	1.23E-02	2.89E-01	2.43E-01	6.09E-01	6.31E-01	1.44E+00	9.61E-01	9.25E-01
La	1.19E-04	6.90E-05	-1.10E-05	1.87E-05	3.67E-06	-3.17E-05	-7.33E-06	6.67E-07
Ce	-2.04E-03	-1.13E-03	1.70E-03	-2.10E-04	2.06E-03	8.65E-04	-6.45E-04	-3.28E-04
U	4.68E-02	3.16E-02	1.27E-02	2.73E-02	3.08E-02	4.30E-02	4.67E-02	3.44E-02
Specimen 3 minus average control								
Sr	-6.77E-04	-2.56E-02	-8.70E-03	1.27E-02	7.67E-03	9.77E-02	7.07E-02	9.30E-02
Cs	1.16E-02	2.08E-01	2.59E-01	5.45E-01	5.13E-01	1.45E+00	8.71E-01	9.33E-01
La	3.33E-07	-9.00E-06	1.80E-05	5.27E-05	7.67E-06	1.18E-04	-2.33E-06	1.17E-05
Ce	-2.58E-03	-1.33E-03	-1.53E-03	-2.79E-04	-8.47E-05	1.10E-03	-2.61E-04	7.67E-06
U	2.82E-02	4.99E-02	1.45E-02	5.77E-02	3.54E-02	5.39E-02	4.42E-02	5.95E-02

Table A7. Leachability index for the leaching of cesium from three grout specimens containing 5 wt % surrogate waste

Mean time (s)	Delta t (s)	Specimen 1		Specimen 2		Specimen 3	
		Diffusivity cm^2/s	Leachability index	Diffusivity cm^2/s	Leachability index	Diffusivity cm^2/s	Leachability index
3.60E+03	7.20E+03	4.52E-09	8.3	5.04E-09	8.3	1.95E-09	8.7
2.97E+04	1.80E+04	7.37E-11	10.1	5.65E-11	10.2	5.72E-09	8.2
1.02E+05	6.12E+04	7.25E-09	8.1	6.78E-09	8.2	4.15E-09	8.4
2.52E+05	8.64E+04	4.33E-11	10.4	2.22E-10	9.7	5.27E-11	10.3
7.12E+05	4.32E+05	3.99E-11	10.4	2.95E-09	8.5	1.54E-09	8.8
1.76E+06	6.05E+05	1.58E-10	9.8	1.23E-09	8.9	2.46E-09	8.6
3.53E+06	1.21E+06	1.47E-10	9.8	1.52E-13	12.8	1.44E-09	8.8
Average leachability index			9.6		9.5		8.8

Table A8. Leachability index for the leaching of cesium from three grout specimens containing 10 wt % surrogate waste

Mean time (s)	Delta t (s)	Specimen 1		Specimen 2		Specimen 3	
		Diffusivity cm^2/s	Leachability index	Diffusivity cm^2/s	Leachability index	Diffusivity cm^2/s	Leachability index
3.60E+03	7.20E+03	5.97E-09	8.2	8.03E-09	8.1	4.22E-09	8.4
2.97E+04	1.80E+04	9.86E-09	8.0	7.49E-09	8.1	8.63E-09	8.1
1.02E+05	6.12E+04	1.44E-08	7.8	1.40E-08	7.9	1.14E-08	7.9
2.52E+05	8.64E+04	1.21E-08	7.9	1.86E-08	7.7	1.25E-08	7.9
7.12E+05	4.32E+05	7.42E-09	8.1	1.10E-08	8.0	1.13E-08	7.9
1.76E+06	6.05E+05	4.60E-09	8.3	6.16E-09	8.2	5.13E-09	8.3
3.53E+06	1.21E+06	1.82E-09	8.7	2.85E-09	8.5	2.94E-09	8.5
Average leachability index			8.2		8.1		8.2

Table A9. Leachability index for the leaching of uranium from three grout specimens containing 5 wt % surrogate waste

Mean time (s)	Delta t (s)	Specimen 1		Specimen 2		Specimen 3	
		Diffusivity cm^2/s	Leachability index	Diffusivity cm^2/s	Leachability index	Diffusivity cm^2/s	Leachability index
3.60E+03	7.20E+03	4.12E-15	14.4	9.35E-17	16.0	2.03E-16	15.7
2.97E+04	1.80E+04	2.79E-20	19.6	2.99E-20	19.5	2.23E-16	15.7
1.02E+05	6.12E+04	3.69E-16	15.4	6.09E-16	15.2	1.27E-16	15.9
2.52E+05	8.64E+04	2.91E-22	21.5	3.56E-19	18.4	1.14E-19	18.9
7.12E+05	4.32E+05	1.16E-21	20.9	2.96E-17	16.5	7.48E-18	17.1
1.76E+06	6.05E+05	1.47E-21	20.8	4.46E-17	16.4	3.97E-17	16.4
3.53E+06	1.21E+06	7.33E-22	21.1	1.09E-16	16.0	4.09E-17	16.4
Average leachability index			19.1		16.9		16.6

Table A10.Leachability index for the leaching of uranium from three grout specimens containing 10 wt % surrogate waste

Mean time (s)	Delta t (s)	Specimen 1		Specimen 2		Specimen 3	
		Diffusivity cm^2/s	Leachability index	Diffusivity cm^2/s	Leachability index	Diffusivity cm^2/s	Leachability lndex
3.60E+03	7.20E+03	1.16E-16	15.9	7.71E-16	15.1	1.95E-15	14.7
2.97E+04	1.80E+04	1.62E-16	15.8	1.66E-16	15.8	2.19E-16	15.7
1.02E+05	6.12E+04	6.29E-16	15.2	2.27E-16	15.6	1.03E-15	15.0
2.52E+05	8.64E+04	2.66E-16	15.6	3.57E-16	15.4	4.79E-16	15.3
7.12E+05	4.32E+05	4.78E-17	16.3	7.87E-17	16.1	1.25E-16	15.9
1.76E+06	6.05E+05	8.98E-17	16.0	1.17E-16	15.9	1.07E-16	16.0
3.53E+06	1.21E+06	5.65E-17	16.2	3.18E-17	16.5	9.64E-17	16.0
Average Leachability lndex			15.9		15.8		15.5