A PLASMA MELTING TECHNOLOGY FOR VOLUME REDUCTION OF NONCOMBUSTIBLE RADIOACTIVE WASTE IN KOREA

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ABSTRACT

In Korea, there is a strong need for the development of radioactive waste volume reduction technology. Korea Electric Power Research Institute (KEPRI) has been searching for ways to reduce the radioactive volume significantly and to produce stable waste forms. In particular, plasma treatment technology has caught KEPRI's attention for treating noncombustible radwaste because this technology may far surpass conventional methods. The potential for greater control of temperature, faster reaction times, better control of processing, lower capital costs, greater throughput and more efficient use of energy is there. For the plasma melting study of noncombustible waste, KEPRI has leased a lab scale multipurpose plasma furnace system and performed preliminary tests. Using simulated noncombustible waste based on field survey data from nuclear power plants, lab scale melting experiments have been carried out. The properties of molten slag vary with additives and noncombustible waste materials. KEPRI's current study is focused on finding an optimum composition ratio of various noncombustible wastes for melting, investigating physical properties of molten slag, and obtaining operating parameters for continuous operation.

1. INTRODUCTION

Twelve nuclear power plants, are under commercial operation and four under construction in Korea. As a result of this nuclear program the amount of radioactive waste has been increasing from year to year. Therefore, an advanced volume reduction technology for low- and intermediate-level radioactive wastes (LILW) is greatly needed. The high concern for the protection of the environment, the difficulty in selecting disposal site and the high cost of disposal in Korea further emphasize the need. KEPRI has regarded the vitrification and melting technologies as being the most promising one of the new LILW treatment technologies. An economic and technical feasibility study by KEPRI concluded that the application of vitrification technology utilizing a cold crucible melter (which heats material via direct induction) for the volume reduction and stabilization of combustible LILW such as paper, protective clothing, wood, vinyl sheets, and spent resins would be appropriate. Furthermore, the study concluded that applying plasma melting technology to the volume reduction of noncombustible wastes such as pipe, metal scraps, glass material and concrete would be advantageous. Hence, KEPRI selected an induction heater for the vitrification of combustible waste, and a plasma torch based furnace for the melting of noncombustible waste. The 80 kW plasma torch based furnace is on loan to KEPRI from the Engineering & Technology Research Center of Industrial Wastes for Advanced Materials at Suncheon University, Korea. With this plasma melter, simulated noncombustible wastes such as concrete, sand, glass and metal scraps were melted and converted into slag by high temperature plasma. For the development of plasma melting technology, slag formulation (which varies with additives and waste materials), along with off-gas treatment for the volatile radioisotopes such as cesium, is an important consideration (Pierce & Robert, 1994). The melting experiments are set to be executed through a five phase plan using the 80 kW plasma torch based furnace. The first phase of the experiments has already been performed. In the first phase, additives for the wasted LD slag were used for the control of basicity and ease of melting (Ban & Park, 1996). This paper focuses on the results from KEPRI's recent experiments which have been focused on finding an optimum mixing ratio of various noncombustible wastes with additives and investigating characteristics of produced slag. The purpose of these experiments was to evaluate the possibility of treating noncombustible wastes with the plasma torch melter.

2. PLASMA MELTING TECHNOLOGY

Plasma is an ionized gas that is conditioned to respond to electromagnetic forces. The plasma arc is created when a voltage is established between two points. The plasma acts as a resistive heating element and maintains very high temperatures. The plasma arc creates a flame with high temperatures and energy densities (Camacho, 1988). Uses of this flame, with these characteristics in conjunction with an ionized and reactive medium, have demonstrated the potential of plasma melting technology to eliminate many waste materials in an environmentally safe and cost-effective manner. The plasma torch is a device that converts electrical energy into intense heat. There are two methods of using the plasma torch by a counter-electrode, namely the transferred and the non-transferred method. Recently, plasma vitrification technology has been considered for treating radioactive waste.

3. EXPERIMENTAL FACILITIES

3.1. Plasma Torch

The lab scale convertible torch which can operate either in the transferred mode or in the non-transferred mode was domestically manufactured and installed. This torch has been operated using argon gas and/or hydrogen gas as the plasma gas. The hollow type electrodes of straight polarity were employed. Because the copper electrode generates an arc from its interior, the gas pressure can be varied periodically while the plasma gas moves with a swirling flow. The location of the point of arc generation is moved around to reduce possible wear of the torch (Camacho, 1988). The plasma torch system consists of the following components: the plasma torch assembly; power supply and control panel; water cooling system; and a gas source. As for the plasma power supply, a constant current DC power unit with 60 kW output (maximum current 500A) is used. The cooling water was demineralized water and was introduced into the torch inside, to minimize corrosion of the electrode. Its supply pressure was maintained in the range of $1kg/cm^2$ during torch operation. Before starting the furnace, the system’s temperatures, pressures, cooling water flows, and gas flows are checked and verified.

3.2. Furnace

The furnace has a cylindrical configuration with an inside diameter of 150mm, outside diameter of 340 mm, and a height of 350mm. This furnace is capable of producing slag at approximately 5kg/h. The furnace was constructed of a CA18 type precastable refractory material with a thickness of 95 mm, which is over 93% AI_2O_3. The furnace is floor mounted and is surrounded by a stainless steel plate to reduce heat loss. The furnace has a manual tilting device that can move upwards and downwards in order to pour the molten slag into the casting mold. This furnace was designed for batch operation (Eschenbach & Schlienger et al., 1996). Forced air-cooling was introduced into the furnace when necessary, and the thermocouple was embedded in the bottom of furnace to measure the temperature of melting slag.

3.3. Off-Gas Exhaust System

The off-gas exhaust system is well furnished. This system is composed of an exhaust hood, exhaust fan, dust collector, and duct. During plasma torch operation, the off-gases leave the furnace through the exhaust hood and enter the exhaust duct (500mmx300mm). By the use of an exhaust fan, off-gases are pulled out from the furnace through the exhaust duct into a dust collecting system. Instrumentation for exhaust gas analysis (e.g. the continuous emissions monitoring system) is to be utilized in the near future.

4. EXPERIMENTS

4.1. Contents of Experiments

The contents and scope of lab scale tests using the 80 kW convertible plasma torch are focused on the following:

1. Development of the basic technique to optimize conditions for continuous operation of the plasma torch melting system .
2. Melting simulation of various noncombustible wastes produced at nuclear power plants to study the slag formulation and to determine an optimum mixing ratio of wastes.
3. Intactness tests such as hardness test and leaching tests to analyze the chemical composition of the produced slag and to examine the capture of cesium within the produced slag.
4. Analysis of the off-gas from the plasma furnace to investigate nuclide volatility.

During the experiments, additives for the wasted LD slag were used in the simulated noncombustible wastes to improve viscosity of the molten slag, and to get a non-leaching final products. The transferred mode of the plasma torch in which energy is delivered directly to the material to be heated was used to ensure good slagging efficiency (Camacho, 1988).

4.2. Constituents of Simulated Noncombustible Waste

The simulated wastes consisted of various noncombustible materials such as metal scraps, glass, sand, and concrete. In order to obtain comparative results in each melting experiment and to investigate the optimum mixing ratio of noncombustible materials with respect to waste types, artificial noncombustible LILW material based on field survey data from nuclear power plants in Korea was made. Table 1 lists the mixing ratio and composition for the simulated noncombustible wastes.

Table 1 Mixing ratio and composition for the simulated wastes.

Simulated waste sample No	concrete (g)	glass (g)	sand (g)	Cs_2CO_3 (g)	pig iron (g)	LD slag (g)
1	600	300	-	9.55	4000	460
2	600	200	200	9.57	4000	564
3	1000	-	-	8.69	4000	420
4	800	200	-	9.00	4000	475
5	600	400	-	9.30	4000	520
6	500	500	-	9.49	4000	550

4.3. Slag Melting

Operation of a plasma melting system had an initial quantity of 4kg of pig iron in the furnace for each run so as to provide for an initial melting bath. Afterwards, simulated noncombustible wastes plus additives were poured into the furnace. The simulated wastes of noncombustible materials were melted in the furnace and maintained in the furnace for about half an hour at temperatures between 1100 and 1300 by controlling plasma output. The melting point of slag varied as a function of the type and amount of additives. In this experiment, in order to easily melt the slag and to protect the refractory of the furnace, the basicity (CaO/SiO_2) of the molten slag was maintained at about 0.5 by calculating the chemical composition of feed material. High viscosity imposes problems for melting and pouring. Therefore, additives are required to control the viscosity of molten slag. Generally, the chemical composition of noncombustible wastes such as concrete, sand, and glass are for the most part, composed of SiO_2 as shown on Table 2.

Table 2 Chemical composition of simulated waste plus LD slag

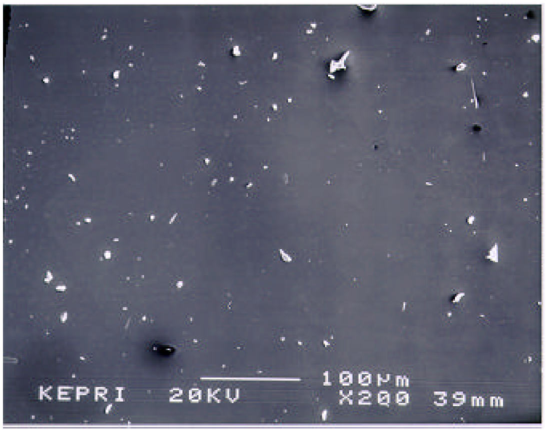
Sample	Weight Ratio(%) (Concrete/Glass/ Sand/Ldslag)	Total Weight	Chemical Composition(%)							
			SiO_2	Al_2O_3	CaO	MgO	Fe_2O_3	Na_2O	K_2O	CaO/ SiO_2
1	44/22/0/34	1560	47.0	7.2	23.5	4.9	8.0	3.3	0.3	0.5
2	38/13/13/36	1562	46.7	8.2	23.3	4.9	8.7	1.8	0.2	0.5
3	70/0/0/30	1420	45.8	11.0	22.9	5.2	8.8	0.01	0.45	0.5
4	54/14/0/32	1470	46.5	8.87	23.2	5.0	8.3	1.9	0.4	0.5
5	40/26/0/34	1520	47.2	6.7	23.6	4.8	7.8	3.7	1.6	0.5
6	32/32/0/36	1550	47.4	5.7	23.8	4.7	7.6	4.5	0.2	0.5

which the chemical composition is shown in Table 3) was used.

Table 3 Chemical composition of wasted LD slag

Species	Chemical Composition								
	SiO_2	Al_2O_3	CaO	MgO	MnO	P_2O_5	S	T.Fe	i.g.loss	CaO/SiO_2
LD slag	14.8	1.5	46.1	6.3	5.4	1.7	0.08	16.4	7.62	3.11

After each run, material which was converted into slag by the high temperature plasma was taken from the furnace. Molten slag was poured into a casting mold by tilting the furnace downwards. Material at the center of the mold will cool more slowly than near the edge. Slow cooling is expected to produce a relatively better slag. As expected, the slag appeared in general to be quite glassy (Eschenbach, 1995). Some slag had a degree of crystallinity as well as glassiness. Figure 1 is a SEM photomicrograph of a slag sample taken from the center of sample 1 of the produced slag.



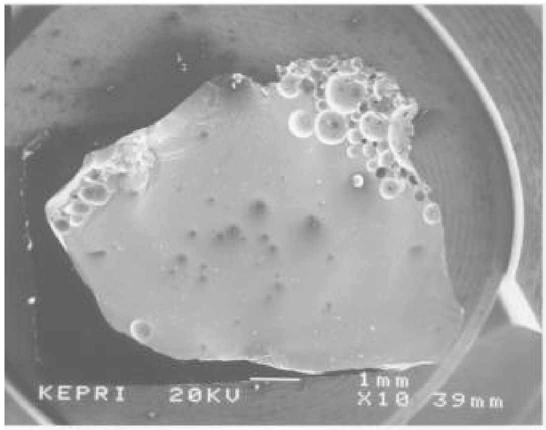


Figure 1 SEM photomicrograph of a slag sample

4.4. Intactness Test

4.4.1 Toxicity Leaching Tests

A leaching test was carried out using the standard Toxicity Characteristics Leaching Procedure (TCLP) tests on melted sample slags from experiments.

The TCLP analysis of the produced slag revealed no significant leaching of any hazardous chemical compound. All compounds screened in the TCLP were below the regulatory detection limits. This means the produced slag has very high resistance to leaching (Eschenbach & Schlienger et al., 1996). Table 4 shows the results of TCLP tests for the produced slag.

Table 4 Results of TCLP tests for the produced slags

Sample	As	Cd	Cr	Pb	Si	B	Na	Mg	Al	K
1	0.12	0.25	0.13	0.13	17.59	0.63	14.69	23.49	11.38	7.01
2	0.16	0.25	0.17	0.13	43.73	0.27	26.20	46.21	17.96	17.14
3	0.18	0.45	0.18	0.18	24.73	1.04	7.19	26.50	17.97	11.48
4	0.17	0.42	0.11	0.17	26.28	0.84	16.62	28.11	14.64	15.65
5	0.16	0.19	0.02	0.11	27.85	0.37	162.53	113.35	5.46	22.73
6	0.22	0.31	0.06	0.15	67.36	0.37	167.39	103.54	13.34	27.83
Sample	**Ca**	**Mn**	**Fe**	**Ni**	**Cu**	**Zn**	**Cs**	**Se**	**Hg**	
1	234.68	8.78	1172.92	0.15	0.04	0.74	1.73	1.55	< 0.03	
2	396.95	16.47	1096.91	0.11	0.03	0.71	2.49	1.62	< 0.03	
3	194.63	10.55	1777.90	0.13	0.03	0.72	1.90	2.50	< 0.03	
4	157.99	10.73	1833.15	0.10	0.03	0.71	1.98	1.90	< 0.03	
5	1714.82	27.10	537.51	0.15	0.03	0.88	< 0.01	0.66	< 0.03	
6	1361.86	32.69	2261.04	0.12	0.03	0.90	2.01	0.91	< 0.03	

4.4.2 Total Content Analysis

Both the nitric acid digestion method and the microwave digestion method were carried out in order to analyze the total content of produced slags. In the case of the nitric acid digestion method, most of the slag samples were observed to become hardened such that further pouring of digestive fluid resulted in splashing. Hence, this method was considered unsuitable. Instead, the microwave digestion method was employed after being found to be suitable. Table 5 shows the results of the total content analysis for the produced slag by microwave digestion method.

Table 5 Results of total content analysis for the produced slag

Sample	SiO_2	Al_2O_3	CaO	MgO	K_2O	Na_2O	Fe	CaO/SiO_2
1	28.57	7.18	21.85	3.36	1.88	4.94	18.31	0.765
2	38.77	10.67	29.57	5.08	2.19	3.82	9.69	0.763
3	34.78	6.58	15.32	2.14	2.26	1.77	21.65	0.440
4	34.89	4.15	10.76	1.28	2.70	4.10	6.83	0.308
5	28.06	6.27	25.05	4.33	2.09	6.05	10.69	0.893
6	33.88	3.99	16.19	3.11	1.31	7.68	7.66	0.478

4.4.3 Specific Gravity Measurements

The samples are taken from the center of the produced slag in order to obtain comparative results. It can be said that measured data are a reflection of the density of the produced slag. Table 6 shows the specific gravity for the produced slag.

Table 6 Results of specific gravity measurement for the produced slag.

Sample	Specific Weight
1	2.60
2	2.49
3	2.44
4	2.60
5	2.64
6	2.61

4.4.4 Other Physical Properties of the Slag

The volume reduction factor was found to be low compared to volume reduction factors obtained from treating combustible waste via pretreatment methods such as pyrolysis.

From observation, the slag appeared to be of sufficient strength for physical durabiity.

4.4.5 Analysis of Tracers

In order to examine the capture rate of cesium within the produced slag, the tracer of cesium carbonate (Cs_2CO_3) was added to the simulated waste. After melting, the input cesium-133 was compared with cesium-133 measured from the slag samples. With respect to the behavior of cesium-133, it was found that a considerably large percent of cesium-133 retained in the simulated waste was captured within the molten slag. Table 7 shows the tracer results of cesium-133 in the produced slag.

Table 7 Tracer results of Cs-133 in the produced slag

Sample	Input Cs (mg/Kg)	Measured Cs (mg/kg)	Capture Rate
1	5000	3364	67.3
2	5000	2362	47.2
3	5000	2404	48.1
4	5000	2446	48.9
5	5000	2708	54.2
6	5000	2276	45.5

5. CONCLUSIONS

The first phase of experiments conducted in this research program was successful in demonstrating the melting of a simulated noncombustible waste. It is believed that the plasma melting technology provides a heat source with high energy density well suited for treating mixed noncombustible materials such as concrete, sand and glass. The simulated waste that was subjected to the plasma arc flame was completely transformed into a vitrified mass of rock-like material. The slags in general appeared to be quite glassy. These slags were classified as non-hazardous substances, and were found to carry sufficient strength. An important factor was the melting point of slag which depends on the type and amount of additives. Generally, slag with lower melting points are less resistant to leaching than slag with higher melting points. It has been observed that difficulties with system operation occur if the viscosity of the molten slag is not proper, and that chemical erosion on the refractory of the furnace occurs if basicity is not proper. To overcome these effects, the composition ratio of noncombustible waste should be carefully considered. All slag produced in plasma melting released a considerably lower amount of hazardous chemical compounds from leaching tests than the regulatory limits.

TCLP tests have indicated that produced slag has very high resistance to leaching. We believe that plasma melting technology offers great potential to achieve volume reduction and stabilization of noncombustible radwaste in an environmentally safe and cost-effective manner for radioactive waste disposal.

In the future, through a series of melting experiments using various types of simulated noncombustible waste plus additives, optimal slag formulation and capture of volatile radioisotopes such as cesium in the produced slag will be examined; product consistency (a leaching test) will be performed; and a diagram of optimal slag composition by waste type will be made. On the basis of these data, a pilot plant of a 200 kW plasma melter will be designed and constructed. The data will eventually be utilized for the design of a commercial plasma melter.

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KEY WORDS

Non-combustible Waste, Plasma Melting, Basicity, Additives, Molten Slag, Leaching