The DOE Office of Environmental Management International Cooperative Program: Overview of Technical Tasks and Results

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ABSTRACT

The DOE Office of Environmental Management (DOE-EM) Office of Engineering and Technology is responsible for implementing EM's International Cooperative Program. Over the past 15 years, collaborative work has been conducted through this program with researchers in Russia, Ukraine, France, United Kingdom and Republic of Korea. Currently, work is being conducted with researchers in Russia and Ukraine.

Efforts aimed at evaluating and advancing technologies to support U.S. high-level waste (HLW) vitrification initiatives are being conducted in collaboration with Russian researchers. Work at Khlopin Radium Institute (KRI) is targeted at improving the throughput of current vitrification processes by increasing melting rate. These efforts are specifically targeted at challenging waste types identified at the Savannah River Site (SRS) and Hanford Site. The objectives of current efforts at SIA Radon are to gain insight into vitrification process limits for the cold crucible induction melter (CCIM) technology. Previous demonstration testing has shown that the CCIM offers the potential for dramatic increases in waste loading and waste throughput. However, little information is known regarding operational limits that could affect long-term, efficient CCIM operations. Collaborative work with the Russian Electrotechnical University (ETU) "LETI" is aimed at advancing CCIM process monitoring, process control and design. The goal is to further mature the CCIM technology and to establish it as a viable HLW vitrification technology.

The greater than two year effort conducted with the International Radioecology Laboratory in the Ukraine recently completed. The objectives of this study were: to assess the long-term impacts to the environment from radiation exposure in the Chernobyl Exclusion Zone (ChEZ); and to provide information on remediation guidelines and ecological risk assessment within radioactively contaminated territories around the Chernobyl Nuclear Power Plant (ChNPP) based on the results of long-term field monitoring, analytical measurements, and numerical modeling of soils and groundwater radioactive contamination.

INTRODUCTION

The Office of Engineering and Technology's international efforts are aimed at supporting EM's mission of risk reduction and accelerated cleanup of the environmental legacy of the nation's nuclear weapons program and government-sponsored nuclear energy research. To do this, EM pursues collaborations with government organizations, educational institutions, and private industry to identify and develop technologies that can address the cleanup needs of DOE. An important element of these collaborations is to leverage international experience in waste clean-up and environmental remediation. In 2009, through EM's international program, collaborative work was conducted with researchers in Russia and Ukraine.

IMPROVED MELT RATES FOR US HIGH-LEVEL WASTE GLASSES – SRNL, PNNL AND KRI

Background and Application

The U.S. Department of Energy (DOE) is currently processing (or planning to process) high-level waste (HLW) through Joule-heated melters at the Savannah River Site (SRS) and Hanford. The process combines the HLW sludge with a pre-fritted glass or mineral additives which are subsequently melted. The molten glass is poured into stainless steel canisters to produce the final waste form.

Recent tank retrieval and blending strategies at both SRS and Hanford have identified increased amounts of high-Al_2O_3 concentration waste streams that are scheduled to be processed through their respective HLW vitrification facilities. Increased concentrations of Al_2O_3 in glass:

- Promote the crystallization of aluminum-containing crystals during slow cooling in the canister, which can decrease glass durability.
- Increase the liquidus temperature of the glass that can interfere with melter operation.
- Have the potential to decrease feed processing rate due to the refractory nature of Al_2O_3 (i.e. slow dissolution into the glass forming melt).

To address these issues (in particular, improving feed processing rate while avoiding detrimental crystallization), SRNL, PNNL and KRI have jointly performed the following studies:

- Small-scale resistance and joule-heated melter testing to evaluate the impacts of glass frit composition on melt rate for simulated SRS feeds with high aluminum and high iron concentrations
- Small-scale melter testing to evaluate the impacts of melt pool agitation (bubbling) on melt rate for simulated SRS feeds
- Continuous feeding, scaled-melter testing to evaluate the effects of glass composition and addition of an internal heat source on the melting rate of simulated high-aluminum Hanford feeds

Experimental Melters

Two melter systems were utilized at KRI in support of this study, the Steklo Metallicheskie Konstruktsii (SMK) melter system and the Electricheskaya Pech-5 (EP-5) melter system. The SMK is a batch melter intended for experiments on the rate of melting (by monitoring cold cap consumption) and crystal accumulation (through sectioning of the melt crucible) of simulated HLW glass. The components of the SMK melter system make it possible to provide for dry or liquid feeding, off-gas treatment, air bubbling, glass product pouring, and vitrification temperature monitoring, with continuous stirring of the feed. The EP-5 melter is configured for longer-term, continuous feeding and batch pouring. It is equipped with systems and mechanisms that provide for the following: dry or liquid feeding, off-gas treatment, air bubbling of the melt, pouring of the glass product, and temperature monitoring of the melting process.

Figure 1. Demonstration of melter operation at KRI during visit by SRNL and PNNL researchers in May 2009.

Melter Testing with SRS Feeds

KRI prepared the sludge simulants in accordance with recommendations provided by SRNL [1-2]. KRI used two types of simulated sludge for the SRS feeds: one which represented a projected, future sludge batch with a high aluminum concentration (referred to as SMR-2), and one which represented a projected, future sludge batch after the implementation of the aluminum dissolution process (referred to as SMR-4), which resulted in increased iron concentrations [3]. Three frits were evaluated for combination with the simulated SRS sludges to evaluate the impact of frit composition on melt rate, and their compositions are given in Table I.

Table I. Frit Compositions for Melt Rate Studies with SRS Feeds

Frit ID	B_2O_3 (wt %)	CaO (wt %)	Li_2O (wt %)	Na_2O (wt %)	SiO_2 (wt %)
551	8	5	7	8	72
550	12	0	8	7	73
418	8	0	8	8	76

Impact of Frit Composition

The first set of experiments using SRS feeds combined the two simulated sludges with the three frit compositions to determine which frit produced the best melt rate with each of the two feeds. The feeds were melted in the SMK melter. During the experimental runs, KRI performed visual observations of the cold cap behavior on the melt surface, and monitored boiling of the slurry on the cold cap and the morphology of the cold cap during its consumption. The glass was sampled for analysis after the melts were poured from the crucible.

Summarizing the results of the glass analysis after pouring and the visual observations, the following conclusions were made for the SMR-2 feed with the three frits:

1. When the cold cap covered 90% of the melt surface, the sludge simulant feeding rates were the same for Frits 418 and 551 and 11% higher for Frit 550.
2. The time required for consumption of the cold cap when it covered 90% of the melt surface was approximately the same for Frits 551 and 418, and 1 to 1.5 minutes shorter for Frit 550.
3. No spinel inclusions were observed in the glass made with Frit 550.

Similarly, the following conclusions were made for the SMR-4 feed with the three frits:

1. When the cold cap covered 90% of the melt surface, the sludge simulant feeding rates were the same for Frits 551 and 550, and 27% lower for Frit 418.
2. The time required for consumption of the cold cap when it covered 90% of the melt surface was approximately the same for Frits 551 and 418, and 1 to 1.5 minutes shorter Frit 550.
3. No spinel inclusions were observed via SEM in the glass made with Frit 550.

The glass pouring rates upon completion of feeding were approximately the same for Frits 551 and 418, and 30% higher for Frit 550 (likely because the melt with Frit 550 was observed to have a lower viscosity). Based on these results, the Frit 550 composition was selected for follow-on testing to study the impact of agitation on melting of the SMR-2 and SMR-4 simulated sludges.

<u>Impact of Agitation</u>

The next series of experiments was designed to evaluate the impact of bubbling of the melt pool on melt rate in the SMK melter using Frit 550. Visual observations and the feed rates necessary to maintain uniform cold cap coverage were recorded during each melter run. In the experiments without bubbling, the sludge simulant feed spread over the melt surface unevenly and “bumps” formed on the surface of the cold cap with their tops shifting to the back wall of the crucible into the feeding zone. This phenomenon affected measurements of time required for the cold cap consumption because the height of these “bumps” was fairly uncertain. When bubbling was applied to the melt, the sludge simulant feed spread over the melt surface more evenly and no bumps in the feeding area were formed. The visual observations of the pouring process showed that, under similar conditions, the SMR-4 melt with Frit 550 was less viscous than the SMR-2 melt with Frit 550.

All of the experiments with bubbling showed that, as compared to the experiments without bubbling, an increase of the sludge simulant feeding rate was required to maintain the 90% cold cap coverage of the melt surface. Bubbling in the SMR-2 simulant experiments caused a significant increase of the velocity of the cold cap consumption (by a factor of 1.5); while this effect was rather unnoticeable in the SMR-4 experiment.

Melter Testing with Hanford Feeds

A previous study [4] demonstrated that it was possible to formulate glasses with up to 26 wt% Al_2O_3 that satisfied property requirements and were processable with Joule-heated melters operated at 1150° C. This corresponded to a roughly 100% improvement over the current 13 wt

% waste loading limit in Hanford Tank Waste Treatment and Immobilization Plant (WTP) glass. However, the observed melting rate of the high-alumina loaded glass from EP-5 melter test at KRI was roughly half of the WTP design capacity per unit surface area. Further testing was recommended to quantify the effect of high Al_2O_3 loading on glass production rate.

A melting rate study [5] is being performed at PNNL to investigate the impacts of glass and melter feed chemistry on production rate in Hanford HLW melters. One of the potential methods to improve the melting rate identified from that melting study was to use nitrates, as source materials for selected additives, and sucrose to provide an internal heat source. The objective of current EP-5 melter tests at KRI was to investigate the effect of the new glass formulation and addition of nitrates plus sucrose on melting rate of high-alumina loaded glass.

Crucible Glass Formulation Study

A glass formulation crucible study was performed following the same glass formulation strategy and applying the same constraints described in Kim et al. 2008 [4]. The test glasses included a replicate of HAL-17 that was selected for scaled melter testing in a previous study [4] and six additional compositions with 26 wt % Al_2O_3 in glass corresponding to 48.6 wt % waste oxides loading. The focus of the composition variation was the effect of CaO, MgO, Li_2O, Na_2O, and B_2O_3 on predicted melt viscosity and crystallization after canister centerline cooling (CCC) treatment and after 24 h heat treatment (HT) at 950° C. Table II summarizes the glass composition and their test results.

Table II. Target Glass Compositions (in Wt%) and Test Results for High-Al_2O_3 Glasses

Glass ID	**HAL-17R**	**HAL-20**	**HAL-21**	**HAL-22**	**HAL-23**	**HAL-24**	**HAL-25**
Al_2O_3	25.89	25.89	25.89	25.89	25.89	25.89	25.89
B_2O_3	16.14	16.14	16.14	16.14	16.14	20.30	16.14
CaO	7.33	4.25	1.16	4.25	7.33	7.33	7.33
Fe_2O_3	6.37	6.37	6.37	6.37	6.37	6.37	6.37
K_2O	2.72	2.72	2.72	2.72	2.72	2.72	2.72
Li_2O	4.00	4.00	4.00	4.50	4.00	4.00	4.50
MgO	0.13	3.98	7.84	3.98	6.32	0.13	0.13
Na_2O	6.07	6.07	6.07	8.79	6.07	6.07	8.72
SiO_2	26.00	25.23	24.46	22.00	19.81	21.84	22.85
Others(a)	5.36	5.36	5.36	5.36	5.36	5.36	5.36
SUM	100.00	100.00	100.00	100.00	100.00	100.00	100.00
CCC glass, crystal phase(b) vol%	Sp 2.1	Sp 3.2	Sp 3.4	Sp 3.6, Np 5.1	Sp 5.0	Sp 1.9	Sp 1.9, Np 10.2
950°C 24 h HT, crystal phase(b) vol%	Sp 1.1	Sp 2.1	Sp 1.8	Sp 2.2	Sp 2.2	Sp 1.1	Sp 1.3

(a) Others include in wt% BaO 0.06, Bi_2O_3 1.24, CdO 0.03, Cr_2O_3 0.56, F 0.72, NiO 0.43, P_2O_5 1.14, PbO 0.44, SO_3 0.22, TiO_2 0.01, ZnO 0.09, and ZrO_2 0.43.

(b) Sp = spinel, Np = nepheline

Out of the six new formulations, the HAL-24 glass with lowest crystallinity after CCC treatment and 24 HT at 950° C, was selected for scaled melter testing. This HAL-24 had an increased B_2O_3 concentration by 4.2 wt% from HAL-17 at the expense of SiO_2, which resulted in a decrease of predicted viscosity from 4.6 to 2 Pa·s [6] yet showed crystallinity after CCC treatment and 24 HT at 950°C comparable to HAL-17.

HAL-24 glass was tested for viscosity (η) and electrical conductivity (ε) to confirm that the glass met requirements for processing in a Joule heated melter operated at the nominal 1150° C processing temperature. The results showed that HAL-24 had η and ε values within the acceptable ranges. The HAL-24 glass was also tested using the Product Consistency Test (PCT) [7] to confirm product quality compliance. Results showed that boron release from the HAL-24 glass was about one order of magnitude lower than the EA glass requirement [8].

EP-5 Melter Tests

The simulant for this study had the same oxide composition as used in a previous study [4], but source chemicals that better simulated the composition of high-Al Hanford HLW were used. Simulated waste was prepared in 6 batches and preparation of each batch was tested for the wt % of suspended solids to confirm that it was within 19 and 21 wt %.

Three feed variations were tested in this study designed to evaluate the effects of new glass composition and addition of nitrates and sucrose. The sources of alkali oxide additives (K_2O, Li_2O, and Na_2O) were either carbonates (in Feeds 1 and 2) or nitrates (in Feed 3). Table III summarizes the recipe for the three melter feeds tested in this study.

Table III. Recipe for Melter Feeds for Hanford Melting Rate Tests (g per 100 g glass)

Feed	Feed 1 (HAL-17, carbonates)	Feed 2 (HAL-24, carbonates)	Feed 3 (HAL-24, nitrates + sucrose)
Waste simulant oxides[(a)]	48.600	48.600	48.600
H_3BO_3	28.303	35.692	35.692
$CaSiO_3$ (wollastonite)	12.777	12.777	12.777
K_2CO_3	3.771	3.771	NA
KNO_3	NA	NA	5.517
Li_2CO_3	9.436	9.436	NA
$LiNO_3 \cdot 3H_2O$	NA	NA	31.412
$Na_2CO_3 \cdot 2H_2O$	5.041	5.041	NA
$NaNO_3$	NA	NA	6.034
SiO_2	14.103	9.944	9.944
Sucrose (for C/N = 1)	NA	NA	10.867
Additives total	73.431	76.660	112.242
Oxides loading, g per g feed	0.300	0.297	0.268

(a) Mass of waste simulant slurry depends on the amount of water used to prepare the simulant. The amount of simulant used was based on a measured oxides loading of the simulant used for each feed.

Melter operational conditions were kept constant for all tests since the objective of the present study was to evaluate the comparative melting rate of the three feeds. The target glass melt temperature was 1150° C. The EP-5 melter was modified for this study to install two bubblers compared to one bubbler used in the previous study [4]. The bubbling rate was 1.5 L/min from each bubbler with a total bubbling rate of 3 L/min. The steady state feed rate was obtained by establishing the feed rate that maintained the cold cap coverage at between 80 and 90%. The steady state feed rate was used to calculate the steady state melting rate. Table IV summarizes the results of melter tests.

Table IV. Summary of Melter Test Results

Feed	Feed 1 (HAL-17, carbonates)	Feed 2 (HAL-24, carbonates)	Feed 3 (HAL-24, nitrates + sucrose)
Amount of slurry fed, L	14	25	38
Steady state feed rate achieved, L/h	2.5	3.0	4.2
Mass of glass poured, kg	5.5	8.2	13.3
Steady state melting rate, $kg/d/m^2$	680	809	1013

The HAL-17 composition from the present study (Feed 1) achieved significantly higher melting rate (680 $kg/d/m^2$) than that obtained in the previous study (380 $kg/d/m^2$) [4]. The observed increase of melting with the same glass composition was attributed to the changes in melter operational condition and simulated waste composition. It is likely that using two bubblers with increased total bubbling rate (3 L/min) compared to the previous test (0.5 – 0.6 L/min from one bubbler) was the main source for increased melting rate. However the melting rate with HAL-17 (680 $kg/d/m^2$) in this study was still below the WTP design capacity of 800 $kg/d/m^2$.

HAL-24 with carbonates (Feed 2) showed a ~20% increase in melting rate compared to HAL-17 by decreasing the viscosity of glass melt, resulting in a melting rate (809 $kg/d/m^2$) that was comparable to the WTP design capacity. Further increase of melting rate by ~25% (1013 $kg/d/m^2$) was achieved by the HAL-24 with nitrates and sucrose (Feed 3) that provided an internal heat source. The observed increase of melting rate by addition of nitrates and sucrose was encouraging because there was the potential that the heat generated by nitrates and sucrose reactions could be lost during an early stage of melting so that the contribution to the melting rate would be minimal. In addition, the decrease of oxide loading per unit feed mass by the addition of nitrates and sucrose (0.268 g per g feed for Feed 3 compared to 0.297 for Feed 2, see Table III) was expected to contribute to a decrease of melting rate.

In summary, it was shown that the melting rate could be increased by formulating glass with lower viscosity and by providing an internal heat source through raw material additives. This study demonstrated that very high Al_2O_3 loading in glass may be realized without sacrificing the production rate by carefully formulating the glass and implementing various methods designed to improve the melting rate.

MAXIMIZING WASTE LOADING FOR APPLICATION TO SAVANNAH RIVER HIGH LEVEL WASTE – SRNL AND SIA RADON INSTITUTE

Background and Application

CCIM technology is being considered as a possible next generation melter technology for vitrification of DOE wastes. An SRS Sludge Batch 4 (SB4) simulant at a waste loading of 50 wt % (versus about 34 wt % in the current Defense Waste Processing Facility (DWPF) melter) was successfully vitrified using Frit 503-R4 frit additive in a large-scale CCIM [9]. The SB4 formulation represented a high aluminum content feed that, as mentioned above, had presented challenges to HLW vitrification processes. One objective of the work completed in 2009 was to determine a practical limit for processing the SB4 feed in the CCIM. A second objective was to evaluate crystal formation and crystal settling behavior in the CCIM during processing. The formation of crystalline phases in the melt and settling of these phases could deleteriously affect long-term stable melter operations and limit the utility of the CCIM to vitrify high waste loading feeds.

Practical Upper Limit for CCIM Processing of SB4

SB4 waste surrogate and Frit 503-R4 additive mixtures were fed as slurries at 60 wt % and 70 wt % SB4 waste loadings into the SIA Radon bench-scale (0.22 m diameter) cold crucible. The testing was initiated with glass from the previous 50 wt % waste loading tests [9] so a range of waste loadings from 50 to 70 wt % could be evaluated incrementally as 60 and 70 wt % waste loading feed was added to the melter. In total, ~569.1 kg of feed was vitrified and ~186.6 kg of glassy product was produced during about 91 hr of operation. The vitrified product obtained in the bench-scale cold crucible was poured into 12 canisters. The products sampled from canisters 1 to 6 were composed of amorphous phase and spinel with traces of nepheline. The glassy products poured in canisters 7 to 12 were composed of glass, spinel and nepheline. Melter parameters (temperature, power, pour rate, etc.) were monitored during the tests. Glass property measurements including viscosity, electrical resistivity and leach resistance were also made in an attempt to determine an upper waste loading limit for processing.

It was observed that as the waste loading increased it was more difficult to process the waste/glass additive mixtures. An increase in melter temperature was necessary when waste loading was greater that 60 wt % and processing at greater than about 66 wt % waste loading was not feasible due to the refractoriness of the feed. It was further observed that the glass production rates were lower than what was observed at 50 wt % waste loading in the previous study. The leach resistance, as measured by the PCT, indicated that normalized boron release from the glass increased with increasing waste loading. The boron release was significantly higher in a glass with ~65 wt % waste loading where substantial amounts of nepheline formed in the glass. Viscosity and electric resistivity also increased with increasing waste loading. Based on melter observations and glass property measurements, the maximum recommended SB4 waste loading was ~55-60 wt % for the SB4 feed when processing in the SIA Radon CCIM. However, when considering melting rate, a lower waste loading may be optimal to provide maximized waste throughput in the CCIM.

Crystal Formation and Settling Behavior in the CCIM

Prior to implementation of a CCIM in a production facility it is necessary to better understand processing constraints associated with the CCIM. The glass liquidus temperature requirement and tolerance to crystal formation for processing in the CCIM is an open issue. Testing was conducted to evaluate crystal formation and crystal settling during processing in the CCIM to gain insight into the effects on processing. The SB4 waste glass composition with known crystal formation tendencies was selected for testing. A continuous melter test was conducted for approximately 51 hours. To evaluate crystal formation, glass samples were obtained from pours and from glass receipt canisters where the glass melt had varying residence time in the melter. Additionally, upon conclusion of the testing, glass samples from the bottom of the melter were obtained to assess the degree of crystal settling. Glass samples were characterized in an attempt to determine quantitative fractions of crystals in the glass matrix. Crystal identity and relative composition were determined using a combination of x-ray diffraction (XRD) and scanning electron microscopy coupled with energy dispersive spectroscopy (SEM/EDS). Select samples were also analyzed by digesting the glass and determining the composition using inductively coupled atomic emission spectroscopy (ICP-AES).

There was evidence of crystal formation (primarily spinels) in the melt and during cooling of the collected glass. Furthermore, there was evidence of crystal settling in the melt over the duration of the melter campaign. Table V provides the chemical composition of spinel forming oxides in the glass from melter pours that were held at temperature for varying times prior to pouring as well as glass sampled from the bottom of the melter (dead zone) after completion of the testing. It was evident that the concentrations of spinel forming oxides in the glass samples were similar irrespective of melter soak time. However, the glass obtained from the bottom of the melter was enriched in spinel formers indicative of spinel phase settling in the melter. The long-term impact of this crystal settling on melter operation requires further testing.

Table V. Concentration of Spinel Phase Constituents in Glass Samples

Pour	Sample ID	Oxide Concentration (wt %)			
		Fe_2O_3	NiO	Cr_2O_3	MnO
1	30 min	16.8	0.732	0.106	4.16
2	40 min	14.7	0.654	0.092	3.54
3	64 min	16.6	0.702	0.093	3.82
4	92 min	16.2	0.706	0.096	3.78
-	Dead Volume	26.3	2.42	0.492	4.87

COLD CRUCIBLE INDUCTION MELTER MODELING AND CONTROL – INL AND ETU "LETI"

Background and Application

INL is collaborating with ETU "LETI" in research and development efforts related to automated control, mathematical modeling, and innovative draining techniques for application to CCIM systems. These efforts have resulted in design and construction of an integrated large scale CCIM installed at ETU "LETI" that includes a dual frequency glass melting and draining capability (figures 2). The current focus is on improvement of the equipment design and overall control of the glass melting process and continued improvement of the measurement techniques for key operational parameters that are necessary for effective control.

Improved Equipment Design and Melting

A large scale melter with an improved design has been developed and installed in the ETU "LETI" laboratory. The melter has a 0.4 m diameter and is capable of processing 75 kg of glass per melter volume. One key feature of this new melter design is that it includes increased crucible sectioning, which enhances the penetration of the electromagnetic field into the melt. The melter system also includes a sealed lid that offers several advantages, besides being more representative of an actual radioactive waste processing application: 1) it allows reduction of the height of the crucible structure, reducing the electrical losses in the stainless steel tubes, 2) it improves the consistency of the operation, and thus the reliability, and 3) it provides the ability to control the pressure within the melter plenum, thus, allowing control of the glass flow during the draining process, including complete stoppage. The melter design includes two separate inductors, which operate on different frequencies. The primary inductor provides the main energy for initiating and maintaining the melt pool. The secondary inductor provides contactless electromagnetic heating for control of the melt pouring process, while also providing additional heating in the central zone of the melt pool, which enhances the mixing and overall homogeneity of the melt. This is accomplished with no direct contact of materials with the molten product, which is a significant advantage over conventional approaches.

(a) (b) (c)

Figure 2. CCIM system installed at ETU "LETI": a) control and data acquisition systems, b) melter vessel in process hood, c) melter pouring.

This integrated installation also includes a system that pulls a vacuum in the melter plenum for control of the flow rate of glass during draining, with the ability to also fully stop the draining process. This system has been successfully demonstrated during proof of principle testing in smaller crucibles, showing reliability and repeatability (see Figure 2c). Additional testing, enhancement, and refinement are needed due to the increased melter diameter, which results in different characteristics of the temperature distribution profile from the small inductor. Additionally, the high frequency of the smaller inductor requires more effective techniques for isolating the high frequency field from the sensors to provide protection and reliable data acquisition.

Mathematical modeling

Axially-symmetric models of the CCIM have been developed using ANSYS Multiphysics software that provide the capability to conduct "what-if" scenarios, sensitivity analyses, and help focus the experimental work to provide a more efficient use of the resources. The models include both single and dual frequency induction heating systems. Current enhancements have resulted in a representative model for the melt initiation process. All of the models include the electromagnetic, thermal and hydrodynamic effects, as well as the ability to account for temperature-dependence of key material properties. The current models were developed specifically for borosilicate glass processing. The models have been validated to be very representative of the actual melter operation and are, thus, used for optimization of the melt pool geometry, crucible and inductor geometry, and drain geometry, frequency selection, and capacity of the power supply. Extension of the model to investigate a full-scale melter (i.e. 1.0 m diameter) is ongoing with good results to date (see Figure 3).

Additional modeling efforts have been initiated using a different software package that provides the capability to model mass flow, and thus provide a predictive tool for the glass draining process and key parameters for control.

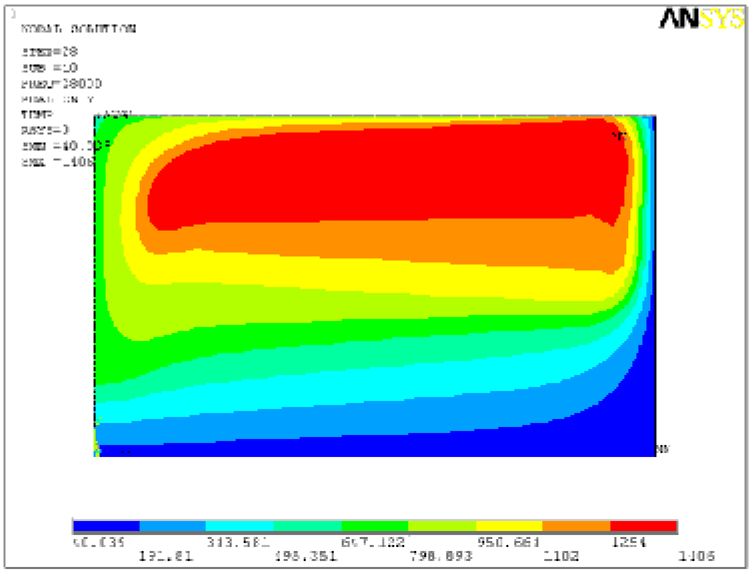

Figure 3. Temperature modeling of the ETU "LETI" CCIM melt pool.

Enhancement of the System for Parameter Measurement and Control

A data acquisition system was developed using various hardware devices and software. This system has been enhanced to include a high precision calorimetry system for all water cooled elements of the installation. These measurements are used to validate the results of the innovative sensors that have been developed for monitoring and control of high frequency induction processing systems. High-precision components have been implemented that result in an overall measurement accuracy of 1.5 %. The data acquisition system also provides for measurement of electrical signals from both generators, high-frequency voltage and current signals of the primary inductor, temperature profile in the melt, current frequency of the generator, pressure in the melter plenum, allowing determination of all of the key electrical characteristics of the CCIM in and automated, real-time, on-line mode (i.e. power factor and efficiency of the inductor, electrical losses, etc.). The system has been significantly enhanced due to improved design of the sensor, improved hardware, and more efficient signal filtration of the high-frequency noise.

Additional efforts are planned to investigate direct measurement of similar signals and characteristics for the secondary high-frequency drain inductor. This will present additional challenges due to the difficulties of isolation of the high frequency signal. Future plans include development of a reliable melt level measurement system, which will allow better control of glass melting draining, offering the possibility to implement a continuous processing capability.

LONG-TERM IMPACTS FROM RADIATION/CONTAMINATION WITHIN THE CHERNOBYL EXCLUSION ZONE – IRL AND SRNL

Background and Application

The following tasks comprised the project: Task 1 **-** Characterization and Monitoring Methods; Task 2 **-** Understanding of the Processes and Modeling of Fate and Transport of Radionuclides through Environmental Pathways (e.g., soils, groundwater, surface waters, microbiota, plants and animals, etc.); Task 3 **-** Evaluation of Risk Assessment Methods; and Task 4 **-** Evaluation and Demonstration of Cleanup Technologies for Radioactively-Contaminated Sites. In turn, these four tasks consisted of 18 subtasks, all of which have been completed. Based on the work described in the subtasks reports, IRL and SRNL collaborators will develop three journal articles, the summaries of which are below. The elements of these tasks provided insight into radioecological conditions in the years following the Chernobyl accident, risk assessment methodologies used and environmental cleanup technologies developed and deployed.

Environmental problems associated with decommissioning the Chernobyl Nuclear Power Plant Cooling Pond

Decommissioning of nuclear power plants and other nuclear fuel cycle facilities is of current interest. There exist significant experience and generally accepted recommendations on remediation of lands with residual radioactive contamination; however, there are hardly any such recommendations on remediation of cooling ponds that, in most cases, are fairly large water reservoirs. The literature only describes remediation of minor reservoirs containing radioactive silt (a complete closure followed by preservation) or small water reservoirs resulting in reestablishing natural water flows. Problems associated with remediation of river reservoirs resulting in flooding of vast agricultural areas also have been described. One of the large, highly contaminated water reservoirs that require either remediation or closure is Karachay Lake near the MAYAK Production Association in the Chelyabinsk Region of Russia where liquid radioactive waste had been deep well injected for a long period of time. Backfilling of Karachay Lake is currently in progress. Another well-known highly contaminated water reservoir is the Chernobyl Nuclear Power Plant (ChNPP) Cooling Pond (Figure 4), decommissioning of which is planned for the near future. This study summarized the environmental problems associated with the ChNPP Cooling Pond decommissioning.

Radioecology of vertebrate animals in the area adjacent to the ChNPP site in 1986-2008

Widespread environmental contamination of the areas adjacent to the ChNPP site attracted a great deal of publicity to the biological consequences of the ChNPP catastrophe. However, only a few studies focused on a detailed analysis of radioactive contamination of the local wild fauna and most of them were published in Eastern European languages, making them poorly accessible for Western scientists. In addition, evaluation of this information was difficult due to significant differences in raw data acquisition and analysis methodologies and final data presentation formats. Most of these studies described fish as an important component of human nutrition found in contaminated water reservoirs in non-evacuated areas and murine rodents as a convenient and traditional element of eco-toxicological research. Data on birds and large mammals were even scarcer while studies of amphibians and reptilians were almost nonexistent. Most studies indicated significant (1-3 orders of magnitude) variations in the radioactive contamination of animals (even for one species inhabiting the same area during the same period of time) due to heterogeneity of radioactive contamination, temporal and spatial variability of biological accessibility of radionuclides, animal migration patterns, variability and specificity of their food patterns, and factors related to gender of animals. Various studies frequently appeared to present contradictory data on ranking species on contamination indicators, seasonal and long-term contamination profiles and sex/age differences. Using an integrated approach for assessment of all available information, the International Radioecology Laboratory scientists showed that the ChNPP accident had increased the average values of the animals ^{137}Cs and ^{90}Sr contamination by a factor of thousands, followed by its decrease by a factor of tens, primarily resulting from a decrease in the biological accessibility of the radionuclides. However, this trend depended on many factors. Plant and bottom feeding fish species (i.e., those that fed from the areas with the maximum adhesion of the radioactive fallout) were the first to reach the maximum contamination levels. No data were available on other vertebrates, but it could be assumed that the same trend was true for all plant feeding animals and animals searching for food on the soil surface. The most significant decrease of the average values occurred during the first 3-5 years after the accident and it was the most pronounced for elk and plant and plankton feeding fish. Their diet included elements “alienated” from the major radionuclide inventory; for example, upper soil layers and bottom deposits where the fallout that had originally precipitated on plants, water and soils gradually migrated. Further radionuclide penetration into deeper layers of soils and its bonding with their mineral components intensified decontamination of the fauna. It took a while for the contamination of predatory fish and mammals (wolves) to reach the maximum followed by its similar slow decrease. Currently, these species are found to be the most contaminated. Species that were environmentally connected with soils or bottom deposits (terrestrial amphibians, rodents, burrowing animals, birds searching for food in soils and underlying layers and wild hogs) also showed relatively high contamination levels. In general, by the mid 1990’s, fluctuations in ^{137}Cs contamination primarily depended on seasonal changes in food patterns, physiology and migration for animal species and on all these factors plus seasonal changes of water temperature and flood patterns for fish species. An increase of ^{90}Sr biological accessibility in soils affected its average accumulation. Currently, due to stabilization of environmental complexes and processes in the region, long-term profiles of radioactive contamination of vertebrate animals mostly indicate a gradual decay of radionuclides, with further changes in biological accessibility of radionuclides being practically unnoticeable due to significant seasonal and geographical fluctuations of the contamination.

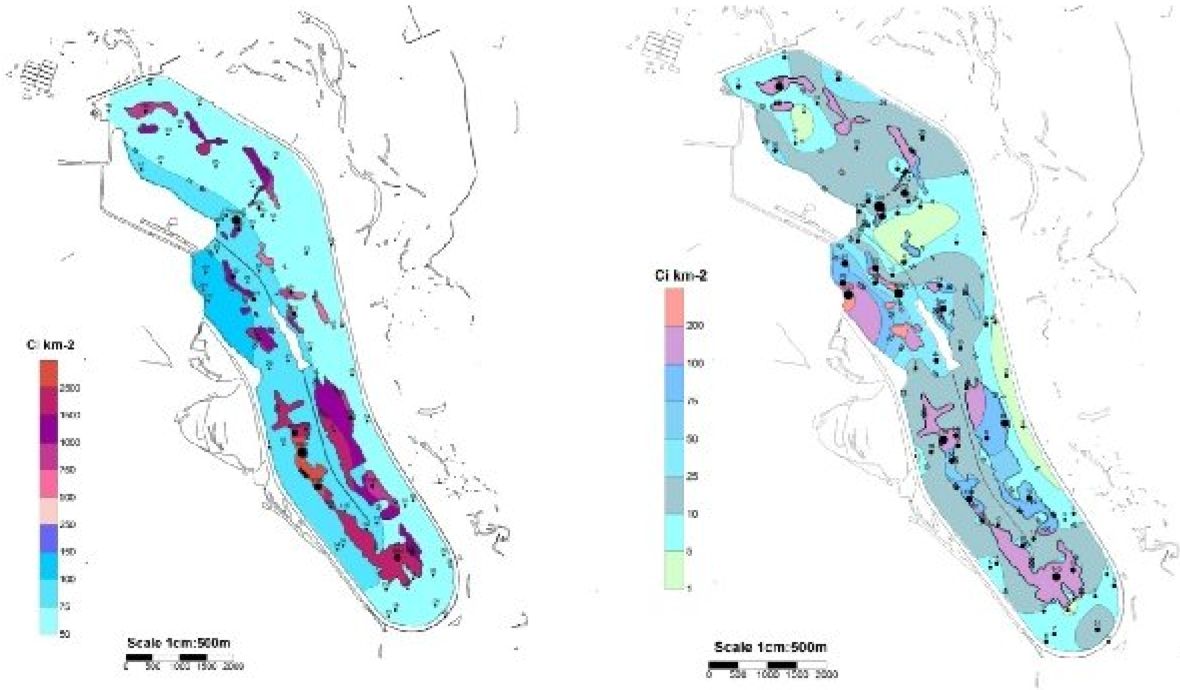

Figure 4. Radioactive contamination distribution for the ChNPP Cooling Pond bottom deposits [10].

SUMMARY

The DOE-EM International Cooperative Program continues to be an important element of the Office of Engineering and Technology's waste cleanup and environmental remediation effort. The international program has provided data to improve HLW vitrification processing through successful melter testing at KRI. The evaluation of the CCIM as a next generation technology for waste vitrification has been facilitated through the joint work with SIA Radon and ETU "LETI". The recently completed collaborations with the IRL provided data on radioecological conditions in areas around the ChNPP and environmental monitoring and risk assessment practices that can be applied to DOE-EM's environmental remediation programs.

The success of these programs provides impetus to continue collaborations with these and other international research partners. Efforts are continuing with researchers in Russia at KRI, SIA Radon and ETU "LETI". The potential for future collaborations with IRL are being investigated through the DOE-EM International Collaborative Program framework as well as evaluating funding opportunities within the DOE-Office of Science or the National Nuclear Security Administration (NNSA).

A project through the DOE-EM International Collaborative Program recently commenced with the Korea Hydro and Nuclear Power - Nuclear Energy and Technology Institute (NETEC). The Ulchin Vitrification Facility (UVF) at the Ulchin Nuclear Power Plant site was designed by NETEC to treat the Low and Intermediate Level Waste (LILW) generated from the Ulchin Power Plant. The CCIM unit recently became radioactively operational. In the current project, NETEC personnel will summarize the process for bringing the unit on-line and provide insight into "lessons learned" during the design, construction and commissioning phases. Dialog is also currently underway with organizations in France, Japan and the United Kingdom regarding collaborative opportunities.

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