



## Detoxification of hazardous dust with marine sediment

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### ABSTRACT

Hazardous electric arc furnace dust containing dioxins/furans and heavy metals is blended with harbor sediment, fired at 950–1100 °C to prepare lightweight aggregates. Dust addition can lower the sintering temperature by about 100 °C, as compared to a typical industrial process. After firing at 950 °C and 1050 °C, more than 99.85% of dioxins/furans originally present in the dust have been removed and/or destructed in the mix containing a dust/sediment ratio of 50:100. The heavy metals leached from all fired mixes are far below Taiwan EPA legal limits. The particle density of the lightweight aggregates always decreases with increasing firing temperature. Greater addition of the dust results in a considerably lower particle density (mostly <math><2.0 \text{ g cm}^{-3}</math>) fired at 1050 °C and 1100 °C. However, firing at temperatures lower than 1050 °C produces no successful bloating, leading to a denser particle density (>2.0 g cm<sup>-3</sup>) that is typical of bricks.

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### 1. Introduction

The steel-manufacturing industry worldwide produces troublesome wastes – slag and dusts – in enormous amounts annually. Slag is generally non-hazardous, while dust is hazardous, as classified by the Taiwan Environmental Protection Administration (<http://waste.epa.gov.tw/prog/IndexFrame.asp?Func=5>), due to the content of both heavy metals and dioxins/furans. Slag can be readily recycled into construction material, but dusts are problematic.

As an island, there are thirteen commercial harbors, five industrial harbors, and one hundred and forty-nine fishery harbors in Taiwan. In order to keep ship navigation safe, some busy harbors that frequently build up bottom sediment on the sea bed require annual dredging. Final disposal of the dredged harbor sediment is a big task because of its huge mass.

If both electric arc furnace dust (EAFD) and harbor sediment are treated as wastes, cost for proper final disposal is high from the aspects of their hazardous and/or massive nature. Thus, based on the idea of waste recycling that will help the society grow in a sustainable way, this study aims to investigate the feasibility of recycling both EAFD and harbor sediment as an important construction material – lightweight aggregates. A thermal process for manufacturing lightweight aggregates seems to be an effective approach to remove the hazard of EAFD because the heavy metal content in it can be either vaporized or immobilized, and the dioxin/furan content can be thermally destroyed (Gonzalez-Corrochano et al.,

2012). Only if the hazardous EAFD is transformed into a nonhazardous form, will it be safe to put the recycled products into use in the construction sector. However, in order to make EAFD a proper raw material for lightweight aggregates, the compositions of EAFD need to be modified to meet the following criteria: 48–70% SiO<sub>2</sub>, 8–25% Al<sub>2</sub>O<sub>3</sub>, and 3–12% flux, as previously proposed (Riley, 1951; Huang and Wang, 2013; Tuan et al., 2013). The flux includes iron oxides, alkaline oxides, and alkaline earth oxides (CaO, MgO, Na<sub>2</sub>O, FeO, and Fe<sub>2</sub>O<sub>3</sub>) (Riley, 1951; Liao et al., 2013; Tuan et al., 2013).

Because an electric arc furnace smelter mainly uses waste iron-containing other items as raw materials, the EAFD discharged from it would be rich in flux components, such as iron oxides (Pickles, 2008). It can be expected to contain little SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>. In contrast, harbor sediment is always rich in SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> (Wei et al., 2011). Thus, if EAFD and harbor sediment are blended together at proper ratios, the mixes would be able to meet the compositional criteria for successful manufacturing of lightweight aggregates.

Practical application of lightweight aggregates is extensive. They can be used in civil construction, horticulture, and environmental engineering (Gonzalez-Corrochano et al., 2009). When used in civil engineering, lightweight aggregates can be a marvelous substitute with a much lower density to ease the amount of steel used for the construction sector without degrading the strength of constructed objects, such as bridges, tall buildings, roads, and others. In the area of horticulture, water can be preserved inside the lightweight aggregates that are usually more porous, as compared with marbles. As for applications in environmental engineering, lightweight aggregates can be used as a substrate for bacteria

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to grow in wastewater treatment units (Dordio and Carvalho, 2013).

Lightweight aggregates acceptable in practical application are generally defined as aggregates having a particle density of  $<2.0 \text{ g cm}^{-3}$  and a 24-h water absorption rate of approximately  $<18\%$  (Cheeseman and Viridi, 2005). Usually, aggregates low in particle density and water absorption rate are desired in the aspects of lessening the weight of constructed objects and enhancing the workability with cement (Posi et al., 2013). Also, it is not difficult to manufacture lightweight aggregates with a density of  $<1.0 \text{ g cm}^{-3}$  and a 24-h water absorption rate of  $<5\%$ . As a comparison, natural marble has a particle density of ca.  $2.8 \text{ g cm}^{-3}$  and a relatively low 24-h water absorption rate,  $<5\%$ . Although lightweight aggregates often have a higher water absorption rate than natural marble, they can be manipulated during their preparation process to become more water-resistant if a glassy phase is formed on their surface. The glassy surface is a result of sufficient sintering of raw materials containing proper compositions. For example, waste glass rich in silica and sodium oxides is beneficial for forming a water-impermeable glassy surface at a lower temperature than a typical process temperature for preparing lightweight aggregates (Tsai et al., 2006). Also, richness in flux components is capable of improving the sintering reaction and helping the formation of glassy surface. However, formation of a glassy surface alone will not make lightweight aggregates; suitable gas-releasing components are simultaneously required to bloat the heated, low-viscosity samples (Anagnostopoulos and Stivanakis, 2009).

Many compounds are capable of releasing gases at elevated process temperatures, such as carbonates, sulfates, oxides, and hydroxides of metals, and others (Bhatty and Redit, 1989; Gonzalez-Corrochano et al., 2009). As long as these gas pressures can be captured inside and overcome the viscosity of the glassy phases, pellet expansion would take place to form lightweight aggregates. The gases may include  $\text{CO}$ ,  $\text{CO}_2$ ,  $\text{SO}_2$ ,  $\text{SO}_3$ ,  $\text{O}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{Cl}_2$ , and others (Bhatty and Redit, 1989; Gonzalez-Corrochano et al., 2009).

## 2. Experimental methods

The experimental flowchart is presented in Fig. 1 and details of the experimental procedures are described below.

### 2.1. Compositional analysis

Harbor sediment used in the present study was collected from the regularly dredged Taichung Harbor. It is one of the busiest harbors of Taiwan and its dredging practice is carried out each year for ship navigation safety. As to EAFD, it was collected from a Taiwanese steel-making plant. Both the as-received sediment and EAFD were dried at  $105^\circ\text{C}$  for 24 h, ground to pass a 50-mesh sieve ( $<297 \mu\text{m}$ ), stored in plastic bottles, and ready for compositional analysis and further experiments. Compositional analyses, except Si,  $\text{Cl}^-$ , and  $\text{SO}_4^{2-}$ , for sediment and EAFD were performed with an inductively coupled plasma-atomic emission spectrometer (ICP-AES, Optima 3000DV, Perkin Elmer, USA) after a microwave-assisted (Microwave oven, MWS-2, Berghof Laborprodukte GmbH, Germany) acidic digestion procedure. Si contents in sediment and EAFD were determined based on the sample weight loss after their reaction at  $550^\circ\text{C}$  with a mixed acid (concentrated HF + concentrated  $\text{HClO}_4$ ) to form  $\text{H}_2\text{SiF}_6$  which is volatile (Taiwan CNS, 1985). To determine the content of  $\text{SO}_4^{2-}$  and  $\text{Cl}^-$  in both materials, a 0.100-g powder sample was placed in a capped Teflon vessel together with a mixed acid (10-mL concentrated  $\text{HNO}_3$  + 3-mL concentrated HF + 1-mL  $\text{H}_2\text{O}_2$ ), digested at  $180^\circ\text{C}$  for 10 min with microwave irradiation, filtrated, and the content of  $\text{SO}_4^{2-}$  and  $\text{Cl}^-$

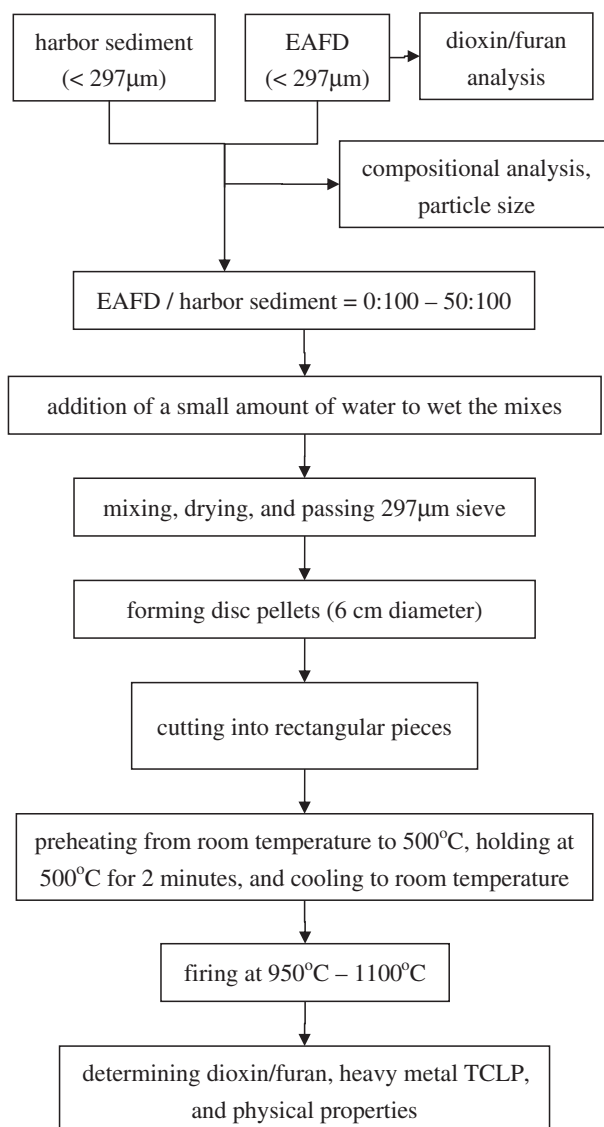


Fig. 1. Experimental flowchart for preparing lightweight aggregates.

in the liquid phase were determined with an ion chromatograph (IC, DX-100, Dionex, USA). The calibration curves for  $\text{SO}_4^{2-}$  and  $\text{Cl}^-$  span  $0\text{--}10.00 \text{ mg L}^{-1}$  and  $0\text{--}5.00 \text{ mg L}^{-1}$ , respectively, with the square of both correlation coefficients being  $>0.999$ . The particle size distribution of both sediment and EAFD were measured with a laser diffraction particle analyzer (Coulter LS230, Beckman Coulter, Germany).

### 2.2. Preparation of lightweight aggregates

The sieved EAFD and sediment were end-to-end mixed at a speed of 30 rpm for 6 h at various ratios (EAFD/sediment = 0:100, 10:100, 20:100, 30:100, 40:100, 50:100). For better mixing, a suitable amount of de-ionized water was added to wet the mixes prior to starting the mixer. After mixing, the mixes were again dried and ground to  $<50$  mesh. Then, fifty grams of each mix was formed into a disc-like green pellet having a diameter of 6 cm. The disc pellets were manually cut into small rectangular pellets (approximately  $2.5 \text{ cm} \times 1.75 \text{ cm}$ ) and fired at  $950\text{--}1100^\circ\text{C}$  for 18 min in an attempt to prepare lightweight aggregates after a de-volatilizing pre-treatment at  $500^\circ\text{C}$  (i.e., heating the rectangular pellets from room temperature to  $500^\circ\text{C}$  at a heating rate of  $100^\circ\text{C min}^{-1}$ ,

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