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# Pozzolanic reaction of a mortar made with cement and slag vitrified from a MSWI ash-mix and LED sludge



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# HIGHLIGHTS

• MSWI ash-mix and LED sludge were melted to form a non-toxic slag.

• The strength of the slag-blended cement mortar (SBCM) is superior to OPCM specimen.

• SBCM reveals the greater strength by nano-particle effect and pozzolanic reaction.

• The proposed method provides an application of recycling MSWI ashes and LED sludge.

## ARTICLE INFO

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# ABSTRACT

Fly ash and scrubber ash that were generated by incinerators were mixed to produce municipal solid waste incinerator (MSWI) ash mix based on the proportions of the produced ash. In addition, 50 wt.% sludge produced during the manufacturing process of light emitting diodes (LEDs) was mixed into the MSWI ash mix, which was then melted to create MSWI-LED slag. This slag was used to replace 0, 5, 10, 20, and 30 wt.% cement and then the  $5 \times 5 \times 5$  cm<sup>3</sup> slag-blended cement mortar (SBCM) specimens were molded. Analyses of compressive strength and other properties were conducted. The results showed that the SBCM specimen at 1-7 days of age possessed a compressive strength close to or slightly higher than that of a ordinary Portland cement mortar (OPCM) specimen, whereas that at 14-90 days had a compressive strength 6-36% greater than that of OPCM specimens. The specimens were analyzed using X-ray diffraction (XRD), thermogravimetric and differential thermal analyses (TG/DTAs), and nuclear magnetic resonance (NMR). The results showed that the pozzolanic reaction in the slag increased compressive strength. Moreover, this study conducted a toxicity characteristic leaching procedure (TCLP) and investigated initial and final setting times and the fluidity of the specimens. The results indicated that MWSI-LED slag prevented heavy metal leaching, increased the compressive strength of specimens of different ages, shortened the initial and final setting times, and only slightly reduced fluidity. Replacing part of the cement in mortar with this slag facilitates waste recycling, enhances environmental protection, and meets the demand for sustainable development.

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## 1. Introduction

In Taiwan, most urban garbage is processed in incinerators, which generates fly ash and scrubber ash that contains multiple types of heavy metals harmful to human health. Incinerator waste is characterized as hazardous industrial waste [1–4] that is commonly solidified using cement and chelating agents before it is buried. However, landfills continue to leach toxic heavy metals

http://dx.doi.org/10.1016/j.conbuildmat.2014.04.088 0950-0618/© 2014 Elsevier Ltd. All rights reserved. [1,2], and the construction of landfills is becoming increasingly difficult in densely populated Taiwan. Thus, recycling the incinerated ash in incinerators is an urgent issue that requires a solution.

In addition, the production of light emitting diodes (LEDs) in the optoelectronic semiconductor industry produces waste sludge during grinding and polishing processes. The sludge is then turned into sludge cakes, which are known as LED sludge and are hazardous industrial waste. The sludge contains SiC, multiple types of oxidants, additives, dispersing agents, and organic and inorganic compounds used in grinding buffers. The amount of SiO<sub>2</sub> at the nanometer scale should not be neglected [3,4]. The particle

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diameter of SiO<sub>2</sub> is between 90 and 100 nm. Without proper handling, these materials not only harm the environment, but also pose a threat to human health. The manufacturing and processing factories of the Taiwanese optoelectronic semiconductor industry in science parks commission industrial waste landfills to handle their LED sludge; however, although the sludge is buried, risks remain. Currently, the development of photovoltaics and LEDs has been promoted globally. With the continuous growth of the LED production capacity in the optoelectronic semiconductor industry, the generated amount of LED sludge has increased dramatically, and the threat to the environment continues to grow. This urgent problem requires a solution.

Of these two types of industrial waste, studies on and applications of recycling incinerator bottom ash have been efficient. Currently, the recycled material can be applied to road bases, bricks, permeable bricks, and building tiles [5–10]. However, research on recycling and reusing incinerator fly ash is in the active development stages. Al-rawas et al. [11] substituted part of cement and sand with incinerator fly ash to conduct a study on cement mortar specimens. The concrete slump decreased when the sand was replaced, and the slump increased when the cement was replaced. The strength of the cement at 28 days was either approaching or slightly higher than that of plain cement mortar specimens. Lin et al. [12–14] pulverized slag that was produced using melted incinerator fly ash. The slag was used to replace 0%, 10%, and 20% cement and placed in a  $1 \times 1 \times 1$  in.<sup>3</sup> cement mortar specimen to conduct a series of analyses, including observation of hydration and exploration of the Al<sub>2</sub>O<sub>3</sub> hydration activity. Moreover, additional experiments on compressive strength were conducted in which slag was used to replace Portland I and II and Belite cements. The results showed that compared with the plain cement mortar specimen, the slag cement mortar specimen demonstrated little increase in compressive strength at the early stage; however, a significant increase in strength was observed at later stages. Although the slag-blended cement mortar specimen demonstrated strength comparable to that of the plain cement mortar specimen at the later stage, the strength in the early stage was inadequate. Lee et al. [15] used 10–40% of the municipal solid waste incinerator (MSWI) fly ash slag to replace Portland I cement and produced a  $5 \times 5 \times 5$  cm<sup>3</sup> mortar specimen, which demonstrated an increase in compressive strength of 75-89% on the 28th day, and an increase of 95-104% on the 90th day. This was similar to previous analysis results of mortar specimens, and replacing portions of cement with recycled MSWI fly ash slag appears possible. Lee et al. [16–18] used evenly mixed MSWI fly ash and scrubber ash and added waste materials that contained substantial SiO<sub>2</sub>. After the mixture was melted, the resulting slag was pulverized into powder, which was used to replace 20-40% of the cement in the specimen. Compared with ordinary Portland cement mortar (OPCM), the compressive strength of this replacement, at 1-14 days, was 64-83% of the OPCM. On Day 28, the strength was nearly identical, and the replacement showed a 102-111% increase in strength between Days 60 and 90. This also indicates that replacing a portion of cement with MSWI fly ash slag is possible. Siddique [19] stated that the addition of MSWI fly ash or slag into cement mortar lowered compressive strength at the early stage. Insufficient strength of concrete at the early stage slows the process of template removal, which is a disadvantage in practical construction. Therefore, the issue of insufficient strength requires an immediate solution. A comprehensive review of the literature indicated that a study on the cement replacement using melted slag composed of MSWI ash and LED sludge has not been conducted. This study pulverized this melted material to replace 5, 10, 20, and 30 wt.% of Portland I cement in a mortar specimen. This method efficiently solved the toxicity and insufficient strength at the early stage of cement placing.

#### 2. Materials and methods

#### 2.1. Materials

*Cement*: Domestic Portland I cement, specific gravity = 3.13, fineness = 3520 cm<sup>2</sup>/g.

Standard sand: Ottawa standard sand complying with ASTM C778 [20], specific gravity = 2.63.

*MSWI fly ash*: Collected from the dust collector cyclone of the Hsinchu Refuse Incinerator, light grey and yellow, specific gravity = 2.91.

*MSWI scrubber ash*: Collected from the dust collector cyclone of the Hsinchu Refuse Incinerator, grey and white, specific gravity = 2.62.

*MSWI ash mix*: An even mixture with a weight ratio of 1:3 of fly ash to scrubber ash that had been sifted by a #100 mesh, specific gravity = 2.69.

*LED sludge*: Produced by an LED factory in Hsinchu Science and Industrial Park, the LED sludge cakes were light blue; these were dried, pulverized into powder, sifted using a #100 mesh, and called LED sludge; ivory, specific gravity = 2.50, specific surface area =  $12,000 \text{ cm}^2/\text{g}$ .

*MSWI-LED slag*: MSWI ash mix was combined with LED sludge at a weight proportion of 1:0.5. The mixture was melted to obtain slag. The cooled slag was pulverized and the powder is grey and white, specific gravity = 2.57, specific surface area =  $5460 \text{ cm}^2/\text{g}$ .

#### 2.2. Methods

#### 2.2.1. The manufacturing process of MSWI-LED slag

After referencing previous studies [16–18], this study mixed the MSWI fly ash and scrubber ash at a proportion of 1:3. Ash mix and LED sludge were evenly mixed at a weight proportion of 1:0.5 and placed in an alumina crucible to be melted using an electric oven. The set program in the oven enabled continuous heating of the mixture until reaching 1350 °C. A constant temperature was maintained for 30 min and then the mixture was cooled by air to obtain black glass slag. The mixture is called MSWI-LED slag after it is pulverized by a ball mill and sifted using a #400 mesh (<38  $\mu$ m).

#### 2.2.2. Newly produced mortar

MSWI-LED slag was used to replace 0%, 5%, 10%, 20%, and 30% of the cement to conduct various experiments. Table 1 shows the mixture proportions. The fluidity experiment on the mortar was in compliance with the ASTM C1437 standard [21]. The mortar solidification experiment included an ASTM C187 cement standard consistency test [22]. The required water was measured before the initial and final setting experiments were conducted using the Vicat method [23] to measure the initial and final setting times.

#### 2.2.3. Compressive strength experiment

The mortar specimen in this study was in compliance with the requirement of the ASTM C109 compressive strength experiment standard [24]. The weight ratio of the cement to standard sand was 1:2.75, and the water-binder ratio (W/B) was 0.485. A 5  $\times$  5  $\times$  5 cm<sup>3</sup> mortar specimen was formed using this mixture (the control group was the ordinary Portland cement mortar, termed the OPCM specimen). Subsequently, the MSWI-LED slag was used to replace 5, 10, 20, and 30 wt.% of the cement in specimens of the same size (the treatment group was slag-blended cement mortar, termed the SBCM specimen), which was represented by SBCM (5%), SBCM (10%), SBCM (20%), and SBCM (30%). The proportions are presented in Table 1. After the specimen was cast, it was placed in a chamber and maintained at a constant temperature and humidity (23 °C; relative humidity 95%) for one day. The specimen was removed from the chamber, and then de-molded and cured in a saturated lime water chamber maintained at 23.0 ± 1.7 °C. The specimens were cured for 3, 7, 14, 28, 60, and 90 days. To comply with the requirements of the ASTM C109 compressive strength test, this study selected three specimens for each curing period to conduct the compressive strength test. The average compressive strength of the three selected specimens was considered the representative strength for that particular period during analysis and evaluation.

# 2.2.4. During the compressive strength tests, mercury intrusion porosimetry (MIP) was used

The OPCM and SBCM specimens for each curing period were separately preserved in methanol solutions to stop the specimens from hydrating. The OPCM and SBCM specimens that were preserved in methanol solutions were removed and placed in a 100 °C baking oven for 24 h. The specimens were then moved to a vacuumed desiccator to cool. Subsequently, the specimens were placed into a vacuumed mercury porosimeter (PoreMaster 60, Quantachrome, USA) for mercury testing and analysis.

#### 2.2.5. Toxicity characteristics leaching procedure (TCLP) test

The MSWI fly ash, ash mix, and MSWI-LED slag specimens were dried and pulverized. Based on the SW 846-1311 standard [25], a TCLP test was conducted to analyze the leaching of toxic heavy metal.

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