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Feasibility study of using brick made from municipal solid waste incinerator fly ash slag

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Abstract

This study deals with the effect of MSWI slag on fired clay bricks. Brick samples were heated to temperatures which varied from 800 to 1000 °C for 6 h, with a heating rate of 10 °C/min. The material properties of the resultant material then determined, including speciation variation, loss on ignition, shrinkage, bulk density, 24-h absorption rate and compressive strength. Toxicity Characteristic Leaching Procedure tests were also conducted. The results indicate that the heavy metal concentrations in the leachates met the current regulatory thresholds. Increasing the amount of MSWI slag resulted in a decrease in the water absorption rate and an increase in the compressive strength of the MSWI-slag bricks. The 24-h absorption rate and compressive strength of the MSWI-slag brick made from samples containing slag sintered at 1000 °C all met the Chinese National Standard (CNS) building requirements for second-class brick. The addition of MSWI slag to the mixture reduced the degree of firing shrinkage. This indicates that MSWI slag is indeed suitable for the partial replacement of clay in bricks. © 2006 Elsevier B.V. All rights reserved.

Keywords: Residues; Sintering; Shrinkage; Compressive strength; Water absorption; Toxicity Characteristic Leaching Procedure

1. Introduction

Incineration has two main advantages: it reduces the volume of municipal solid waste by about 90% and it reduces the reactivity of this material by the nearly complete destruction of all organic compounds contained therein. However, the incineration of municipal solid waste residues is still presenting problems, mainly with the bottom ash (about 250-300 kg/1000 kg of waste) and fly ash (about 25-30 kg/1000 kg of ash) still presents problems [1]. Furthermore, finding a suitable place for traditional landfill sites has been getting more and more difficult in Taiwan [2]. The incinerator ashes often contain large amounts of hazardous materials such as heavy metals and dioxins. It has been found that the Cd leaching concentration in fly ash exceeds the ROC EPA's current regulatory thresholds, and meaning it should be thus classified as hazardous waste [3]. Therefore, these hazardous materials can endanger the environment if it cannot be carefully treated.

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The melting process is probably the best method to resolve these problems. Melting treatment is a vitrification technology which has been identified as a potentially effective tool for immobilizing heavy metals into a nonleachable slag. This technology involves subjecting waste material to temperatures of around 1300-1400 °C, so that nonvolatile species become chemically bonded into the resultant matrix, rendering then nonleachable [4]. The high temperatures and lengthy times involved in such processes lead to the complete destruction of toxic organic compounds. However, the initial investment and operating costs are still expensive. So as to render the method more practical, the melting process should include the option of producing recyclable products. This is a goal of a resource recycling society and should help to extend the life of existing landfill sites. The process has already been employed to produce such diverse products as ceramics, metals and various composite materials. Recently, the process has also been applied to the reuse of MSWI slag in aggregates [5], asphalt paving [6], water-permeable brick and flagstones, sand [7], cement [8] and other construction materials.

Generally, the crystallization involved in the vitrification treatment could take place in different modes. (1) Crystallization occurs during very slow cooling; various minerals can be formed

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when the melt is gradually cooled. The crystallization of igneous rocks, such as granite, belongs to this type. (2) Crystallization occurs upon supercooling. In order to avoid forming high temperature crystalline phases, such as magnetite or olivine, the melt is rapidly cooled to certain temperatures to obtain individual mineral phases. (3) Crystallization takes place during vitrification heat treatment [9]. To form a better glass-ceramic the crystalline behavior of these minerals needs to be encouraged by higher heat treatment temperatures. Both the porosity and water absorption rate properties are improved with increasing heat treatment temperatures [10]. The sintering process consists of a thermal treatment for coherently bonding particles, in order to enhance the strength and the other engineering properties of the compacted particles. It is an effective alternative treatment for the resource recycling of waste. The thermal heating destroys organic residue and stabilizes inorganic material and metals by incorporating oxides from the elemental constituents into a ceramic-like material [11–14]. In this study, employed heating temperatures varied from 800 to 1000 °C for 6 h with heating rate of 10 °C/min and investigated the feasibility of combined clay and MSWI slag.

2. Materials and methods

2.1. Raw materials

The fly ash used in this study was collected from the cyclone of a mass-burning incinerator located in the northern part of Taiwan. The incinerator, capable of processing 1350 metric tonnes of local municipal solid waste per day, is equipped with air pollution control devices (APCD) consisting of a cyclone, a semi-dry scrubber system and a fabric baghouse filter. Mostly cyclone ash and the scrubber ash (defined as fly ash) were used in this study. The physical properties of the clay and MSWI slag, including pH, density, moisture content and loss on ignition are summarized in Table 1. The particle size distribution of the clay and MSWI slag is also shown in Fig. 1.

The MSWI fly ash slag was prepared by first melting it at 1400 °C for 30 min. The molten slag was then water-quenched to produce a fine slag. The water-quenched slag was then further pulverized in a ballmill until the particles could pass through a #16 mesh sieve. The MSWI slag was homogenized and then the chemical composition was characterized. The clay sample was obtained from a local brick manufacturing plant. The size distributions of the raw materials are listed in Fig. 1. In order to get a uniform particle size, both the MSWI slag and the clay

Characteristics	of clay a	and MSWI	slag

Table 1

Characteristics	Clay	MSWI slag
pH	8.2 ± 0.1	9.8 ± 0.1
Density (g/cm ³)	2.5 ± 0.1	2.7 ± 0.2
Moisture content (%)	2.1 ± 0.3	0.3 ± 0.0
Loss on ignition (%)	6.8 ± 0.4	0.03 ± 0.0

Mean \pm standard deviation (n = 3).



Fig. 1. Particle size distribution of clay and MSWI fly ash slag.

were crushed and ground into particles that could pass through a #16 mesh sieve. They were dried prior to further applications. The specimens were prepared for XRF analysis by mixing 0.4 g of the sample and 4 g of 100 Spectroflux, at a dilution ratio of 1:10. Homogenized mixtures were placed in Pt–Au crucibles, and then treated for 1 h at 1000 °C in an electrical furnace. The homogeneous melted sample was recast into glass beads 2 mm thick and 32 mm in diameter. The chemical composition of the raw materials, as analyzed by XRF, is shown in Table 2. The heavy metal concentrations in the MSWI ash and slag samples were confirmed by ICP-AES. The samples were crushed, and the

Table 2

Composition, heavy metals and leaching concentrations in the clay, MSWI fly ash and slag

	Clay	MSWI fly ash	MSWI slag	Taiwan TCLF regulatory limits
Compositi	on (%)			
SiO ₂	61.5 ± 0.2	35.8 ± 4.4	35.0 ± 3.3	
Al_2O_3	15.8 ± 0.2	9.8 ± 0.8	16.5 ± 2.9	
Fe ₂ O ₃	6.1 ± 0.1	4.9 ± 0.2	3.8 ± 0.9	
CaO	0.4 ± 0.0	14.7 ± 1.1	25.0 ± 2.3	
MgO	1.3 ± 0.0	0.8 ± 0.0	2.9 ± 0.0	
SO ₃	0.1 ± 0.0	2.2 ± 0.6	1.4 ± 0.4	
K_2O	2.7 ± 0.9	5.3 ± 0.3	1.3 ± 0.2	
Cl-	-	5.0 ± 0.5	0.01 ± 0.00	
Total meta	l (mg/kg)			
Cu	22.8 ± 1.2	1409.3 ± 89.1	1001.8 ± 0.2	
Zn	106.4 ± 0.6	7115.8 ± 163.2	2720.6 ± 6.5	
Pb	71.7 ± 0.30	1284.0 ± 14.8	520.2 ± 0.1	
Cr	56.1 ± 2.1	811.6 ± 24.6	590.6 ± 2.0	
Cd	0.7 ± 0.30	80.2 ± 1.7	2.3 ± 0.02	
Leaching of	concentration (r	ng/L)		
Cu	0.1 ± 0.2	0.6 ± 0.1	0.3 ± 0.1	15.0
Zn	0.9 ± 0.1	16.2 ± 0.1	9.1 ± 0.3	_
Pb	0.1 ± 0.1	0.7 ± 0.0	0.36 ± 0.1	5.0
Cr	ND ^a	4.3 ± 0.3	ND	5.0
Cd	ND ^b	1.8 ± 0.2	ND	1.0

Mean \pm standard deviation (n = 3).

^a Detection limits < 0.016 mg/L.

^b Detection limits < 0.012 mg/L.

Table 3
Chinese National Standard Methods for chemical and physical tes

Analysis method
CNS 5090-A3089
CNS 2887
CNS 1127-R3042
CNS 1127-R3042
X-ray diffraction analysis
NIEA R201.13C, NIEA R302.20T (Cd), NIEA
R303.20T (Cr)
NIEA R305.20T (Cu), NIEA R306.20T (Pb),
NIEA R307.20T (Zn)

heavy metals were extracted by acid (HF:HClO₄:HNO₃ = 2:1:1). At least three samples were tested in each experiment.

2.2. Specimen processing

The feasibility of using MSWI slag in brick making was investigated. The MSWI slag content in the clay–MSWI slag mixture was varied from 0 to 40% (by weight). The mixtures were then homogenized in a blender and molded under 60 kg/cm^2 of pressure to form $50 \text{ mm}(L) \times 25 \text{ mm}(W) \times 50 \text{ mm}(H)$ bars. The results obtained from the laboratory tests can be applied to the commercial size brick since it was constructed by the same component and process. Therefore, the scale up of the brick sample should be acceptable in this study. The molded specimens were air-dried at room temperature for 24 h, and then oven dried at 80 °C for another 24 h to remove the water content. The dried specimens were then heated to a designated temperature (800, 900 and 1000 °C).

2.3. Analysis methods

The brick samples then underwent a series of tests including firing shrinkage, weight loss on ignition, water absorption, bulk density and compressive strength, to determine their quality in comparison to the Chinese National Standard (CNS) methods (Table 3). Crystalline phases of all the heat-treated samples and the untreated ground mixture were identified by X-ray diffraction (XRD) analysis.

3. Results and discussion

3.1. Clay and MSWI slag characteristics

The Atterberg limits of clay and MSWI slag are listed in Table 4. The Atterberg limits of clay were higher than those



Fig. 2. XRD patterns of clay, MSWI slag and brick samples.

of the clay-MSWI slag mixture used in this study. The percent reduction of liquid limits for the clay-MSWI slag mixtures were 5, 14 and 22% at MSWI slag substitution levels of 10, 20 and 40%, respectively. Table 2 shows the composition of the clay and the MSWI slag. The XRF analysis shows that the major components in the clay were SiO₂ (61.5%), Al₂O₃ (15.8%) and Fe₂O₃ (6.1%). The next most abundant components were K₂O (2.7%), MgO (1.3%) and CaO (0.36%). Further X-ray diffraction analysis revealed that the clays used in this research mainly consisted of SiO₂, Al₂O₃ and Fe₂O₃, which are suitable for the following sintering process. The major components observed in the MSWI slag were SiO₂ (35%), CaO (25%) and Al₂O₃ (16.5%). The next most components were Fe₂O₃ (4.9%), K₂O (3.2%) and MgO (2.9%). Fig. 2 shows that the MSWI fly ash slag did not show any crystallization peak.

The TCLP test results are shown in Table 2. High concentrations of chromium and cadmium were observed in the fly ash samples. The cadmium concentration was 1.8 mg/L, which exceeded the Taiwan EPA's regulatory threshold. The raw MSWI fly ash thus has to be treated before final disposal. The TCLP leaching concentrations for the target metals in the clay and the MSWI slag met the EPA's current regulatory thresholds, and are presented in Table 2. The resultant slag was stabilized because many heavy metals were immobilized in the glassy Si–O matrix

Table 4 Atterberg limits of clay and MSWI slag used in the study

	Clay	90% clay + 10% slag	80% clay + 20% slag	70% clay + 30% slag	60% clay + 40% slag
Liquid limit (%)	38.2 ± 0.3	36.2 ± 0.1	32.6 ± 0.3	30.3 ± 0.4	29.6 ± 0.6
Plastic limit (%)	25.2 ± 0.1	24.1 ± 0.3	22.3 ± 0.5	21.6 ± 0.3	21.0 ± 0.2
Plasticity index	13.0 ± 0.2	12.1 ± 0.2	10.3 ± 0.3	9.3 ± 0.2	8.6 ± 0.3

Mean \pm standard deviation (n = 3).



Fig. 3. Weight loss on ignition of the brick.



3.2. Weight loss on ignition of bricks

The weight loss which occurs in a monolith after sintering is related to the development of the porosity and the densification, and eventually has an affect on the compressive strength of the thermally treated samples [15,16]. The weight loss on ignition for a normal clay brick is 15%. Fig. 3 shows the weight loss on ignition of brick and the amount of MSWI slag added to the mixture at various heating temperatures. For a normal clay brick, the weight loss after heating to a temperature of 800, 900 or 1000 °C attributed to the organic matter content in the clay is 4.8, 5.6 and 6.0%, respectively. The results show that the addition of MSWI slag resulted in only an unapparent decrease in the weight loss on ignition. It is assumed that as the temperature was increased, the carbonate in clay became deformed into CO₂, so the weight loss in the brick was reduced. The weight loss on ignition in the bricks made for this study all met the CNS criteria. This observation indicates that clay and MSWI slag are compatible ingredients, so that MSWI slag can be used as a clay substitute.

3.3. Shrinkage of bricks

The quality of brick can be further measured by examining the shrinkage of brick. Based on the regulation of Taiwan, the shrinkage of brick is exhibited of less than 8%. Fig. 4 shows the amount of shrinkage after firing at various heating temperatures. The shrinkage of a normal clay brick is -1.0, 0.5 and 6.9% after heating to temperatures of 800, 900 and 1000 °C, respectively. When the MSWI slag content in the mixture varied from 0 to 40%, the brick shrinkage changed from -0.83 to 1.17, 1.05 to 1.47 and 3.1 to 3.6% with respect to heating temperatures of 800, 900 and 1000 °C, respectively. It is indicated that addition of MSWI slag should not apparently decrease the weight loss, by melted at 1400 °C.



3.4. Bulk density of bricks

During sintering, open and closed pores are usually formed. The minimum density corresponds to the maximum volume of closed pores in the sample. Densification is a pore-filling process that occurs during the liquid-phase flow and by pore shrinkage [17]. The measurements of the bulk density of samples with different proportions of ash and fired at three different temperatures are shown in Fig. 5. Clay bricks normally have a bulk density of $1.8-2.0 \text{ g/cm}^3$. The results indicate that increasing the temperature results in an increase in the bulk density (Fig. 5). The heating temperature can also affect the bulk density of the bricks. When the amount of MSWI slag was higher than 10% and the brick was fired at 1000 °C, the bulk density met the desired criteria. The results indicate that the bulk density of the bricks increased as the MSWI slag content increased. In Table 1, it is indicated the density of MSWI slag is greater than the clay.

3.5. Water absorption rate of the bricks

The water absorption rate, which refers to the weight of moisture in the pores compared to the sintered specimen's weight,



Fig. 5. Bulk density of the brick.



Fig. 6. Water absorption rate of brick.

is an effective index for evaluating the brick quality. The less water that infiltrates the brick, the greater its durability and resistance to the natural environment are expected. Fig. 6 shows the results of the water absorption tests for various MSWI slag-clay mixtures fired at the three different temperatures. The water absorption was from 17.8 to 20.1, 16.9 to 19.0 and 16.4 to 14.3% with respect to temperatures of 800, 900 and 1000 $^\circ \text{C},$ respectively. The specimen without MSWI slag, heated to a temperature of 1000 $^\circ$ C, met the first-class water absorption standard. The results indicate that when the MSWI slag content was decreased the water absorption of the bricks increased. However, when the amount of MSWI slag was higher than 10%, even although it was fired at 1000 °C, the water absorption of the brick only met the water absorption criteria for second-class brick. In addition, as the heating temperature increased, the amount of water absorption in the brick decreased. The smaller water absorption rate that occurred after heating at the higher temperature (1000 °C), suggests that local liquid-phase sintering occurred, which contributed to a decrease in the pore volume and thus the water absorption rate. Apparently the bonding ability of the mixture is related to the amount of MSWI slag added to the mixture.

3.6. Compressive strength of bricks

The compressive strength is the most important engineering quality index for building materials. The compressive strengths of the bricks made from MSWI slag–clay mixtures all met the CNS 1127-R3042 standards: 150 kg/cm^2 for a first-class brick and 100 kg/cm^2 for a second-class brick.

The results of compressive strength testing of the bricks made from both clay and MSWI slag mixtures are shown in Fig. 7. When the heating temperature increased to 800 and 900 °C, the compressive strength of the brick gradually increased. When the heating temperature was higher than 900 °C, the compressive strength of the clay and MSWI slag mixed brick samples met the criteria for first-class brick. When amounts of up to 40% MSWI slag were added to the brick heated to 1000 °C, the strength was similar to that of normal clay bricks. The results indicate



Fig. 7. Compressive strength of the brick.

that the optimum heating temperature for maximum compressive strength was 1000 °C. It is indicated that MSWI slag could indeed be converted into a glass–ceramic using this processing route. Similar crystallization behavior has been observed in glassy slag from MgO–Al₂O₃–SiO₂ systems [18]. It is concluded that MSWI slag can be blended with clay to produce brick.

3.7. X-ray diffraction patterns of the bricks

Fig. 2 shows the X-ray diffraction pattern of bricks heated at $1000 \,^{\circ}$ C. The MSWI slag was composed of calcium silicate and unknown crystal phase components. Clay is mainly composed of SiO₂, Al₂O₃ and Fe₂O₃. The amount of SiO₂ in the brick samples increased when the percentage of the added MSWI slag was increased. When amounts of MSWI slag were added to the bricks, heated to a temperature of $1000 \,^{\circ}$ C, there was an increase in the height of the Ca(Mg, Al)(Si, Al)₂O₆ peak, which indicates that increasing amounts were crystallized. MSWI slag is a product of a high temperature process and is comparatively stable, which prevents the formation of new crystal phases. A MSWI slag–clay brick mixture, sintered at $1000 \,^{\circ}$ C, can be considered to be a material which may have potential for use in the ceramic industry.

3.8. SEM observation of the bricks

SEM investigations were conducted in order to get a better understanding of the morphology of the microstructure. SEM images of the brick samples heated to 1000 °C at various substitute levels are shown in Fig. 8. It was found that mineral crystals in the brick samples increased when the percentage of added MSWI slag was increased. When amounts of up to 40% MSWI slag were added to the bricks heated to 1000 °C, the mineral crystal increase is similar to that of normal clay bricks. This is due to the higher driving force and the increasing crystal growth rate. This allowed fine grains to form, which caused an increase in the grain boundary area. Therefore, the physical and mechanical properties yielded better characteristics.



(a) 0%

(b) 10%





(d) 30%



(e) 40%

Fig. 8. SEM images of brick samples heat-treated at 1000 $^\circ C$ with various substitute levels.

4. Conclusions

This study has demonstrated a feasible way for using MSWI slag as a clay substitute to produce good quality bricks. Brick samples were heated to temperatures which varied from 800 to $1000 \,^{\circ}$ C for 6 h, with a heating rate of $10 \,^{\circ}$ C/min. The con-

clusions derived from the aforementioned experiments are as follows:

1. The TCLP leaching concentrations for the target metals in the clay and the MSWI slag all met the EPA's current regulatory thresholds.

- 2. The results show that the addition of MSWI slag resulted in an unapparent decrease in the weight loss on ignition.
- 3. The results indicated that the bulk density of the bricks increased when the MSWI slag content increased.
- 4. When amounts of up to 40% MSWI slag were added to the brick heated to 1000 °C, the strength was similar to that of normal clay bricks.
- 5. The heating temperature of 1000 °C, produced a significant densification, resulting in a total shrinkage in volume, a decrease in the water absorption rate and an increase in the density and compressive strength.
- 6. The correlation between the compressive strength, firing shrinkage and apparent density showed that compressive strength increased as the firing shrinkage and density increased, however, the compressive strength decreased as the water absorption rate increased.
- 7. The advantages of less firing shrinkage, less weight loss on ignition and greater compressive strength of MSWI slag–clay brick should stimulate the use of MSWI slag as a brick additive in the near future. It indicates that MSWI slag has potential as a material for brick components.
- 8. Keeping in view the saving of energy and maximum use of slag, it is apparent that mixture with 40% MSWI ash fired at 800 °C was optimal for meeting the brick quality standard.

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