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Production of glass-ceramic from incinerator fly ash

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Abstract

Vitrification is one of the technological options for treating toxic waste. This paper reports the results of the feasibility study of utilizing fly ash from municipal waste incinerators by powder sintering technology. This process was used to transform vitrified incinerated fly ash into a $CaO–Al_2O_3–SiO_2$ system of glass ceramic products. The major phase exhibited is gehlenite ($Ca_2Al_2SiO_7$), which belongs to the melilite group. The glass-ceramics exhibited attractive physical and mechanical properties. The products can be fabricated at 900 °C/2 h for construction purposes and have large application potential. © 2002 Elsevier Science Ltd and Techna S.r.l. All rights reserved.

Keywords: A. Sintering; D. Glass-ceramics; Vitrification

1. Introduction

Current methods of hazardous waste disposal generally include dumping in landfills and incineration. The integrity of dumps can be breached, thereby causing materials to leach into surrounding water tables. Incineration also has its problems. The ash residue, after incineration of municipal solid wastes, contains a large amount of hazardous materials such as dioxins and heavy metals, and therefore needs further treatment before it becomes reasonably safe for the environment. It is estimated that over two million tonnes of incinerated ash will be generated annually in Taiwan after the year 2003. Thus, a viable competing immobilization technology is needed. Prior studies on treating toxic waste [1–9] have shown that the high temperature vitrification process provides satisfactory results. Conversion of incinerated ash into a stable glass through vitrification is an attractive technology for waste treatment, because it can achieve large waste volume reductions, destroy organic compounds more effectively and create a durable waste form. A disadvantage of vitrification is that this energy-intensive process involves relatively high costs. Therefore, its use should aim for products of higher quality and optimized properties. Incinerated ash containing a large amount of CaO, SiO_2, and Al_2O_3 can be a good raw material for $CaO–Al_2O_3–SiO_2$ system of glass-ceramic production. A judicious choice of raw mix along with appropriate heat treatment one can produce various crystalline phases, thus facilitating the immobilization of heavy metal ions in the framework of silicates.

In order to better understand the feasibility of treating the incinerated ash, this study was directed toward characterization of heat treatment using the powder sintering process for as-quenched slag generated from thermal molten technology. The products were characterized by Toxicity Characteristic Leaching Procedure (TCLP) for testing the hazardous materials, X-ray diffractometry (XRD) for crystal structure determination, and scanning electron microscopy (SEM) for microstructure/morphology observation. In addition, other properties, such as compressive strength, four-point bending strength, porosity, water absorption rate, density, electrical resistivity, thermal expansion coefficient and chemical resistance were also investigated.

2. Experimental procedures

The incinerated ash from one of the municipal solid waste incinerators in Taipei was used in this investigation. The incinerated ash has a wide particle size ranging from 0.2 to 500 μm with D_{50} of 28.4 μm. The chemical composition of the incinerated ash and as-quenched glass are shown in Table 1. Incinerated ash sample of ~3.5 kg was placed in a graphite crucible and induction-heated in air. The molten sample was kept at

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Table 1
Chemical composition of incinerated ash and as-quenched glass

Chemical composition	Incinerated ash (wt.%)	As-quenched glass (wt.%)
SiO_2	18.18	37.99
Al_2O_3	9.34	16.23
CaO	19.19	28.69
TiO_2	1.87	2.84
Fe_2O_3	1.83	0.26
MgO	2.74	3.83
Na_2O	8.51	4.02
K_2O	7.36	0.83
ZnO	3.25	0.05
PbO	0.88	0.03
Cr_2O_3	0.09	0.06
MnO	0.16	0.21

1400 °C for 20 min to ensure complete melting. Subsequently, the melt was rapidly quenched by pouring it into water, followed by drying and grinding to minus 200 mesh. The ground sample was pressed at 150 kgf/cm^2 into a 4×1.5×0.7 cm stainless mold without using any binder. The green body thus formed was then sintered at different temperatures (850, 900, 950, 1000 and 1050 °C, respectively) for 2 h, and then cooled to room temperature. The experimental procedures are shown in Fig. 1. The glass–ceramic materials were thus formed. A Hitachi S-4100 scanning electron microscope was used to examine the glass-ceramics. XRD was done with a Rigaku D/MAX-VB diffractometer with CuK_α radiation at 4°/min scanning speed. Crystalline phases were identified by comparing the intensities and positions of Bragg peaks with those listed in the Joint Committee on Powder Diffraction Standards (JCPDS) data files. Physical property evaluations such as density and porosity were performed according to the methods described elsewhere [10]. Four-point bending and compressive strength tests were carried out with a Testometric 220D bending test machine. Five grams of crushed samples with particle sizes of 0.5–1.0 mm were boiled in 50 ml acid/alkali (20 %wt.) solution for 1 h. Weight loss was then measured for evaluation of chemical resistance.

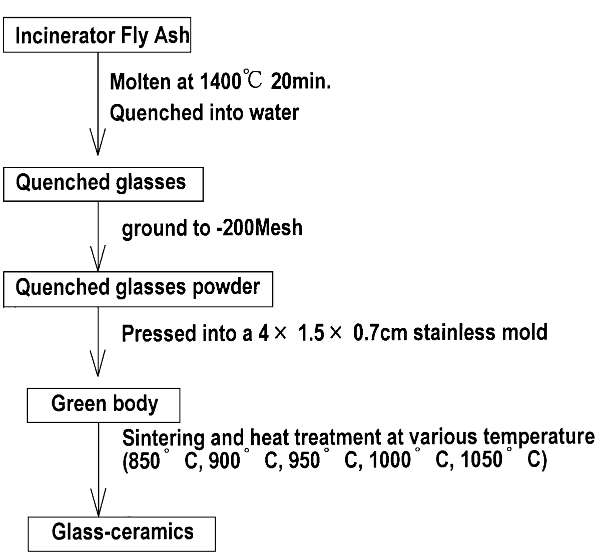

Fig. 1. Experimental procedure.

3. Results and discussion

3.1. Toxicity characteristic leaching procedure (TCLP) results

Toxicity characteristic leaching procedure (TCLP) results of the incinerated ash and as-quenched glass used in this study are given in Table 2. Each sample analyzed has insignificant leachability characteristics for Cr, Pb, Ni, Cu, and Cd. It is obvious that the vitrification in the form of as-quenched glass improves the chemical resistance. The extracted amounts of heavy metals are lower than the limits required by the EPA of Taiwan. The leachability characteristics for heavy metals are low because the heavy metal ions replaced Ca^{+2} or Al^{+3} ions and are held in the framework of glass.

3.2. Glass–ceramics microstructure characterization

X-ray diffractometry (XRD) analysis of the bulk-treated glass-ceramic samples (2 h of heat treatment at various temperatures) showed different tendencies towards crystallization as a function of thermal treatment temperatures. The results are shown in Fig. 2. Crystallization was detected from 850 °C, and the extent of crystallization was temperature-dependent. The major phase present shows a melilite group mineral—gehlenite ($Ca_2Al_2SiO_7$). Similar results were also reported for a slag-based glass-ceramic in Turkey [11] and the slag mixed with incinerator bottom ash in Italy [2]. SEM microstructural examinations after heat treatments at different temperatures were performed and the results are shown in Fig. 3. It can be seen that the microstructures of materials were not thoroughly homogeneous. In general, the crystal size (0.2–2 μm) increased with increasing heat treatment temperature.

Table 2
TCLP results for incinerated ash and ash-quenched glass

Elements	Incinerated ash leached (ppm)	As-quenched glass leached (ppm)
Zn	25.28	6.90
Cd	16.91	0.16
Pb	2.48	ND[a]
Cu	0.35	ND
Ni	0.15	ND
Cr	20.26	ND

[a] ND indicates not detected.

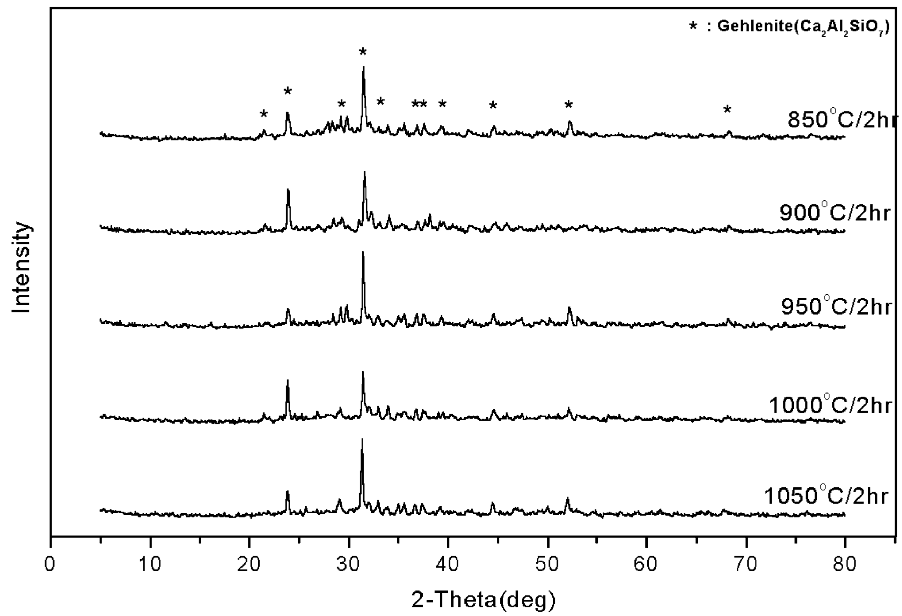

Fig. 2. XRD results of the glass-ceramics at different heat treatment temperature for 2 h.

Table 3
Properties of the glass-ceramics

Properties	850 °C	900 °C	950 °C	1000 °C	1050 °C
Density (g/cm^3)	2.03	2.07	2.17	2.26	2.21
Porosity (%)	25.67	22.66	19.29	11.76	11.92
Water adsorption (%)	12.63	10.82	7.41	5.18	5.87
Compressive strength (MPa)	41.44	56.29	53.96	38.75	31.31
Four-point bending strength (MPa)	19.96	22.57	17.00	12.09	11.99
Electric resistivity ($10^8\Omega$-cm)	11.72	4.59	3.23	2.95	1.05
Thermal expansion coefficient (10^{-6} mm/°C) (25–450 °C)	8.63	8.61	9.19	9.18	10.21

3.3. Physical and mechanical properties of the glass-ceramics

Various physical and mechanical properties of glass-ceramics obtained from vitrified incinerated fly ash with sintering at different temperatures are listed in Table 3. Densities of the glass-ceramics were in the range of 2.03–2.26 g/cm^3, and increased with increasing sintering temperature. Both porosity and water absorption correlated well each other and decreased with the increase of the sintering temperature. For mechanical properties, the compressive strength and four-point bending strength of glass–ceramics is a function of different heat-treatment temperature. The compressive strength and bending strength were observed to increase with increasing temperature from 850 to 900 °C. The maximum strength needed 22.57 MPa for four-point bending test and 56.29 MPa for compressive test at 900 °C. The strength then gradually decreases with the increase in heat-treatment temperature. The average coefficients of thermal expansion and electrical resistivity that are very important for high resistance applications in presence of temperature changes were 9.2×10^{-6} mm/°C and 4.7×10^{8} Ω-cm, respectively. Generally, the glass-ceramics produced by heat treatment at 900 °C show the best character.

Table 4
Chemical resistance for glass-ceramics

Chemical (20% wt.)	wt.% Loss				
	850 °C	900 °C	950 °C	1000 °C	1050 °C
CH3COOH	3.41	4.23	3.65	3.02	4.26
HCL	15.12	11.57	11.06	10.29	11.72
H2SO4	0.15	0.77	0.99	1.17	1.57
NaOH	0.74	1.92	0.55	0.55	5.19

3.4. Chemical resistance properties of glass-ceramics

Results for chemical resistance of the glass-ceramics are shown in Table 4. It can be seen that, in general, the durability of glass-ceramics does not correlate with the heat-treatment temperature but shows acceptable

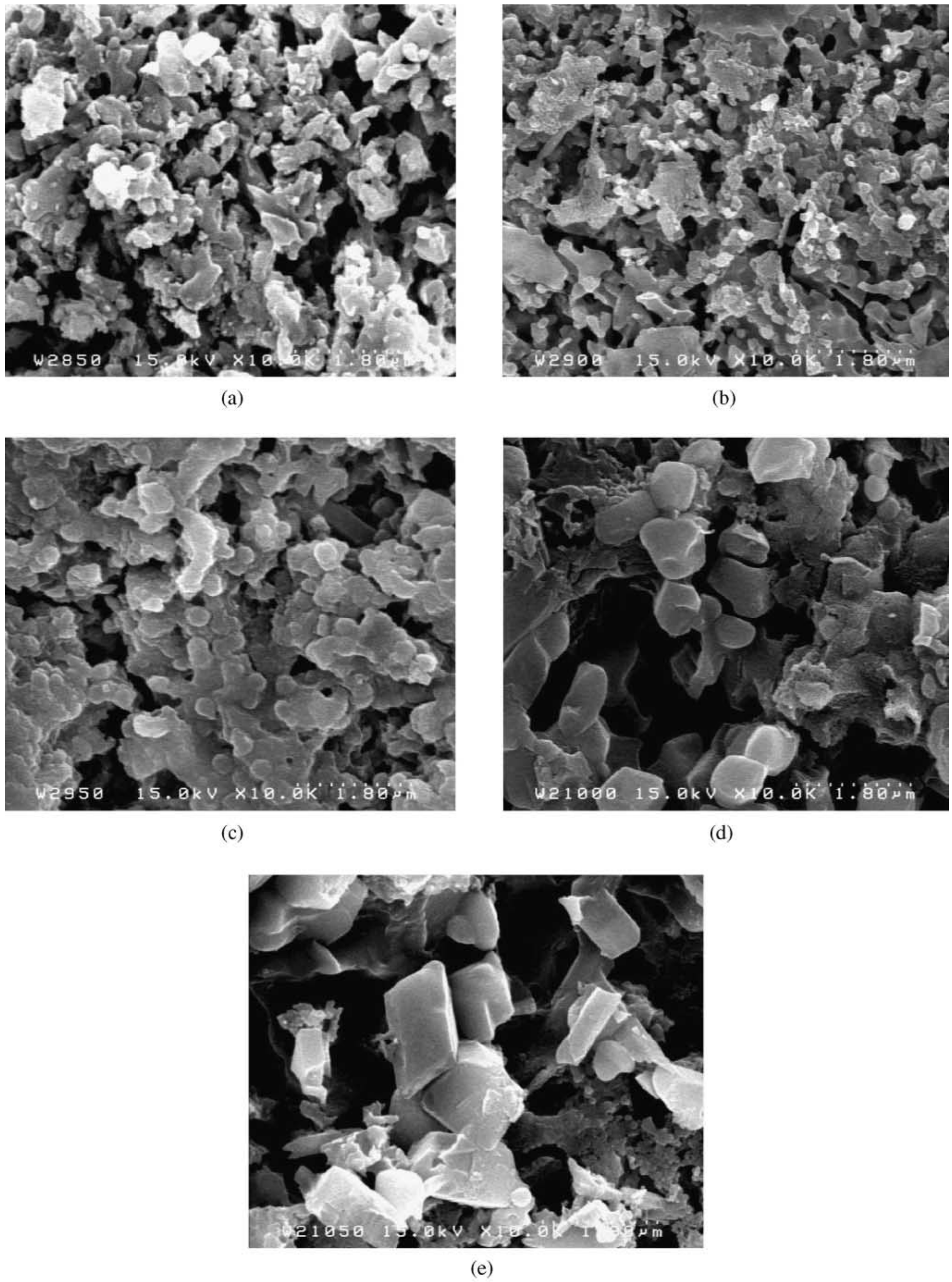

Fig. 3. SEM micrograph of microstructure material sample heat-treated for 2 h at various temperatures: (a) 850 °C, (b) 900 °C, (c) 950 °C, (d) 1000 °C, (e) 1050 °C.

behavior. However, relatively high weight losses for the HCl durability test may be caused by the gelatinization of gehlenite in HCl solution [12]. Improvement in chemical durability, especially with HCl attack, requires further investigation.

4. Conclusions

It was possible to produce glass-ceramic products of acceptable quality (except for relatively high weight losses attack by hydrochloric acid) by a vitrification of incinerated fly ash. The major constituent of the product was gehlenite. This product has good potential to serve as construction materials.

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