Waste Management xxx (2018) xxx–xxx

Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

Change in re-use value of incinerated sewage sludge ash due to chemical extraction of phosphorus

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A R T I C L E   I N F O

Article history:
Received 23 August 2017
Revised 27 November 2017
Accepted 3 January 2018
Available online xxxx

Keywords:
Incinerated sewage sludge ash
Characterization
Chemical extraction
Phosphorus
Heavy metals
Pozzolanic activity

A B S T R A C T

The potential of six different extractants to recover phosphorus (P) from incinerated sewage sludge ash (ISSA) was evaluated. Secondary effects such as the co-dissolution of Zn and Cu were also considered. The residual ISSA from each study was assessed in particular detail, focusing on the leachability of remaining Zn and Cu, major element composition, crystalline phases and overall degree of crystallinity and particle size distribution. The residual ISSA was also evaluated as a pozzolanic material using a Strength Activity Index (SAI) test with mortars containing Portland cement with a 20% substitution by ISSA. All results were compared to tests with untreated ISSA. Overall, the use of 3 of the 6 extractants could be ruled out due to poor P recovery potential and/or a serious compromise of the potential reuse of residual ISSA in Portland cement-based materials. The results highlight the added value of considering the potential reuse of residual ISSA when trying to optimize P recovery from ISSA by wet methods.

1. Introduction

Sludge-to-energy incineration technology is increasingly becoming the preferred choice for sludge management all over the world, especially in Europe and Hong Kong, China (Samolada and Zabaniotou, 2014; Zhao et al., 2016). The world's largest sewage sludge incineration facility to date, capable of processing 2000 tonnes/day sewage sludge dry solids, was commissioned in 2015 in Hong Kong (T-Park). However, in the incineration process, a considerable amount of sewage sludge ash (ISSA), amounting to about 10% of the dewatered sewage sludge mass, is generated (Donatello and Cheeseman, 2013; Li and Poon, 2017). It is estimated that at least 1.7 million tonnes of ISSA are produced annually worldwide and is likely to increase in the future (Donatello and Cheeseman, 2013). In Hong Kong in particular, if no reuse option is identified, landfill disposal of ISSA will impose a heavy burden on waste management infrastructure. Potentially high concentrations of soluble salts and heavy metals in ISSA, especially Zn and Cu, add to concerns about landfill disposal (Franz, 2008; Xu et al., 2012; Nowak et al., 2013; Lynn et al., 2015).

To tackle this environmental problem, countries worldwide are exploring suitable ways for recycling and beneficial resource recovery of ISSA. Previous researches mainly focused on the use of ISSA as raw materials for manufacturing cement, concrete, bricks and tiles due to the fact that the major elements present (SiO_2 and Al_2O_3) are similar to the clays and pozzolanic materials used in these products (Kosior-Kazberuk, 2011; Ottosen et al., 2013; Donatello and Cheeseman, 2013; Baeza et al., 2014). Some studies have already found that mortars and concrete containing ISSA as binders could exhibit good mechanical properties after a longer curing age (Chang et al., 2010). According to the research of Fontes et al. (2004), 30% of cement replacement by ISSA did not lower the 28 days compressive strength of the mortars. The research results by Baeza et al. (2014) showed that significant pozzolanic reactions occurred with ISSA at a level comparable to fly ash, which increased with curing age and ISSA addition.

Nevertheless, it has also been reported that the application of ISSA as a supplementary cementitious material in construction product is hindered by the physical and chemical properties of ISSA. Data on the effect of using ISSA as a binder on 28 day compressive strength of mortars or concrete showed a reduction with increasing ISSA content, on an average at the rate of 1% for 1% ISSA replacement of Portland cement (Lynn et al., 2015). One possible

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https://doi.org/10.1016/j.wasman.2018.01.007
0956-053X/

reason for poor performance is the presence of heavy metals and salts in ISSA and their effect on Portland cement hydration. Images from SEM analysis reveal that ISSA particles have an irregular morphology, with large pores forming in grains where organic matter has combusted rapidly and where particles are held together in loose agglomerates by either partial sintering or the rapid precipitation of inorganic compounds. The presence of such particles in cement mortars causes an increase in water demand for achieving a given workability. The negative effect of ISSA on strengths can be partially or completely overcome by grinding ISSA (Pan et al., 2003), by the use of superplasticizer (Coutand et al., 2006) or by adding nano-materials (Lin et al., 2008a).

However, all of the above mentioned applications represent a loss of the valuable phosphate content in ISSA, which is characterized by typically by high P_2O_5 content of about 8.9–35.9%, comparable to that of a low grade phosphate ore (Xu et al., 2012). The ISSA has a significant potential to be used as a secondary source of phosphate for the production of fertilizers and phosphoric acid. Many papers have been published in recent years about the recovery of P from ISSA (Herzel et al., 2016; Havukainen et al., 2016; Li et al., 2017). Approaches to P recovery include electrodialytic process (Guedes et al., 2014), thermochemical treatment (Havukainen et al., 2016) and chemical extraction (Ottosen et al., 2013). Among these, chemical extraction is the most widely used method due to its simple process and low cost (Donatello and Cheeseman, 2013; Shiba and Ntuli, 2016). Various extractants have been investigated for their P extraction potential, including inorganic acids, organic acids, alkalis and chelating agents, however, the study on the effects of these extractants on the leaching characteristics of P and trace elements in ISSA is scarce. Previous studies indicated that inorganic acids (especially sulfuric acid) could achieve a better P extraction efficiency (more than 90%) (Petzet et al., 2011; Xu et al., 2012), but also revealed that this led to the leaching of high concentrations of heavy metals from the ISSA (Donatello and Cheeseman, 2013). Therefore, P purification is another inevitable problem to be resolved (Franz, 2008; Biswas et al., 2009; Petzet et al., 2012).

Pressure on economical P reserves and continued increases in P demand will increasingly promote P recovery from ISSA. However, P recovery by chemical extraction will create a residual fraction which may simply be sent to landfill. As with the original ISSA, possible reuse applications for the P-extracted ISSA should be investigated in order to prevent its disposal to landfill, but research on this area is fairly scarce.

It is known from previous studies that the extractants employed for P and heavy metals extraction would affect oxide minerals (especially for the Fe/Mn/Al oxides) and subsequently change the mobility of P and heavy metals (Lee et al., 2016; Beiyuan et al., 2017). Chemical extraction of P not only changes the elemental composition of ISSA, but also the particle size distribution, microstructure of particles and pozzolanic activity. All extraction methods tend to render a significant increase in ISSA specific surface area due to the removal of soluble compounds, such as $CaCO_3$ and alkali metal chlorides, to the bulk solution (Wang et al., 2009; Fedje et al., 2010). According to Donatello et al., (2010a), in terms of mortar strength, acid washing and milling had a net positive effect with ISSA-the opposite of what was observed when experiments were repeated with coal fly ash (FA), metakaolin (MK) and a sand reference material. However, no consistent effect has been reported in the literature about the effect of P recovery by different extractants on the pozzolannic activity of ISSA. As reported by Feng et al. (2004), the pozzolanic activity of rice husk ash pretreated by hydrochloric was not only stabilized, but also enhanced. Both Donatello et al., (2010a) and Ottosen et al. (2013) found that a high amount of gypsum crystals were formed after extraction in H_2SO_4. The importance of gypsum content on early Portland cement hydration chemistry (and thus strength) should not be underestimated (Skapa, 2009). A better understanding of the effect of extractants used for P recovery on the properties of residual ISSA is needed before it can be determined if this is something that can be optimized together with the P extraction efficiency.

Therefore, this work studied the extraction efficiency of total P, Zn and Cu, and the residual ISSA samples were evaluated for the leachability, particle size distribution, chemical composition and pozzolanic activity. The morphology of ISSA before and after chemical extraction was characterized by scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDX) and crystalline phases were assessed by X-ray diffraction (XRD). The aim of this research is to identify potential use of residual ISSA (treated or untreated) as a pozzolanic material in cement mortar/concrete.

2. Materials and methods

2.1. Sample collection

A total of about 500 kg ISSA samples were obtained from the bottom of multicyclone in the sewage sludge incinerator over a period of 2 h for this research. In the incineration process, dewatered sewage sludge is fed to a fluidised bed system with a combustion zone temperature maintained at 850 °C in excess oxygen. About 10 kg of the as-received ISSA samples were coned and quartered until around 1000 g of material remained which were subsequently oven dried at 105 °C.

The physical properties and chemical compositions of ISSA are presented in Table 1. All the measurements were carried out in triplicate, and the average values with standard deviations less than 5% were reported. The ISSA used in the tests had a specific gravity (water pycnometer method) of 2.49, pH (1:5 dry sample: DI Water) of 8.45, and loss on ignition (1000 °C after 120 min) of 0.99%. XRF data (using a Rigaku Supermini200 type X-ray fluorescence (XRF) spectrometer) showed that the major elements in ISSA are Si, Al, Fe, Ca and P, followed by Ti, K, S, Zn, Cl and Cu.

To measure the total heavy metals concentrations in the ISSA, the aqua-regia digestion technique was adapted from EN 13657 (2002). In descending order, the concentrations of the heavy metals in ISSA were: Zn (2198 mg/kg), Cu (621 mg/kg), Ba (170 mg/kg), Cr (137 mg/kg), As (107 mg/kg), Ni (85 mg/kg), Pb (70 mg/kg), Cd (4.5 mg/kg) and Hg (0.1 mg/kg), which were then used in further calculations for extraction rate. However, results achieved by aqua-regia digestion method was generally lower than the XRF method, and the same results were found by Hoffmann et al. (2010) who attributed this to poor calibration of XRF method.

2.2. Chemical extraction of P from ISSA

Samples of ISSA were mixed with one of six different chemical agents (two organic acids, two inorganic acids and two chelating agents) at a liquid-to-solid ratio of 10 L kg^{-1} and 30 rpm in an

Table 1
Physical and chemical analysis of ISSA.

Items		Value
Physical properties	Specific gravity	2.49
	pH	8.45
	Mean particle size (μm)	30.0
	Loss on ignition (%)	0.99
Chemical composition (%)	Na_2O: 2.42; MgO: 1.66; Al_2O_3: 11.88; SiO_2: 31.15; P_2O_5: 9.27; Cl: 0.24; SO_3: 4.08; K_2O: 3.53; CaO: 9.73; TiO_2: 0.61; Cr_2O_3: 0.07; MnO: 0.14; Fe_2O_3: 23.95; CuO: 0.24; ZnO: 0.89	

Please cite this article in press as: Li, J.-s., et al. Change in re-use value of incinerated sewage sludge ash due to chemical extraction of phosphorus. Waste Management (2018), https://doi.org/10.1016/j.wasman.2018.01.007