



Destruction of inorganic municipal solid waste incinerator fly ash in a DC arc plasma furnace

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ABSTRACT

Due to the toxicity of dioxins, furans and heavy metals, there is a growing environmental concern on municipal solid waste incinerator (MSWI) fly ash in China. The purpose of this study is directed towards the volume-reduction of fly ash without any additive by thermal plasma and recycling of vitrified slag. This process uses extremely high-temperature in an oxygen-starved environment to completely decompose complex waste into very simple molecules. For developing the proper plasma processes to treat MSWI fly ash, a new crucible-type plasma furnace was built. The melting process metamorphosed fly ash to granulated slag that was less than 1/3 of the volume of the fly ash, and about 64% of the weight of the fly ash. The safety of the vitrified slag was tested. The properties of the slag were affected by the differences in the cooling methods. Water-cooled and composite-cooled slag showed more excellent resistance against the leaching of heavy metals and can be utilized as building material without toxicity problems.

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1. Introduction

With the amount of municipal solid waste increasing drastically in China during the past several decades, and landfill site selection in major cities becoming increasingly difficult, the optimal method of disposal for most such wastes has been high-temperature incineration. According to the China Statistical Yearbook 2009 [1], 74 MSWI Plants were built in China before the end of 2008. Their total treatment capacity is 51,606 ton/day. However, problems associated with MSWI fly ash, such as contamination with dioxin, furan, and heavy metals have arisen.

Many alternative methods for hazardous fly ash treatment have been suggested and developed. One of these is the use of thermal plasmas, such as electric arcs. Many prior studies have shown that the thermal plasma is a promising technology for vitrification of hazardous fly ash [2–8]. It achieves detoxification and volume-reduction due to its high-temperature; and the plasma is simple to generate and control.

Plasma-based systems have a number of advantages over conventional high-temperature incinerators. The energy source in plasma reactors is electricity, rather than the energy released by combustion, and is therefore independent of the waste being destroyed. The higher temperatures and energy densities that are possible in plasma systems promote chemical reactions, allow shorter residence time, and make large throughputs possible in a

pilot-scale furnace. Their relatively small size allows plasma systems to be integrated into manufacturing processes with relative ease, allowing on-site destruction of wastes. The main drawback of the application of thermal plasmas is the relatively high cost, due in particular to the cost of electrical power and the plasma gas. Nevertheless, there are many applications in which plasma systems are economically viable [2–9]. The ASIPP process is an example of successful application of thermal plasma inorganic waste destruction technology.

The objective of the present study was to improve the process, and evaluate, on a laboratory scale, the reduction in volume and detoxification (removal of hazardous elements) of fly ash, its conversion to a stable slag, and the behavior of hazardous materials when subjected to leaching tests. In order to investigate the future practical application, the fly ash was treated without any additive. By testing the physical properties of the slag, we confirmed that the hazardous substances are immobilized in the slag and do not leach out. So it is safe and could be used as a fill for roadbed material, as a component in asphalt mixtures, in concrete structures, and in secondary products such as interlocking structural blocks [10].

2. The new system description

Fig. 1 shows the process flow diagram of the experimental apparatus we used for plasma vitrification. The system consisted of a plasma furnace, a feed subsystem, a slag gathering system, a metal gathering system, waste gas pollution abatement devices, and automatic control system. The plasma furnace is fired by a 100 kW direct current (DC) plasma arc, which is more steady and

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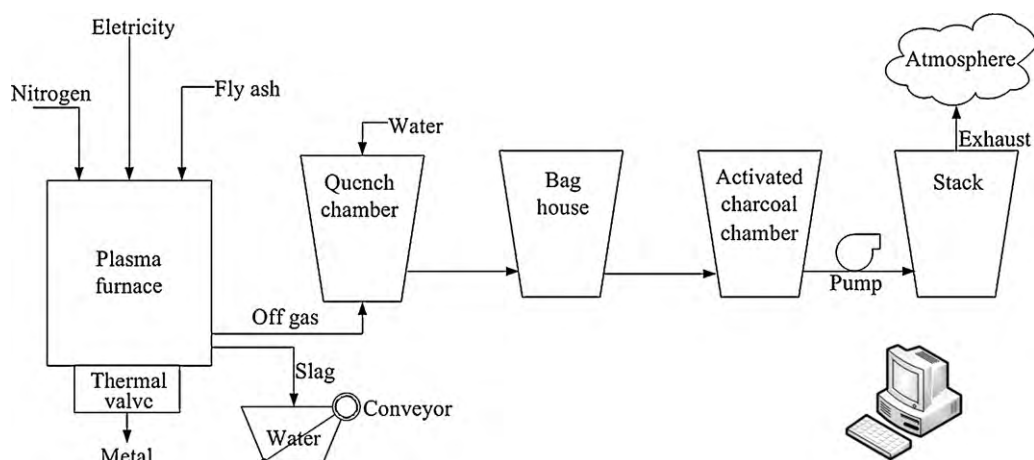


Fig. 1. Process flow diagram of the plasma vitrification.

efficient than an alternating current (AC) arc. Plasma gas is supplied through a hole along the axis of the graphite cathode and high-temperature plasma is generated by the electric discharge. The energy of the plasma is transferred to the anode of the furnace to melt fly ash, which is carried into the vessel continuously. Both heat and ultraviolet radiation from the arc can decompose dioxins into small molecules. Once melted, the slag continuously escapes from the furnace, with the hot waste gas. At the same time, the metals sink to the bottom of the furnace due to their specific gravity and are discharged from the furnace periodically. The waste gas enters the pollution abatement devices to be scrubbed [6,7].

2.1. The innovative design of the vitrification furnace

The furnace shown in Fig. 2 is comprised of a waste feeder system, a graphite crucible, a graphite cathode, four graphite anodes, a gas-cooling mechanism for the cathode, an external surface water-cooling for the anodes and a tapping hole for continuous slag and waste gas removal.

To depress the evolution of NO_x and keep air out of the vessel, a new segmented-wheel gravity feeder was developed after many experiments. The feeder system consists of a motor that drives a segmented-wheel hopper intended for feeding solid waste having a maximum particle size of 1 cm. Fly ash is carried into the vessel through the segmented-wheel hopper vertically, keeping air from the vessel. The feed rate is adjustable by varying the speed of the motor. The feed rate is continuous and very steady, compared to a screw conveyor feeder.

Bottom of the graphite crucible is cone-shaped in order to treat fly ash more sufficiently and drastically, compared to a flat-bottomed one. A commercial CFD code FLUENT was used and modified to model a DC plasma arc and melting slag by solving the fully coupled conservation equations. The boundary conditions are consistent with the experimental data. As shown in Fig. 3, right side of the figure is a cone-bottomed crucible and left is a flat-bottomed crucible. The comparison of temperature distributions is shown by the numerical simulation of the plasma arc and the inner vessel shown in Fig. 3, which demonstrates that the temperature of slag is higher and more regular in a cone-bottomed crucible than in a flat-bottomed crucible.

The slag escapes continuously from the tap hole located on the left side of the crucible, close to the bottom of the vessel. The design of the tap hole and its shield is very skilled, because the tap hole and shield must separate molten fly ash from untreated fly ash during the continuous operation, ensuring that residence time is sufficient

for thorough destruction of the hazardous wastes. Only molten slag can flow around the shield and reach the tap hole. Hot waste gas flows out of the vessel through the same passage as the molten slag; this prevents the slag temperature from dropping.

Typically in the plasma vessel, the temperature near the plasma arc may reach several thousand kelvins while the average temperature in the vessel is somewhat more than 1900 K. The furnace is lined with a suitable high-temperature refractory material so its surface does not need water-cooling, which raises the total energy efficiency.

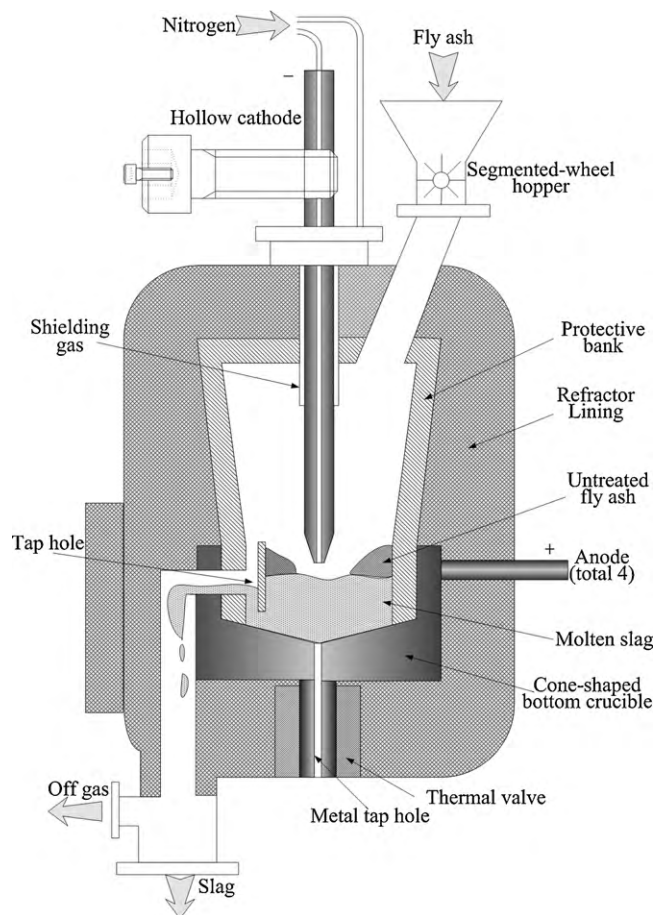


Fig. 2. Schematic diagram of vitrification furnace.

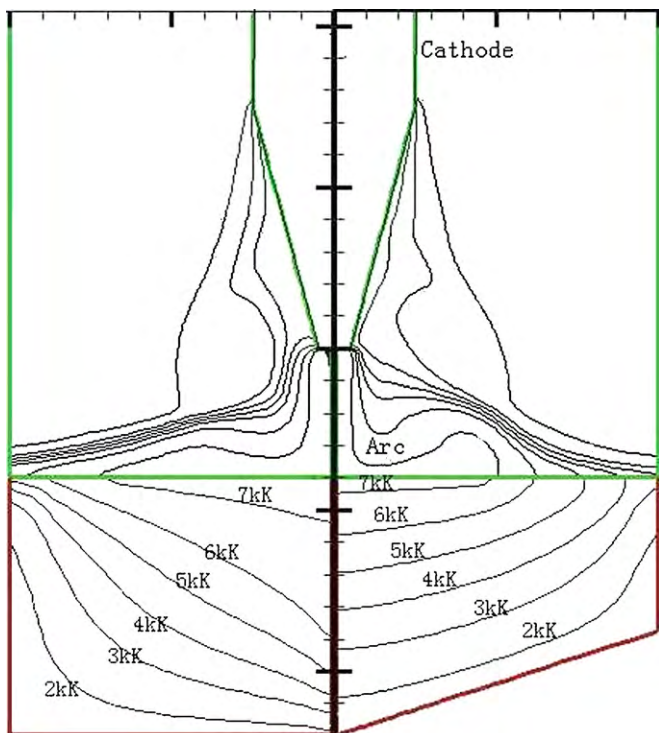


Fig. 3. Comparison of temperature distributions in a cone-bottomed crucible (right side of the figure) and a flat-bottomed crucible (left side of the figure).

2.2. Thermal valve

The thermal valve at the bottom of the crucible is used primarily to remove metal from the vessel, as shown in Fig. 2. The temperature of valve body is normally much lower than that of the melting slag, so a plug of frozen slag forms in the valve body and keeps molten material from escaping the vessel. When metals need to be discharged from the furnace, heat energy from an induction coil built into the thermal valve melts and breaks the thin layer of the slag plug. This technique was found to be also effective at tapping, when the vessel needs to be cleaned.

2.3. Pollution abatement devices

As shown in Fig. 1, a water quench chamber is located at the outlet of the waste gas. Its role is to cool the waste gas quickly to approximately 378 K, so as to minimize any production of dioxins, furans or other compounds. It also removes water-soluble components of the waste gas including hydrogen halide gas and most oxides of sulphur. The quench vessel uses two atomizing nozzles to quench the waste gas.

After quenching, the cooled waste gas passes through a bag-house dust catcher to remove suspended particles, and then through an activated charcoal chamber to remove acid gases and halogens. Finally, it is released into the atmosphere.

The waste gas, whose composition is regularly monitored, typically contains nitrogen and carbon dioxide and, in some cases, water vapor [3,7].

3. Experimental process

3.1. Fly ash pre-treatment

The fly ash used in this investigation was obtained from fabric filters in an air pollution control device at a municipal solid waste incinerator located in Zhejiang Province, China. In order to investi-

Table 1
Experimental conditions.

Item	Specification
Melting furnace type	Graphite-electrode plasma melting furnace
DC plasma arc	100 kW (100 V × 1000 A)
Plasma and shielding gas	Nitrogen
Plasma gas flow rate	12 L/min
Ash throughput	100 kg/h (0.0278 kg/s)

gate the future practical application, the fly ash was treated without any reductants or slagging agents. Fly ash is an inorganic powder, whose average particle diameter is only 42.14 μm. The fly ash, consisting of very fine particles, would be blown away by the plasma gas, escape treatment in the plasma and be carried out by the waste gas, which would lead to secondary pollution, so it could not be placed on the crucible in its natural state. In order to be sure that the fly ash reacted thoroughly, it was pre-treated in a dry granulate machine that compressed it into particles whose average diameter was about 5 mm.

3.2. Operation

Pelletized fly ash without any additive was carried into the vessel via segmented-wheel gravity feeder at a constant speed. This configuration ensures that the feed is introduced directly into the high energy-density anode zone of the arc, where rapid heating and reaction take place, and that the losses of material are minimized. The heating rates of fly ash were controlled to about 10 K/min. The molten slag was kept above 1700 K for 10 min. Operating procedures were used to maintain the operating temperature by controlling the DC plasma power. Standard chemical engineering diagnostics such as monitoring of input gas flow rates were used. Gas chromatography was used routinely to measure the waste gas composition as process parameters were varied. In addition, a laser-scattering technique and several thermocouple thermometers were used to measure temperatures in the vessel. Gas temperatures were measured at the exit of the vessel [10–14]. Experimental conditions are shown in Table 1.

3.3. Slag cooling

The properties of slag after its melting are influenced by cooling method, so we employed various ones. “Water-cooling” refers to cooling in a water tank. The slag can be quenched in a water tank to produce granulated slag, collected in a slag mold to form ingots, and the quenching water simultaneously works as water-seal, which keeps air from the vessel.

“Air-cooling” refers to cooling in natural convectional air. “Composite-cooling” implies that slag was cooled in air for 10 min and then quenched in water [10].

The various cooled slags were separately pounded and ground into small pieces with a mortar and pestle for material characterizations. An XRF-1800 X-ray fluorescence spectrometer was used to examine the fly ash example and some specially polished microstructure materials. The leachate solution was analyzed for Zn, Cd, Cr, Pb, As, and Hg by following the standard leaching procedure. The mobility of hazardous metal species in fly ash and slags was evaluated by the Toxicity Characteristic Leaching Procedure (TCLP) [10]. The concentrations of metal species in extracts, including As and Hg were determined by Atomic Fluorescence Spectroscopy (AFS). The concentration of Zn in the leachate solution was analyzed using a plasma atomic emission spectrometer. Others were determined with a Graphite Furnace Atomic Absorption Spectrometer (GFAAS).

Table 2
XRF analysis of fly ash and various slags (wt.%).

Item	Fly ash	Water-cooled slag	Air-cooled slag	Composite-cooled slag
CaO	26.8418	35.8537	37.8803	36.6723
SiO ₂	16.0115	35.4918	34.2402	36.2157
Cl	14.5447	0.3053	0.7633	0.2729
SO ₃	13.7206	0.7297	0.7495	0.8004
K ₂ O	9.7911	0.8157	1.0741	0.7253
Al ₂ O ₃	4.3900	13.2816	10.9115	13.8906
Fe ₂ O ₃	3.4887	3.6530	4.3990	2.2067
NaCl	3.4451	0.7177	0.8326	0.6926
P ₂ O ₅	2.0601	1.1561	1.5555	0.6434
MgO	1.7615	2.8018	2.6752	2.7985
ZnO	1.3061	0.1817	0.4544	0.1626
TiO ₂	1.2495	1.6991	2.0113	1.5103
PbO	0.4562	ND	0.0319	ND
MnO	0.1836	0.2680	0.3251	0.2693
CuO	0.1518	0.0309	0.0588	0.0234
SnO ₂	0.1501	0.0394	0.0521	0.0414
BaO	0.0991	0.1406	0.2354	ND
SrO	0.0977	0.1382	0.1671	0.1446
Cr ₂ O ₃	0.0784	0.1557	0.2201	0.1356
ZrO ₂	0.0425	2.5219	1.3286	2.7539
Y ₂ O ₃	ND	0.0360	0.0247	0.0402

Note: NA – not available, ND – not detected.

Table 3
Metal loading test results.

Samples	Loading test results (mg/kg)					
	Zn	Cd	Cr	Pb	As	Hg
Fly ash	8100	172	182	2100	81	1.11
Air-cooled slag	3383	6.49	840.9	418.1	1.17	0.011
Water-cooled slag	1973	4.12	825.5	210	3.26	0.039
Composite-cooled slag	1230	4.22	474.1	131	2.88	0.015

Note: NA – not available, ND – not detected.

4. Results and discussion

4.1. The properties of untreated fly ash

4.1.1. XRF

Table 2 shows the chemical composition of fly ash by XRF. Metal oxide composition was estimated from the total amount of analyzed elements and the basicity from the CaO/SiO₂ ratio [15]. The major constituents are CaO, SiO₂, SO₃, K₂O, Al₂O₃, Fe₂O₃ and NaCl. High Na concentration is probably linked with NaCl, the typical salt in fly ash. Also, important contributions of P₂O₅, MgO, ZnO, and TiO₂ were detected. The SiO₂ gives resistance to leaching since heavy metals are fixed in the net structure and are not easily extracted [15].

4.1.2. Metal loading and leaching test

The metal loading of fly ash samples is shown in Table 3. The major constituents (>1000 mg/kg) in fly ash samples were Zn and Pb. The minor constituents (10–1000 mg/kg) include Cd, Cr, As

Table 4
Leaching test results of fly ash and various slags.

Samples	Toxicity leachate (mg/L)					
	Zn	Cd	Cr	Pb	As	Hg
Fly ash	0.38	0.0013	1.41	12.19	0.011	0.011
Air-cooled slag	0.02262	0.001455	0.002168	0.104	0.0055	<0.0005
Water-cooled slag	0.02660	0.001134	0.000814	0.0229	0.0006	<0.0005
Composite-cooled slag	0.03165	0.000197	0.000258	0.0315	NA	NA
Upper limit by law	100	1	5	5	5	0.1

Note: NA – not available, ND – not detected.

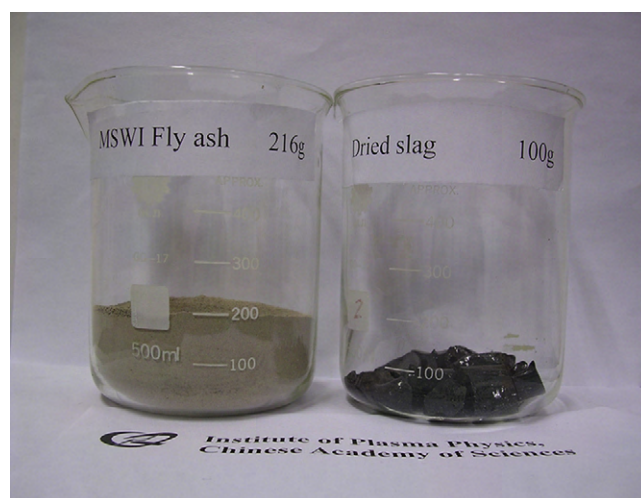


Fig. 4. Volume comparison of fly ash and air-cooled slag after the plasma treatment.

and Hg. Fly ash samples are characterized by high concentrations of volatile metals deposited over their large specific surface area [10,12]. Huang et al. [13] attributed the high concentrations of these metals to the non-recycle policy of waste before incineration. The concentrations of toxic elements such as Pb after leaching experiments of fly ash samples were much higher than the regulated limits of the Toxicity Characteristic Leaching Procedure (TCLP).

The leaching test was used to estimate the elution of hazardous waste. The leaching fraction of fly ash samples listed in Table 4 represents the percentage of metal transferred to the liquid phase during extraction. The leaching tests showed that heavy metals, such as Pb, in fly ash leached out at above the standard limit. This corroborates the high leaching potential of toxic metals from MSWI fly ash, which would be easily released in landfill [11]. So fly ash should be detoxified by plasma vitrification before its final disposal.

4.2. Volume and weight reductions after treatment

The fly ash feeds in general have large volume-reduction ratios (average 80.2%) after the plasma treatment, as shown in Fig. 4. However, the weight reduction ratio is about 36% on average. This could be due to the fact that the fly ash had less combustible or organic constituents [2].

4.3. Comparison of cooling methods

4.3.1. Surface characteristics

The cooling method influences the properties of vitreous slag. All cooled slags have strong physical properties akin to ceramics. Water-cooled slag is granulated, porous, and friable, and the physical hardness decreased, while air-cooled or composite-cooled slag is hard and has better vitreous appearance.

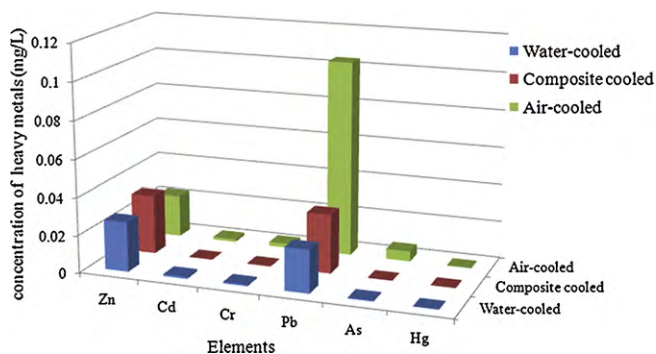


Fig. 5. The results of slag leaching test with respect to the different cooling methods.

4.3.2. Metal compositions of slag

Table 3 illustrates the metal compositions in slags with different cooling methods. It can be seen from the table that the concentrations of Zn, Cd and Pb in air-cooled slag were significantly higher than in slags cooled by the other two methods. Cr is almost same in the quenched and air-cooled slags, while the As and Hg are the opposite, because they are lower in the air-cooled than in the water-quenched and composite-cooled slag.

This could be due to the fact that the compounds of Zn, Cd and Pb in slag dissolved in water, resulting in lowering their total amount of heavy metal immobilized. As and Hg are extremely volatile metals, and easy to gasify, but they are not water-soluble. So some of them will spread into the air during air-cooling process, and their contents are lower than that of water-quenched slag.

A previous study reported that the basicity of the fly ash would affect vitrification, and when the basicity >0.990, it is not favorable for vitrification, whether by air-cooling or water-cooling [16]. In order to investigate the future practical application, the fly ash was treated without any additive. The basicity of the fly ash is about 1.676, so this investigation is to research fly ash on this basicity. It clearly demonstrates that water-cooling actually has a significant effect on enhancing the formation of glassy amorphous structures. The formation of glassy amorphous structures drastically reduces the specific surface area [17,18]. Therefore, it could be expected that water-quenched slag would have a smaller surface area than air-cooled and composite-cooled slags, and this would affect the leaching behavior of metals.

4.3.3. Metal leaching behavior of slags

Table 4 shows the compositions of the fly ash and the slags to serve as a reference to evaluate the leaching behaviors of heavy metals. As shown in Fig. 5, the concentrations of Cr, Cd, Pb and As in leachates from air-cooled slag were relatively higher than those from water-quenched and composite-cooled slags. A similar result found that Cr would be more mobile with a slower cooling rate due to a more intensive surface oxidation of the slag to form more leachable Cr^{6+} [17]. For Zn, it was observed in these slags that the concentration in the extract decreased. In composite-cooled slag, leaching of As and Hg was undetectable. Fig. 5 shows that the concentrations of Pb leached from slags were all under 0.5 mg/L, revealing that the cooling method had no notable influence on the mobility of Pb. After vitrification, leachate concentrations of Zn were <0.05 mg/L in all slags, indicating that its availability was drastically reduced [17].

The amount of heavy metal decreased when the water-cooling and composite-cooling methods were used. The fact indicates that the amorphous structure enhances the problem of leaching of heavy metals. The low leachability characteristics for Cr, Pb, and Cd thus presumably were because the heavy metal ions replaced parent ions (Al^{3+} and Ca^{2+} in this study) and enclosed in the framework

of silicates. The metal silicates were stable because of its complex formation mechanism, as detailed by Cheng et al. [2]. Upon water-cooling, the physical hardness decreased, but it had a higher value of Vickers hardness than the ceramic [10].

Table 4 lists the leaching values of some important elements. The test was based on slag reclaimed from the plasma process, and compared to normal fly ash from MSWI. The data show the higher efficiency of removing hazardous substances from the leachates, i.e., the concentrations of the hazardous substances in the leachates are less than 10% of the input amount. These values clearly show that the properties of slag from the plasma process are well below the limits given by the environmental standard. This means that the slag can be recycled into materials used for building.

5. Conclusions

The fly ash was treated in a pilot-scale plasma furnace with melting capacity of 100 kg/h and with the maximum plasma power of 100 kW. The process was improved in many details in order to maximize the efficiency. The leaching test was conducted for fly ash and various slags. We conclude that the heavy metals in the slags leached by a value below the standard level upon plasma vitrification. Experiments using various cooling methods were also performed.

The results obtained can be summarized as follows:

- (1) To depress the evolution of NO_x in the plasma gas, an operational method that keeps the furnace atmosphere reducing was established.
- (2) To ensure that the chemical reaction rates were sufficiently large relative to residence times, the tap hole and shield were improved.
- (3) To reduce secondary pollution, the fly ash in the form of fine power was pre-treated into small pellets.
- (4) The leaching tests showed that heavy metals, such as Pb, in fly ash leached out at above the standard limit. So fly ash must be detoxified by plasma vitrification before its final disposal [10].
- (5) The melting treatment metamorphoses ash to granulated slag, which is reduced to one-third in volume with no toxic problems.
- (6) The results of studying the various cooling methods show that a lower degree of leaching occurs from water-cooled and composite-cooled slag than from the other. After the high-temperature melting, microstructure materials were formed and consisted of predominantly a melilite group solid solution of gehlenite and akermanite. It was found that the crystallization was improved as the slag was water-cooled or composite-cooled. Heavy metals originated in the incinerated ash are confined in the treated silicate framework, hence yielding very low leachability results [2,5,10,15]. It also presented better resistance to the decomposition by an acid than the crystalline structure. Therefore, thermal plasma technology can be an effective method to treat incinerated ashes, and slag with preferred properties has the potential to serve as a viable alternative for construction applications [19–22].

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