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Active demonstration of the thermal treatment of surrogate sludge and surrogate drums using the GeoMelt™ In Container Vitrification (ICV) melter installed in NNL Central Laboratory

S Clarke[1], C Scales[1], N Patel[2], J Roe[1] and A W Banford[2]

[1] National Nuclear Laboratory, Central Laboratory, Sellafield Works, Seascale, CA201PG, United Kingdom
[2] National Nuclear Laboratory, 5th Floor Chadwick House, Birchwood Park, Birchwood, Warrington, WA3 6AE, United Kingdom

sean.clarke@uknnl.com

Abstract. Under the auspices of the THERAMIN project, the GeoMelt™ in-container vitrification (ICV) melter system installed in the National Nuclear Laboratory (NNL) Central Laboratory at Sellafield, UK, has been used to demonstrate the immobilisation of two surrogate feeds. The streams selected for demonstration were a cementitious stream representative of legacy drums or failing cemented wastes packages and a wet waste stream made up of a surrogate for Magnox storage pond sludge and clinoptilolite, an ion exchange material, as a way of demonstrating co-immobilisation. The distribution of waste species throughout the vitrified block were assessed to determine the efficacy of the mixing during the thermal treatment process. Analysis of samples from the off-gas system was carried out to establish an activity balance across the process. This paper will outline the process set up, provide operational data from the melt concluding with an assessment of the applicability of the trials results to thermal treatment of these and similar waste streams.

1. Introduction

In the UK, High Level Waste (HLW) and Intermediate Level Waste (ILW) have been successfully immobilised over many decades. HLW has been treated using vitrification and some operational ILW streams by immobilisation in a cement matrix.

Cementitious encapsulation was selected as the baseline treatment technology for many ILW was selected as the baseline treatment technology as it is simple, readily available and is a low-cost material compatible with wet wastes. However, ILW destined for cementation does not generally undergo pre-treatment. In the absence of this stage, organic plutonium contaminated material (PCM) and/or reactive metals could retain chemical reactivity within cementitious products; thus, additional considerations are required for interim storage and final disposal.

Thermal treatment offers an alternative waste treatment process. The application of heat in a thermal process can pacify the waste metals and provide significant volume reduction, this leads to a product that is potentially more stable and more cost effective for storage and disposal.

In the UK, NNL plays a key role in the technical integration and maturation of technologies through the Technology Readiness Levels (TRLs), towards subsequent deployment by end users. The

GeoMeltTM system at the NNL Central Laboratory [1] at Sellafield was commissioned to treat waste and prove the concept that thermal treatment technologies could be chosen as a viable alternative to grout encapsulation for the immobilisation of some ILW streams. Progress made in recent years in this area has included advances with respect to not only the operation of such a system in an active environment, but also advances with respect to the safety case approach required to do so. NNL has published a position paper on thermal treatment of ILW which provides further background [2]. Learning from this experience within NNL formed a key element of NNL's leadership of the thermal treatment demonstration work package (WP3) of the THERAMIN project.

2. In Can Vitrification and GeoMelt™ at NNL Central Laboratory

GeoMelt™ incorporates a range of patented and proprietary vitrification processing technologies that are configured in a variety of ways depending on the waste to be treated and the disposal route (in-situ or containerised). GeoMelt™ transforms hazardous chemical and radioactive wastes into an inert vitreous monolith. Hazardous organics are destroyed by pyrolysis, and heavy metals and radionuclides are immobilized within the glass matrix.

The basis of heating within the GeoMelt™ thermal system is joule-heating via four electrodes. The joule-heating principle operates by internal resistance heating of conductive material as the electrical current passes through. The GeoMelt™ process is initiated at a relatively high resistance requiring a higher voltage potential and lower current. As the melt progresses and resistance decreases, lower voltage taps allow increased current to the melt.

The GeoMelt™ configuration is illustrated in Figure 1. The melter (1) is powered by a 150kW transformer. As the melt progresses, additional material can be fed using the feed while melt system (FWM - 2,3) to take advantage of the space left by volume reduction of the initial waste and reduce voidage within the CRB. The entire system is held under ventilation depression during melting using a blower with a contingency back up (12,13), air is drawn through a filtered inlet line (4) The first stage of the off-gas treatment system is a sintered metal filter (SMF - 5). This is back pulsed during operation to prevent blockage and the associated increase in differential pressure.

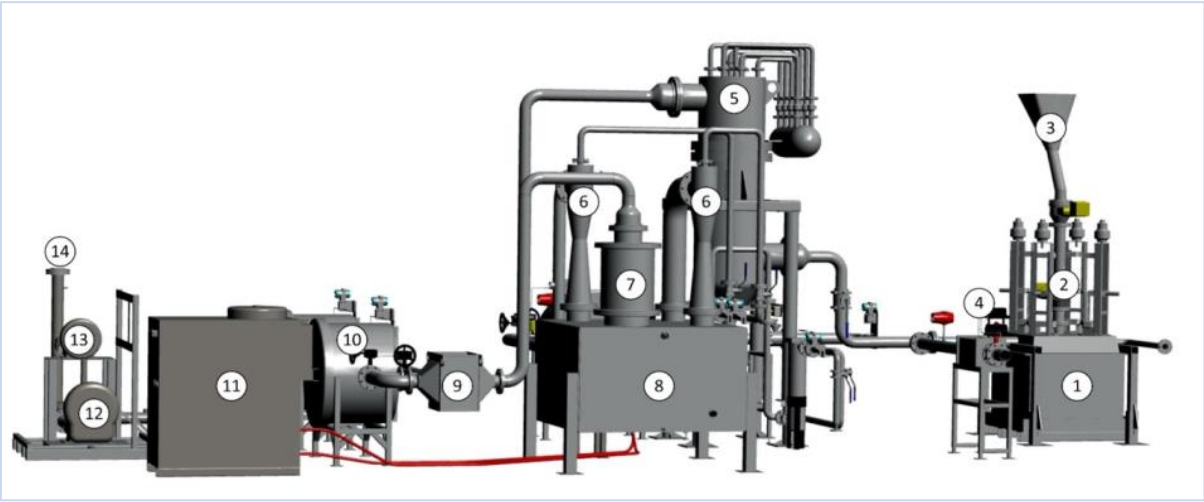

Figure 1. GeoMelt™ ICV System.

Following the SMF, the off-gas stream passes through a cooled caustic scrubbing system (6,7,8,11). The off-gas stream requires heating following scrubbing to avoid moisture dropping out and corroding the pipework, it is heated in line (9). The final stage of filtration is provided by two in line HEPA filters (10) before connecting to the building ventilation network (14). The NNL Central Laboratory GeoMelt™ ICV installation is shown in Figures 2 and 3.

Figure 2. GeoMelt™ ICV at NNL Central Laboratory.

Figure 3. GeoMelt™ ICV installed at NNL Central Laboratory.

3. THERAMIN Demonstration Trials

3.1 Trial Feed Composition

Two specific waste streams were selected for the THERAMIN experimental programme based on the THERAMIN WP2 assessment. Both the wastes tested were identified as being of interest both in the UK and internationally and are referred to subsequently as TH-01 and TH-02.

- TH-01 - A cementitious stream representative of drums or failing cement wastes packages
- TH-02 - A wet waste stream made up of a naturally occurring zeolite (clinoptilolite), sand, Magnox storage pond sludge and miscellaneous contaminants known to arise in a range of UK feed streams.

These are detailed in Table 1.

Table 1. Feed Compositions of GeoMelt™ trials TH-01 and TH-02.

Trial TH-01	Mass (kg) unless stated otherwise	**Trial TH-02**	Mass (kg) unless stated otherwise
Mild steel tins	3.15	CMgS Sludge (45 % v/v solids)	50.25
Cement	13.35	Clinoptilolite	83.8
Aluminium	2.45	Aluminium	1
Plastic (PVC)	3.05	Iron	1
		PAG Oil	1
Glass Formers			
Quarry soil, Boric acid, Soda carb + VTR frit blend	176.6	Silica	8.4
		Sodium Carbonate	16.7
		Boric Acid	8.4
Tracers & Starter Path			
Cs-137 (Active)	25 MBq	Cs-137 (Active)	25 MBq
$SrCO_3$ (Non-active)	(8.42 g)	Sr-85 (Active)	16.1 MBq
CeO_2 (Non-active)	(6.14 g)	EuO_2 (Non-active)	5.8 g
		CeO_2 (added with feed via FWM system)	6.14g
Glass frit (with starter path)	20	Glass frit (with starter path)	20
PRE-STAGE TOTAL	**218.6**		**190.55**
Feed While Melting			
Glass frit	50	Glass frit	12
Soil	10.65		
Total Material	**279.25**		**202.55**

3.2 Operational Observations

3.2.1 TH-01. Melt TH-01 was carried out over a 15 h period. In total 279.25 kg of material was processed at an equivalent rate of 18.6 kg/h. The melt video can be seen here [8].

At several points throughout the melt significant gas evolution was observed at the surface, this was consistent with the decomposition of grout and PVC. This was observed in phases consistent with the melting of each successive layer of tins/cans. At one point, tins were observed to be floating on the surface of the melt, which could have potentially caused a problem of electrical shorting across the electrodes. However, addition of cold feeds dispersed the tins which mitigated this risk. The SMF pressure differential peaked at ~15 inWG with the blower operation peaking at ~80% of its maximum output. Peak power was 34 kW/phase, this was slightly higher than predicted but the higher power was required to reach the target temperature of 1350oC. The melter hood temperature was maintained below its operational control point high of 550oC and the electrical parameters for both phases were well balanced throughout the melt. Operator observation, via the infrared (IR) camera, indicates that adequate convection and movement occurred suggesting good mixing within the melt pool.

3.2.2 TH-02. An initial attempt to melt the feed material was unsuccessful due to the rapid volume reduction of the starter path material which led to a drop of melt surface material away from the electrode tips before they could be lowered. This led to the loss of the conductive path and melt progression. Once cooled the CRB was prepared for re-melt through the introduction of a fresh starter path [3].

The TH-02 re-melt was carried out over 15 h, in total 202.55 kg of material was processed at an equivalent rate of 13.5 kg/h. The melt video can be seen in [9].

At 10.5 h into the melt one of the joule heating phases went into fault. It was not immediately obvious at the time what had happened. The operational decision at this point was to continue processing using the remaining operational phase. Only post-melt was the cause of the fault determined. On retrieval of the product it could be seen that the lowest segment of one of the electrodes had fallen away from the segment above it, breaking the electrical circuit. Due to the loss of one heating phase, no further feeding was undertaken, this was to minimise the challenge on the melter. The power on the remaining phase was increased to 34 kW to compensate for the absence of the second phase, the melt reached its target temperature of 1260oC. The most probable cause of the detachment of the electrode segment is that that the segments were not coupled together robustly during the electrode installation phase of the melt cycle.

4. Results

4.1 Product Appearance

Figures 4 and 5 are pre and post melt images from experiment TH-01, Figures 6 and 7 for experiment TH-02.

Figure 4. TH-01 during loading.

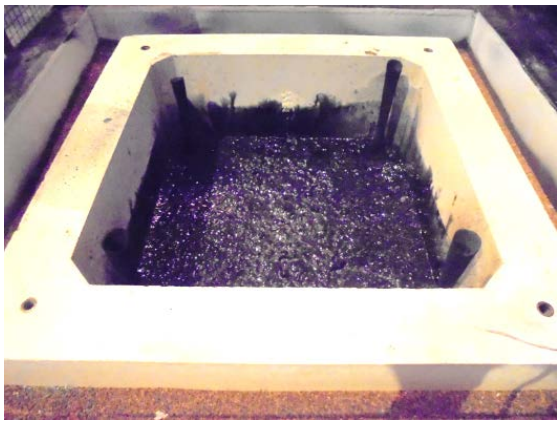

Figure 5. TH-01 vitrified product.

Figure 6. TH-02 starter path.

Figure 7. TH-02 vitrified product.

4.2 Quantities of Product and Secondary Waste

4.2.1 TH-01. Melt TH-01 was cooled in situ and then the ICV and product was removed from plant. On removal of the product, the off-gas pipe connecting to the melter hood was inspected for obstructions, but none were found. Solids were removed from the SMF filter, in total 1.085 kg of material was recovered. In addition, a sample of the scrub tank liquor was taken. A 200 ml sample was taken from a bulk tank volume of 2000 l. SMF solids and scrub tank liquor were submitted for radiochemical and chemical analysis.

The bulk product appeared to have a relatively flat glassy surface, looked broadly homogenous and visually appeared to be a satisfactory product. The electrodes had thinned significantly; this is normal and is observed in virtually all melts carried out with this equipment. The mass of the glass monolith within the CRB was measured to be 236 kg.

The reduction of volume due to the thermal process and the ability to feed additional material during the melt can result in a net overall waste volume reduction for disposal. The potential life cycle benefits of reducing waste volumes for geological disposal during reactor decommissioning was identified by Wallbridge et al [4].

4.2.2 TH-02. As in TH-01, melt TH-02 was cooled in situ and the ICV and product was removed from plant. Solids were removed from the SMF filter, in total 0.16 kg of material was recovered. This amount of material can be considered normal for this process. A sample of the scrub tank liquor was taken. A 200 ml sample was taken from a bulk tank volume of 2000 l. SMF solids and scrub tank liquor were submitted for radiochemical and chemical analysis.

When the product monolith was presented for sampling, the bulk product appeared to have a glassy surface and appeared to be a satisfactory product. As in TH-01, the electrodes had thinned significantly. The mass of the glass monolith within the CRB was measured to be 197 kg. The relatively low product mass resulted in a product container which approximately half full, this is not typical but was due to the inability to feed more than 12 kg during the melt cycle due to the failure of one of the heating phases as described earlier.

4.3 Radioactivity Balance

Glass product and off gas samples taken from melts TH-01 and TH-02 were prepared for Gamma spectroscopy analysis. For each glass sample point a 10g aliquot was finely ground and gamma scanned to determine the Cs-137 and Sr-85 activity concentrations. Similarly, a 10g aliquot of the filtered solids collected in the SMF and 10ml aliquot of the scrub tank liquor were also gamma scanned to determine the Cs-137 and Sr-85 activity concentrations. Table 2 below outlines the results of this analysis. Whilst not available for the TH-01 and TH-02, muon tomography has also been used to provide additional analytical data on GeoMeltTM products from other trials as detailed by Simpson et al [5].

Table 2. Radioactivity analysis across GeoMelt™ System following TH-01 and TH-02.

Melt, Radioisotope	Activity in glass monolith (%)	Activity in SMF solids (%)	Activity in Scrub tank liquor (%)	Difference (%)
TH-01 Cs-137	76.0	1.9	1.2	20.9
TH-02 Cs-137	76.9	0.01	1.2	21.9
TH-02 Sr-85	76.9	>0.01	0.38	22.8

Activity measurements in both TH01 and TH02 trials have yielded Cs retention levels consistent with expectations for a one-off unoptimized melt. It is considered that operational changes along with glass composition modification could significantly increase Cs retention. Sr-85 losses appear high and may be due to errors in the gamma spectroscopy measurements or could indicate entrainment of solids rather than volatility.

Most of the activity in the off-gas system is confined to the SMF, which allows for capture and reprocessing through subsequent melts. Mass and activity analysis across the glass blocks show consistent results indicating that the product is homogeneous, and that waste feeds and additives have been well reacted during the melt process.

The difference in activity identified in Table 2, is probably due to activity retained with the rig system possibly due to a combination of plate out and particulates. This will be investigated in future trials.

5. Conclusions

The active trials carried out by NNL using the GeoMelt™ system installed at the NNL Central Laboratory in Sellafield have successfully demonstrated that thermal treatment can in principle be used to treat surrogate ILW wasteforms. The trials also demonstrate the ability to successfully immobilise both heterogeneous and homogeneous wastes through thermal treatment.

The findings and learning from this proof of concept programme are promising, however it must be stressed that the thermal treatment of wasteforms such as this could be further optimised. The key area of focus would be the optimisation would be changes to the process required to achieve a much higher activity retention % in the final product. If thermal treatment was chosen to treat a given wasteform an in depth and sustained programme of trials would be required to fully understand the feed, optimise the process and maximise the waste loading and volume reduction. Previous experience within NNL in support of the thermal treatment of highly active liquor from spent fuel reprocessing to produce vitrified HLW for Sellafield Ltd. is a useful relevant example of this recommended approach [6].

THERAMIN trials TH-01 and TH-02 using GeoMelt™ has provided an opportunity to demonstrate progression up the Technology Readiness Level (TRL) scale for both feeds from 3 – 6 [7].

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