Thermal Treatment of UK Intermediate and Low Level Radioactive Waste: A Demonstration of the GeoMelt Process Towards Treatment of Sellafield Waste - 10507

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ABSTRACT

Impact Services, Inc., GeoMelt® Division (GeoMelt), has performed Proof of Concept Testing (Trials) using its proprietary In Container Vitrification (ICV)™ technology to demonstrate thermal treatment of Low and Intermediate Level Waste (LLW and ILW) located at the Sellafield Nuclear Site (Sellafield) in the UK. Sellafield Limited (SL) is evaluating the potential of GeoMelt ICV to treat a range of waste forms and provide a viable alternative approach to the Sellafield Site baseline waste treatment technique (i.e., grout encapsulation). Two Trials were performed in early 2009 for SL at the GeoMelt Horn Rapids Test Site (HRTS) in Richland, WA. The two-fold purpose of the Trials was to, 1) demonstrate that thermal treatment is effective and, 2) dispel some of the myths surrounding the process. Two representative SL waste stream simulants were treated in each Trial using a 200 liter GeoMelt ICV system, addressing four SL waste streams. Representative samples of the GeoMelt product, swipes of the hood and off-gas pipe, and a HEPA filter were analyzed. The waste stream simulants were successfully treated and each of the Trial objectives were met, demonstrating the efficacy of the process towards treating the diverse waste streams currently stored at Sellafield.

INTRODUCTION

Thermal treatment processes offer a potential alternative approach to the SL baseline techniques, such as grout encapsulation. GeoMelt contracted with SL to demonstrate its ICV technology as a viable alternative approach to these baseline techniques.

The purpose of the Trials is to gain confidence in the potential of the technology to treat a range of waste forms. These waste forms can be categorized radiologically as primarily ILW, but also as LLW. The physical/chemical forms of these wastes vary from legacy Magnox sludge through to process wastes e.g. zeolites, soils, metals, etc.

Waste to be treated is mixed with glass formers and other additives (as needed) and is processed in a refractory-lined metal container using a proprietary GeoMelt joule-heating treatment process. The waste mixture is heated to temperatures ranging from 1000 to 1800°C (depending on the waste stream) effectively destroying organics, decomposing salts, and melting and homogenizing the remaining materials. Upon cooling, a vitreous form is produced that immobilizes radioactive and other elementally hazardous constituents that remain.

Off-gases generated during processing are contained by enclosing the process in a sealed container and off-gas hood that is maintained at below-atmospheric pressure. These off-gases are then conveyed through a treatment system that traps and removes particulates and scrubs condensable gases (e.g., water vapor) and non-condensable gases (e.g., NOx) from the flow stream. The treated off-gas emissions are then safely released to the atmosphere. Secondary waste (arisings) captured by the off-gas treatment system can either be recycled to subsequent GeoMelt treatments or disposed of separately.

PROJECT OVERVIEW

Testing was performed at the GeoMelt, HRTS, located just south of the US Department of Energy's Hanford Site, in Richland, Washington, USA. The first Trial was performed in late February 2009, and the second in early March, 2009.

Trial 1

Trial 1 incorporated the GeoMelt 'Hybrid' treatment process, which treats waste in two stages. The first stage treats waste after it has been placed into an ICV melt vessel. Treatment is initiated at the base of the staged material and progresses upward. With this "Bottom-up" melting process, a 'starter path' (melt initiator) is arranged just above the base cast refractory block (CRB) surface, with four electrodes staged within the container in contact with the starter path. An initial pile of the glass former/waste material is placed on top of the starter path. Power is then applied to the electrodes, gradually melting the starter-path material via joule-heating. This heat propagates upward and outward, eventually melting the surrounding glass former/waste mixture, establishing a more electrically conductive molten mass. The second stage, referred to as Feed-while-melt (FWM), involves feeding additional waste material into the ICV container with power continually applied to the melter. Waste material continues to be added and melted until the container is filled to a predetermined height. Because it continually compensates for the volume reduction inherent in a melting process, the FWM technique maximizes the mass and volume of material processed in the ICV container and produces a full container of vitrified product. A graphic of the process is shown in Fig. 1.

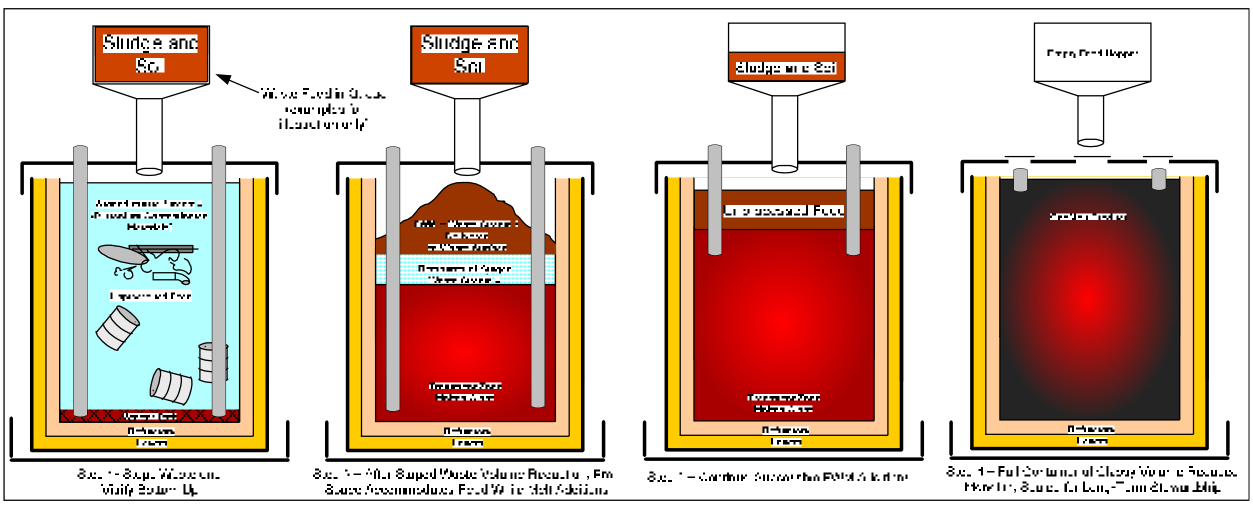

Fig. 1. GeoMelt "Hybrid" ICV Approach

For this Trial, simulated plutonium contaminated materials (PCM), and simulated SIXEP Magnox Sludge, as listed in Table 1, were treated.

Table 1. Trial 1 Formulation

<table>
<tr><th rowspan="2"></th><th colspan="3">Staged - PCM Simulant</th><th colspan="3">FWM - SIXEP Magnox Sludge Simulant</th></tr>
<tr><th>Simulant Component</th><th>Mass of Feed (kg)</th><th>% of Total</th><th>Simulant Component</th><th>Mass of Feed (kg)</th><th>% of Total</th></tr>
<tr><td rowspan="8">Waste Stream Simulants</td><td>Carbon Steel Drum Fragments (representing 200 liter drums)</td><td>78.9</td><td>23.62</td><td>De-mineralized Water</td><td>63.96</td><td>15.05</td></tr>
<tr><td>Stainless Steel (representing 500 liter drums)</td><td>7.1</td><td>2.13</td><td>Misch Metal (Ce, La, Nd, Pr Mix)</td><td>2.93</td><td>0.69</td></tr>
<tr><td>Misch Metal (Ce, La, Nd, Pr Mix)</td><td>0.49</td><td>0.15</td><td>Brucite $Mg(OH)_2$</td><td>82.26</td><td>19.36</td></tr>
<tr><td>PVC as gloves</td><td>24.1</td><td>7.22</td><td rowspan="5"></td><td rowspan="5"></td><td rowspan="5"></td></tr>
<tr><td>Rubber</td><td>9.7</td><td>2.90</td></tr>
<tr><td>Polyethylene as polyethylene grocery bags</td><td>5.4</td><td>1.62</td></tr>
<tr><td>Portland Cement</td><td>18.7</td><td>5.60</td></tr>
<tr><td>Cellulose, Bottle glass, Concrete</td><td>27</td><td>8.08</td></tr>
<tr><td>Other Additives</td><td>Local soil and fluxant</td><td>163</td><td>48.80</td><td>Local soil</td><td>276.25</td><td>65.00</td></tr>
<tr><td colspan="2">Total</td><td>334</td><td>100</td><td></td><td>425</td><td>100</td></tr>
</table>

The steel, PVC, rubber, cellulose, and polyethylene was partially packaged into 48 small carbon steel (tin coated) cans and compacted. The compacted cans were then placed into six inch long thin-wall four inch OD stainless steel tubing and Portland cement was then poured around the compressed cans. These stainless steel tubes with Portland cement-encased cans were placed into the melter at several elevations. They represented scaled versions of 200 liter SL carbon steel waste drums loaded with PCM debris that have been compacted and then grouted inside 500 liter stainless steel over-pack drums.

As shown in Fig. 2, the pre-staged mixture of 334 kg of bulky metallic and organic simulated PCM waste stream simulants and glass formers were first treated. After the staged material had volume reduced by approximately 30%, FWM operations commenced. During the FWM stage, 425 kg of Magnox sludge simulant (35 wt%) and glass formers were treated, ultimately producing a full ICV container of glass product. Fig. 3 shows a typical feed pile of glass formers and Magnox sludge simulant inside the ICV container during FWM operations.

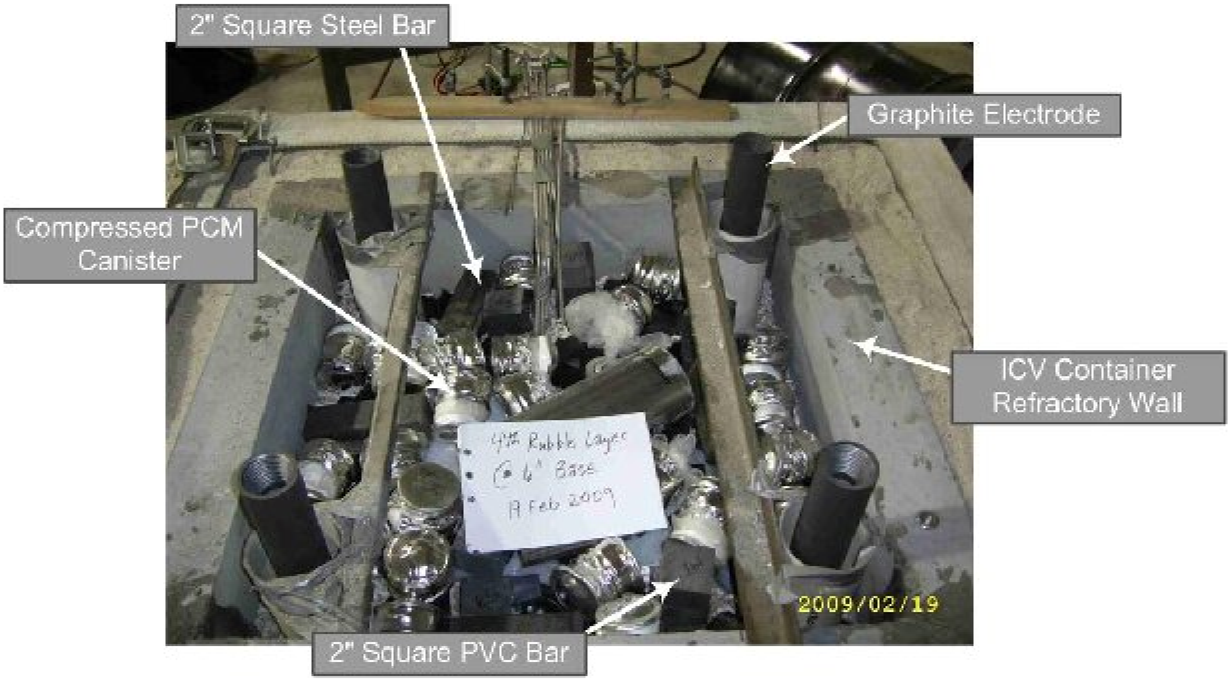

Fig. 2. Sellafield GeoMelt Treatment Trial 1 Staged Feed Material

Fig. 3. Sellafield GeoMelt Treatment Trial 1 FWM Material

A full ICV container of glass product, including glass, weighing approximately 450 kg, and a lower layer of metal primarily composed of iron, weighing 104 kg, were produced. The formation of the separate metal layer was expected, given the high metal fraction (~27%) in the waste simulant.

Non-radioactive isotopes of Ce, Cs, Sr, Co, Re, and Eu were added as tracers to the feed material to simulate the trace chemistry of the radiological species found in the SL waste streams. Representative samples of the glass product, swipes of the hood and off-gas pipe, and a section of the first stage HEPA filter were obtained and analyzed.

Analytical results from these samples were used to determine the overall distribution of the tracers within the system. Finite sample results were extrapolated to represent the entire glass block, off-gas hood and pipe surface area, and the HEPA filter.

Table 2 provides a mass balance summary of the isotope deposition through the treatment system, with each zone presented in weight percentages of the total deposition recovered.

Table 2. Trial 1 Zonal Isotope Wt% Distribution

	GeoMelt Product				Off-gas System			
Simulant Tracer	Glass	Metal Layer	Total		ICV Hood	Piping	HEPA Filter (Available for recycle)	Total
Ce	99.972	0.000	99.972		0.001	0.000	0.028	0.029
Sr	98.946	0.000	98.946		0.003	0.002	1.052	1.057
Cs	68.283	0.000	68.283		0.200	0.101	31.616	31.917
Re	4.540	88.844	93.384		0.161	0.268	6.348	6.777
Eu	99.659	0.000	99.659		0.001	0.001	0.340	0.342
Co	4.743	95.114	99.858		0.032	0.083	0.059	0.175

As seen in the table, approximately 99% of the recovered Ce, Sr and Eu were contained within the glass block, whereas the majority of the Re and Co migrated into the metal layer at the base of the glass block. If the fractions from these two GeoMelt products are summed, 99% of Co is accounted for and 93% of Re is accounted for as immobilized. The remainder of the Cs is primarily recovered on the HEPA filter. The off-gas system accounts for 1% or less of all isotopic deposition, except for Cs and Re, at 31.9% and 6.8% respectively. However, this HEPA filter, containing the balance of Cs and Re, could be removed in entirety and recycled into a subsequent ICV melt treatment if desired. A portion of the filter from the Trial was in fact removed for inclusion in the 2nd Sellafield Trial.

Glass durability testing (e.g., Product Consistency Test, Toxicity Characteristic Leaching Procedure) was not performed since it was outside the scope of the Trial; however, bulk analyses of samples from different locations within the glass block were analyzed. The results showed that distribution of analytes was consistent, demonstrating chemical homogeneity and suggesting mechanical homogeneity throughout the glass block.

Process data obtained during the Trial, combined with post-melt examination of the GeoMelt product, indicated that all materials were successfully treated, each of the Trial Objectives were met, and that the second Trial (Trial 2) should commence as planned.

Trial 2

For Trial 2, simulated SIXEP sand/clino and simulated reactor fuel cladding silo waste were treated using a batch Top-down GeoMelt approach. Similar to the Trial 1 staged feed treatment, the Top-down batch approach involves first filling the ICV treatment vessel with waste stream simulants and glass formers. A starter path material is then placed on the surface of the top surface followed by placement of the graphite electrodes. After the ICV container is secured and made ready for processing, melt power is initiated. Power continues to be applied until all of the material has been fully treated.

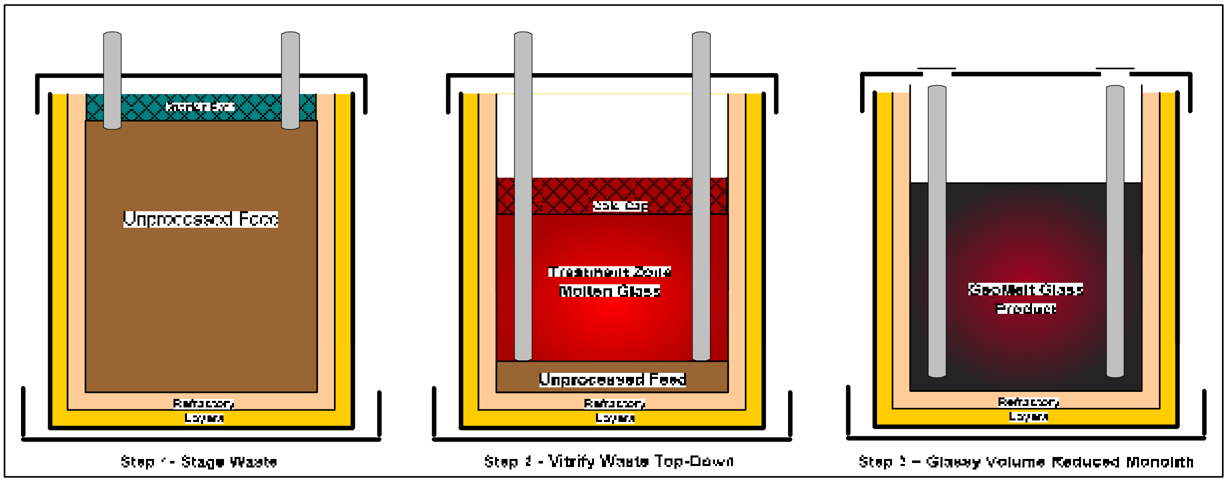

Fig. 4. GeoMelt "Top-down", Staged" ICV Approach

For this Trial, simulated Pile Fuel Cladding Silo waste and simulated SIXEP Sand/Clinoptilolite waste, as listed in Table 3, were treated.

Table 3. Trial 2 Feed Formulation

Pile Fuel Cladding Silo Simulant			SIXEP Sand/Clinoptilolite Simulant		
Simulant Component	Mass of Feed (kg)	% of Total	Simulant Component	Mass of Feed (kg)	% of Total
C - Graphite	13.5	3.31	Demineralized Water	34.1	8.37
CaCO3 - Limestone	5.5	1.35	Clinoptilolite	42.4	10.41
Fe - Steel	14.8	3.63	Silica Sand	10.5	2.58
Mg - metal (rods)	16.0	3.93	Misch Metal (Ce, La, Nd, Pr Mix)	0.8	.20
Mg(OH)2 - Brucite	4.3	1.06	Mg(OH)2 - Brucite	4.0	.98
Al2O3 - Alumina	4.3	1.06	Al2O3 - Alumina	0.1	.02
Cellulose - Trial 1 Recycled HEPA and Scrubfilter	2.2	.54	CaO - Quicklime	0.4	.10
Hydraulic Oil	0.3	.07	Fe2O3 - Hematite	0.1	.02
			Na2CO3 - Sodium Carbonate/Soda Ash	0.2	.05
			K2O - Potash	0.2	.05
Pile Fuel Cladding Silo Simulant Subtotal	61	15	SIXEP Sand/Clinoptilolite Simulant Subtotal	92.2	23
Soil/Hematite Glass Forming Additives				253.55	62
Final Total				407.25	100

The waste stream simulant feed was added to the ICV container in stages. A one inch layer of glass former mixture was first placed on the base silica sand liner surface. A waste stream stimulant of reactor fuel cladding silo waste was then loaded with one inch diameter x six inch long Mg metal rods were placed upright, at varying elevations, as shown in Fig. 5, with the remaining simulant materials then added around and above them, as seen in Fig. 6 (note the carbon steel rods, the recycled HEPA filter material (cellulose), and the used scrub water filter bag from Trial 1).

Fig. 5. Sellafield GeoMelt Trial 2 Staged Mg Rod Waste Stream Simulant

Fig. 6. Sellafield GeoMelt Trial 2 Steel Bar and Recycled Filters from Trial 1 as Waste Stream Simulant

SIXEP Sand/Clinoptilolite waste stream simulant was then added, filling the ICV container, as shown in Fig. 7. This shows the waste stream simulant just prior to placing the starter path and electrodes. Note the liquid pooling of the material in the center of the photo.

Fig. 7. Sellafield GeoMelt Trial 2 SIXEP Sand/Clino Waste Stream Simulant

As with Trial 1, non-radioactive isotopes of Ce, Cs, Sr, Co, Re, and Eu were added as tracers to the feed material to simulate the trace chemistry of the radiological species found in the SL waste streams. Representative samples of the glass product and swipes of the hood and off-gas pipe were obtained and analyzed.

Rhenium was added via the residual metals deposited on the recycled HEPA filter from Trial 1. This relatively small amount (0.1584 grams), when distributed throughout the system, was insufficient to meet the laboratory minimum detection limits (~0.1 mg/kg) for samples obtained from the glass block and metal deposits. However, adequate quantities of material were not available to meet the detection limit 0.2 ug/sample) for the ICV lid swipes, pipe surface and pre-HEPA filter samples. Rhenium distribution results are therefore presented as a range of values, based on typical GeoMelt tracer distribution, including those results from Trial 1.

As seen in Table 4, greater than 99% of the recovered Ce, Sr, Cs, and Eu were contained within the glass block, whereas the majority of the Co (83%) migrated into the metal phase at the base of the glass block. Summing the fractions of these two GeoMelt products, 96% of the Co is retained within the ICV container. This is not unexpected and is similar to Trial 1 results in which most of the Co migrated into the metal phase. The small remainder of the Co entering the off-gas is deposited on the pre-HEPA filter or, to a lesser extent, on the pre-HEPA piping. For each of the tracer isotopes added, 0.13 wt% or less were found past the pre-HEPA filter.

Table 4. Trial 1 Zonal COPC Wt% Distribution

	Product			Off-gas System				
Simulant Tracer	Glass	Metal Phase	Total	ICV Hood	Pre HEPA Piping	Post HEPA Piping	Pre-HEPA Filter (Recyclable)	Total
Ce	99.718	0.000	99.718	0.226	0.016	0.000	0.039	0.282
Sr	99.392	0.000	99.392	0.018	0.065	0.002	0.523	0.608
Cs	99.125	0.000	99.125	0.289	0.109	0.070	0.407	0.875
Eu	99.591	0.000	99.591	0.039	0.150	0.001	0.219	0.409
Re	<11	<92	80-95	<53	<10	<24	<12	5-20
Co	12.999	83.409	96.408	0.534	1.972	0.130	0.956	3.592

The Trial was completed on schedule and fully treated the 407 kg of feed material, producing approximately 281 kg of glass and reduced metal.

CONCLUSION

Both Trials were completed on schedule and fully treated the waste stream simulant staged or fed to the ICV container. Process data obtained during the Trials, analytical laboratory results, combined with post-melt examination of the GeoMelt product, indicates that both of the Trial Objectives were completed. These results demonstrate the efficacy of the process towards treating the diverse waste streams currently stored at Sellafield.

Approximately 99% of the recovered Ce, Sr and Eu were contained within the glass product for both Trials. The Re and Co migrated into a metal phase that developed at the base of the glass block. A 99% retention of Co and 80-93% of Re was obtained in the combined fractions of the glass and metal phase within the ICV container.

Although only four distinct waste forms were addressed during the Trials, these successful results are indicative of the performance obtained during GeoMelt treatment of other wastes forms, including those that will need to be addressed at Sellafield, such as contaminated soils, sludges, sediments, etc.