



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



Jiang-shan Li^{a,b}, Zhen Chen^a, Qi-ming Wang^a, Le Fang^a, Qiang Xue^b, C.R. Cheeseman^c, Shane Donatello^d, Lei Liu^b, Chi Sun Poon^{a,*}

^a Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong

^b State Key Laboratory of Geomechanics and Geotechnical Engineering, Institute of Rock and Soil Mechanics, Chinese Academy of Sciences, Wuhan 430071, China

^c Department of Civil and Environmental Engineering, Imperial College London, London, United Kingdom

^d Cements and Materials Recycling Department, Eduardo Torroja Institute of Construction Sciences, Consejo Superior de Investigaciones Científicas (CSIC), c/ Serrano Galvache, 4, Madrid, Spain

ARTICLE INFO

Article history:

Received 23 August 2017

Revised 27 November 2017

Accepted 3 January 2018

Available online 5 January 2018

Keywords:

Incinerated sewage sludge ash

Characterization

Chemical extraction

Phosphorus

Heavy metals

Pozzolanic activity

ABSTRACT

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.

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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 (SiO₂ and Al₂O₃) 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

* Corresponding author.

E-mail address: cecspon@polyu.edu.hk (C.S. Poon).

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 electro-dialytic process (Guedes et al., 2014), thermochemical treatment (Havukainen et al., 2016) and chemical extraction (Ottofen 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 pozzolanic activity of ISSA. As reported by Feng et al. (2004), the pozzolanic activity of rice husk ash pretreated by hydrochloric acid was not only stabilized, but also enhanced. Both Donatello et al., (2010a) and Ottofen 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 ₂ O: 2.42; MgO: 1.66; Al ₂ O ₃ : 11.88;	
	SiO ₂ : 31.15; P ₂ O ₅ : 9.27; Cl: 0.24; SO ₃ :	
	4.08; K ₂ O: 3.53; CaO: 9.73; TiO ₂ : 0.61;	
	Cr ₂ O ₃ : 0.07; MnO: 0.14; Fe ₂ O ₃ : 23.95;	
	CuO: 0.24; ZnO: 0.89	

end-over-end mechanical rotator for 2 h. According to previous studies, these conditions seemed optimal for P extraction (Fang et al., 2017). Details of the six experiments are summarized in Table 2. Extractant concentrations were selected based on previous literatures (Kleemann et al., 2016; Wang et al., 2016; Donatello et al., 2010b; Biswas et al., 2009), which had been demonstrated to be effective and should be more practical and relevant to field applications.

After mixing, the slurry was centrifuged at 4000 rpm for 10 min and filtered via 0.45- μm mixed cellulose esters membrane filters. The filtrate was then digested by the addition of concentrated HNO_3 at a volume ratio of 5:2 (sample: HNO_3) followed by ICP-OES (SPECTROBLUE) analysis.

The extracted P was measured colorimetrically based on the reaction with ammonium molybdate and development of the 'Molybdenum Blue' colour. The absorbance of the compound was measured at 882 nm in a spectrophotometer and is directly proportional to the amount of P extracted from the ISSA. The retained solids were oven dried at 105 °C and, due to the agglomeration of particles, were dry-ground for 2 min before using for further analysis.

2.3. Test methods

2.3.1. Leachability of heavy metals in residual ISSA

The leachability of residual heavy metals in the residual ISSA that remains after P extraction was examined by the toxicity characterization leaching procedure (TCLP, US EPA, 1992), which is often used as a performance indicator or acceptance criteria for soil remediation. The residual ISSA was mixed with 0.1 M acetic acid, which was adjusted to 2.88 ± 0.05 by 1 M NaOH, at a liquid-to-solid ratio of 20 L kg^{-1} and rotated at 30 rpm in an end-over-end rotator for 18 h.

2.3.2. Particle size distribution

The size distribution and specific surface area of the different ISSA samples were measured using a Malvern MS3000 laser diffraction particle size analyser in wet mode using distilled water as a dispersing agent.

2.3.3. Pozzolanic activity test

The pozzolanic activity of ISSA samples was assessed by the strength activity index (SAI) test based on BS 3892 (1997). The control mortar was prepared with 100% ordinary Portland cement (CEM-I) at a w/b ratio of 0.5 and a sand/binder ratio of 3.

The test mortars were prepared with ISSA replacing 20% of CEM-I (instead of the 30% specified in BS 3892). As per BS 3892, the w/b ratios were altered so that all the mixes had the same workability according to EN1015-3 (1999). The amount of water required for the untreated ISSA mixture was 1.15 times of that in the control mortar. For mortars containing the different residual ISSAs, the same w/b ratio was used as the untreated ISSA to avoid the results being influenced by changes in water content. In any

case, the workability of these mortars was similar. The mortar was mixed in a planetary orbital mixer for 5 min then cast into 50 mm cubic moulds with the aid of a vibrating table. The specimens were demoulded after 24 h and cured in a water bath for 6 and 27 days before being subjected to compressive strength tests (model Testometric CXM 500-50KN universal testing machine, loading rate of 0.3 mm min^{-1}). Strength results are reported as the average of three specimens and the SAI was calculated as the ratio of compressive strength of the ISSA containing mortars to the strength of the pure cement mortars at the same age.

2.3.4. Mineralogy and microstructure of untreated and residual ISSA

The crystalline-phase mineralogy of the ball milled ISSA samples was evaluated by using a high resolution powdered X-ray diffractometer (Rigaku SmartLab, Japan) using $\text{Cu K}\alpha$ ($\lambda = 1.54059 \text{ \AA}$) radiation obtained at 40 kV and 30 mA. Scans were taken between 5 and 80° 2 θ with 0.02° step sizes. The peaks were identified by the Rigaku's software (PDXL) using a pattern data base ICDD PDF 2010 (International Centre for Diffraction Data, Powder Diffraction File 2010). After identification of the crystalline phases, the degree of crystallinity was further analyzed by the whole powder pattern fitting (WPPF) method.

The surface characteristics of gold-coated ISSA samples were examined using a Tescan-vega3 scanning electron microscope with energy dispersive X-ray spectroscopy (SEM-EDX) to analyze the changes in shape, size, and morphology.

3. Results and discussion

3.1. Effectiveness of chemical extraction

This study mainly focused on the extraction of P and the two heavy metals present in highest concentrations (Zn and Cu). From Fig. 1 it is clear that most of the P (>70%) was leached within 2 h of extraction by acids and the acid types had only a little effect on P extraction. The fast P extraction rate may be attributed to the acid soluble nature of the ISSA as well as the small particle size (Biswas et al., 2009). This conclusion was also been well-supported by Hong et al. (2005) who reported that the P would be present in the ISSA in the form of highly soluble salts. Due to a similar pH from 0.85 to 1.70 for the four of the six extractants, the extracted P concentration only changed marginally, which was consistent with the previous finding that the extracted P reached a plateau level at a pH of less than 1.8 (Franz, 2008). Sulphuric acid obtained better results compared to nitric acid at the same concentration probably due to the double concentration of H^+ ions (Fang et al., 2017). The unextracted P may be physically occluded from direct contact with the acidic aqueous phase if present in the central matrix of ISSA particles (Hong et al., 2005).

With the other two extractants (the chelants), P extraction efficiency was less than 30%, some two thirds lower than the acid extractants (see Fig. 1). This may be because chelants have a high affinity to metal ions, resulting in partial dissolution of P. More-

Table 2
Experimental design of chemical extraction of ISSA.

Extractants	Chemical formula	Concentration (mol/L)	pH		
			Before extraction	After extraction	
Inorganic acids	Nitric acid	HNO_3	0.5	0.85	0.97
	Sulfuric acid	H_2SO_4	0.5	0.89	1.14
Organic acids	Citric acid (CA)	$\text{C}_6\text{H}_8\text{O}_7$	0.5	1.70	2.55
	Oxalic acid (OA)	$\text{C}_2\text{H}_2\text{O}_4$	0.5	1.01	1.03
Chelants	EDTA	$\text{C}_{10}\text{H}_{16}\text{N}_2\text{O}_8$	0.05	4.72	5.46
	EDTMP	$\text{C}_6\text{H}_{20}\text{N}_2\text{O}_{12}\text{P}_4$	0.05	1.32	2.53

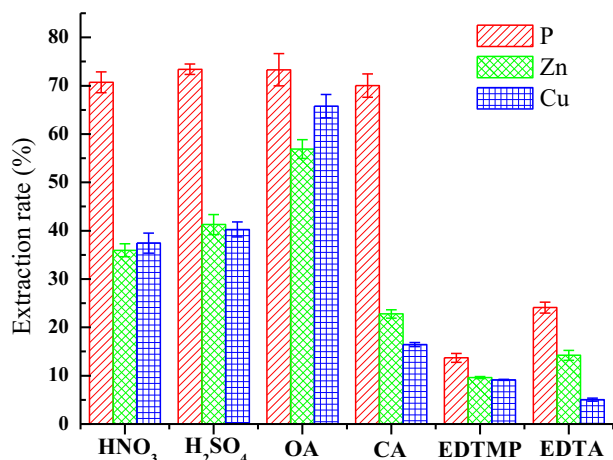


Fig. 1. Extraction rate of P, Zn and Cu from ISSA.

over, the results show the optimal P extraction agent was sulfuric acid, which had been also extensively used in other research due to its comparatively low price (Donatello et al., 2010a).

It is quite well known that the heavy metals and P in the ISSA are co-dissolved during acid extraction (Biswas et al., 2009). Fig. 1 shows that the solubility of Zn and Cu in the ISSA was much lower than that of P which indicates that P is present in the ISSA as an acid soluble form while the solubility of the Zn and Cu compounds is relatively lower. The relatively low leaching rate of Zn and Cu was also due to the fact that parts of metals were immobilized in acid insoluble glassy phase and present as nucleation particles in the central matrix of the ISSA (Stark et al., 2006; Biswas et al., 2009).

The extraction efficiencies of Zn and Cu cannot be explained by pH alone. Oxalic acid achieved the highest extraction rate of Zn (56.89%) and Cu (65.77%) but was not as acidic as HNO₃ or H₂SO₄. EDTA (pH 4.7) had low extraction efficiency of Zn but not as low as EDTMP (pH 1.0). It was clear that oxalic acid has a higher complexation and dissolution capacity of Zn and Cu. Even though results for EDTA were low, it is clear that it preferentially dissolves Zn instead of Cu. Generally speaking, the findings were consistent with the report by Wang et al. (2016), who revealed inorganic acids were the most efficient extractants for metal extraction because acid washing facilitated the dissolution of acid-extractable fraction (metals) and minerals (which primarily hosts the metals) in the ISSA. Contrasting results obtained from various extractants, even under the similar initial pH, can be attributed to the difference in the complexing ability and the final pH of the extraction solutions after reaction (Li and Poon, 2017).

3.2. Chemical compositions of ISSA before and after extraction

As known from previous studies, P usually exists in the form of whitlockite (Ca₃(PO₄)₂), aluminum calcium phosphate (Ca₉Al(PO₄)₇), hydroxyapatite (Ca₅(PO₄)₃(OH)), aluminum phosphate (AlPO₄), and iron phosphate (FePO₄), derived from the reaction of the phosphate in the sewage and the flocculant agent during incineration (Ohbuchi et al., 2008; Donatello et al., 2010b).

Results of XRF analysis of ISSA before and after P extraction are presented in Table 3. Different trends were noted for the five major elements present in untreated ISSA (i.e. SiO₂, Al₂O₃, CaO, Fe₂O₃ and P₂O₅) after extraction.

It was clear that SiO₂ and Fe₂O₃ remained to a significant degree in the residual ISSA regardless of the extractant used and degree of P extraction achieved. The low solubility of iron oxide in ISSA under acidic conditions has previously been observed (Stark

et al., 2006). For SiO₂, which was decreased after treated by H₂SO₄, but increased in the residual ISSA treated by other five extractants, this may be due to the significantly higher solubility of quartz in H₂SO₄ than other extractants.

The levels of CaO in residual ISSA decreased when the levels of P extraction increased but with three important exceptions. In the case of H₂SO₄, the CaO concentrations remained stable and with OA, CaO concentrations even increased in the residual ISSA. These effects can be attributed to the formation of gypsum and calcium oxalate precipitates, which is supported by XRD evidence presented later. The other exception was EDTMP, where even though P₂O₅ and CaO were dissolved, levels of P₂O₅ in residual ISSA actually increased due to P already present in the extractant. This latter observation rules out EDTMP as a single extractant for P recovery from ISSA.

With Al₂O₃, each of the acidic extractants caused a notable decrease in the residual ISSA, suggesting that pH was the dominant influence rather than P recovery. ISSA treated with H₂SO₄ had substantially lower levels of Al₂O₃ than the others. Perhaps this can be attributed to the fact that all acids had the same molar concentration but that H₂SO₄ is diprotic, thus contributing twice as many H⁺ ions than HNO₃ at a given concentration. Therefore, although the initial and final pH of HNO₃ is close to H₂SO₄ (see Table 2) due to the gradual ionization process of H₂SO₄, which also resulted in the higher acidity and solubility of H₂SO₄ in the extraction process.

Even though results were quite low (<3%), the XRF data revealed that residual ISSA consistently had lower contents of Cl, Na₂O and, with the exception of H₂SO₄ treated ISSA, of SO₃. This trend suggests a high dissolution of salts present in ISSA. Levels of CuO and ZnO in residual ISSA were generally lower although it is difficult to draw conclusions from XRF data when all results are <1% when expressed as oxides.

3.3. Leachability of heavy metals in residual ISSA

The original ISSA showed low leachability according to the TCLP (see Fig. 2). Extractable Zn (11.72 mg/L) and Cu (2.11 mg/L) concentrations were well below the 100 mg/L Chinese national regulatory levels (CN-GB, 2007) – indicating that the waste can be classified as a non-hazardous material. To put the results in context, if all the Zn and Cu measured in untreated ISSA had leached (i.e. 2198 and 621 mg/kg respectively), this would equate to TCLP results of 109.9 mg/L Zn and 31.1 mg/L Cu. Less than 10% of the Cu + Zn leached under TCLP conditions – suggesting that most of the metals exist in phrases that are only soluble in more acidic solutions (pH < 3) or are immobilized in glassy matrices.

The results in Fig. 2 also show the leachability of any Zn and Cu that remains in the residual ISSA after treatment with extractants. Due to the fact that significant amounts of Zn and Cu were removed by treatment with the extractants, it would be logical to presume that the leachable Zn and Cu in residual ISSA would be less than that in the untreated ISSA.

However, this was not always the case. Treatment with H₂SO₄ and OA substantially increased the leachable levels of Zn in residual ISSA and treatment with H₂SO₄, HNO₃, OA and CA did the same for Cu. In these cases, the extractant is not only co-dissolving a certain fraction of Zn and Cu with P, but is also altering the non-soluble fractions of Zn and Cu in such a way that it increases their leachability under the conditions of the TCLP test. The different metal leachability of ISSA before and after chemical extraction may be attributed to the different destabilization of the metals by these extractants.

One possibility is that a certain amount of Zn and Cu dissolved by the extractant is weakly held on ISSA particle surfaces via electrostatic interactions (Tsang and Hartley, 2014). Such adsorbed Zn

Table 3
Chemical compositions of ISSA (%).

Compositions	Original	H ₂ SO ₄	HNO ₃	OA	CA	EDTMP	EDTA
Na ₂ O	2.42	0.34	1.75	0.57	0.81	0.98	1.69
MgO	1.66	1.98	1.13	0.63	1.29	1.14	1.24
Al ₂ O ₃	11.88	4.90	8.91	7.88	8.68	12.22	12.14
SiO ₂	31.15	26.60	41.02	42.54	43.29	31.16	36.26
P ₂ O ₅	9.27	1.35	1.83	1.53	5.29	18.04	8.22
SO ₃	4.08	25.96	1.15	0.59	0.32	0.64	0.48
Cl	0.24	0.02	0.05	0.03	0.06	0.05	0.05
K ₂ O	3.53	2.55	3.80	3.55	3.76	3.31	3.89
CaO	9.73	9.34	1.78	15.48	5.46	5.61	7.32
TiO ₂	0.61	0.62	0.89	0.96	0.75	0.65	0.65
Cr ₂ O ₃	0.07	0.09	0.09	0.07	0.07	0.08	0.08
MnO	0.14	0.11	0.13	0.07	0.13	0.10	0.14
Fe ₂ O ₃	23.95	25.07	36.02	25.11	29.09	24.94	26.69
CuO	0.24	0.17	0.20	0.12	0.17	0.21	0.22
ZnO	0.89	0.76	1.05	0.58	0.64	0.71	0.78

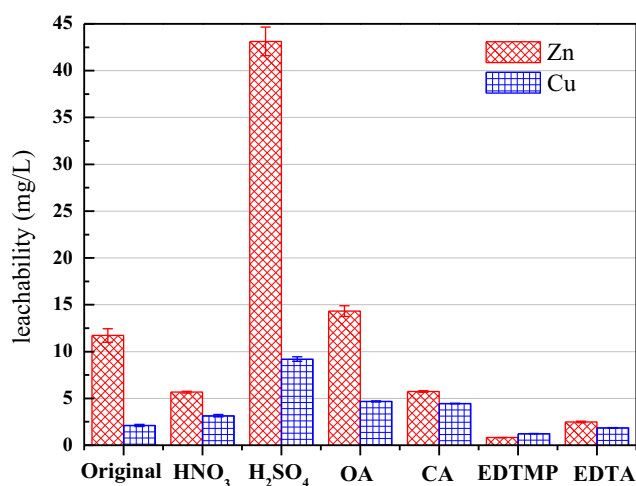


Fig. 2. TCLP leachability of Zn and Cu from untreated (original) and residual ISSA.

and Cu would be easily leachable, leading to an undesirable increase in environmental concern.

According to Fig. 2, the extractant which causes the greatest concern in this respect was H₂SO₄. The EDTA and EDTMP chelants did not follow this type of behavior, arguably because any dissolved Zn and Cu would be chelated and thus less available to interact via electrostatic forces with ISSA particle surfaces. This agreed well with previous findings that EDTA significantly decreased the TCLP leachability of residual Cu, Zn and Cd (Pociecha and Lestan, 2009; Udovic and Lestan, 2009). Considering these results, prior to any landfill disposal or reuse, it is recommended that residual ISSA left after P extraction should be tested for metal leachability.

3.4. Changes in morphology due to extraction

Fig. 3 shows the SEM micrographs of ISSA before and after chemical extraction. It can be seen that the particles of ISSA were very irregular with many plate-like silicon oxides and rough with open pores (Figs. 3a and S1 in supplementary material). After extraction by inorganic acids (e.g. H₂SO₄ in Fig. 3b), the ISSA particles and silicon oxides were broken down to smaller granules. This can be attributed to the removal of soluble salts which helped to hold together some of the particles and the action of hydraulic flushing action caused by an end-over-end vibration (Bruckard et al., 2005). In addition, the heavy metals in smaller granules of the extracted ISSA can react more effectively with the leaching agents.

According to the Figs. 3b and S2 in supplementary material, the formation of secondary compound (rod-like CaSO₄) on the particle surfaces of H₂SO₄-extracted ISSA was shown, which indicated the interaction between H₂SO₄ and CaPO₄ in the ISSA (Donatello et al., 2010a). As shown in Fig. 3c, the organic acids (e.g. OA)-extracted ISSA particles presented amorphous shapes and particle agglomeration with looser structure and smoother surface. The silicon oxides were precisely identified, but the fact that calcium oxalate crystals were hard to find might be due to only very small crystals forming throughout the ISSA particles. Compared to the SEM images of the untreated ISSA (Fig. 3a), chelating agent extraction seemed to have only a marginal effect on the micro-structure and mineral phases of ISSA particles (e.g. EDTMP in Fig. 3d). Despite the clearly elevated P content in the EDTMP residue, the P containing minerals were not identified due to the amorphous structure.

From the EDX results in Table 4 on the entire SEM images it can be seen that O, Si, Fe, Ca, Al and C were mainly present over the particle surfaces of ISSA. P was not seen in the acid-extracted ISSA, indicating the high extraction efficacy of the acid agents, which supported the results of Section 3.1. After chemical extraction by H₂SO₄, the Si and Al in ISSA was significantly decreased, while Fe marginally decreased, suggesting P extraction was mainly from Ca-P and Al-P. The main elements in H₂SO₄-extracted ISSA surface were O, S and Ca, no doubt linked to precipitation of CaSO₄. The C, O and Ca were mainly presented in the oxalic acid-extracted ISSA, implying the precipitation of calcium oxalate. The EDTMP extraction posed little effect on the distribution of the elements of ISSA, especially for Fe, indicating non-reactivity of Fe containing phases in ISSA, in agreement with XRF results presented earlier. But the P content was highest, thereby hindering its application as a supplementary cementitious material for mixing with Portland cement to produce composite cement (Maghsoodloorad et al. 2014).

3.5. Particle size distribution

The particle size distributions of untreated ISSA and residual ISSA after P extraction are illustrated quantitatively in Table 5 together with results of specific surface area analysis.

The data in Table 4 clearly shows that chemi-mechanical treatment with all extractants, with the notable exception of EDTMP, caused the proportion of fine particles to increase and thus also the specific surface area of the residual ISSA. The slightly decrease in specific surface area of EDTMP treated ISSA may be attributed to the low dissolution and high complexation of EDTMP, this is in agreement with the SEM results presented earlier. The extraction procedure also resulted in the agglomeration of particles, which resulted in a substantial increase in the particle fractions >100

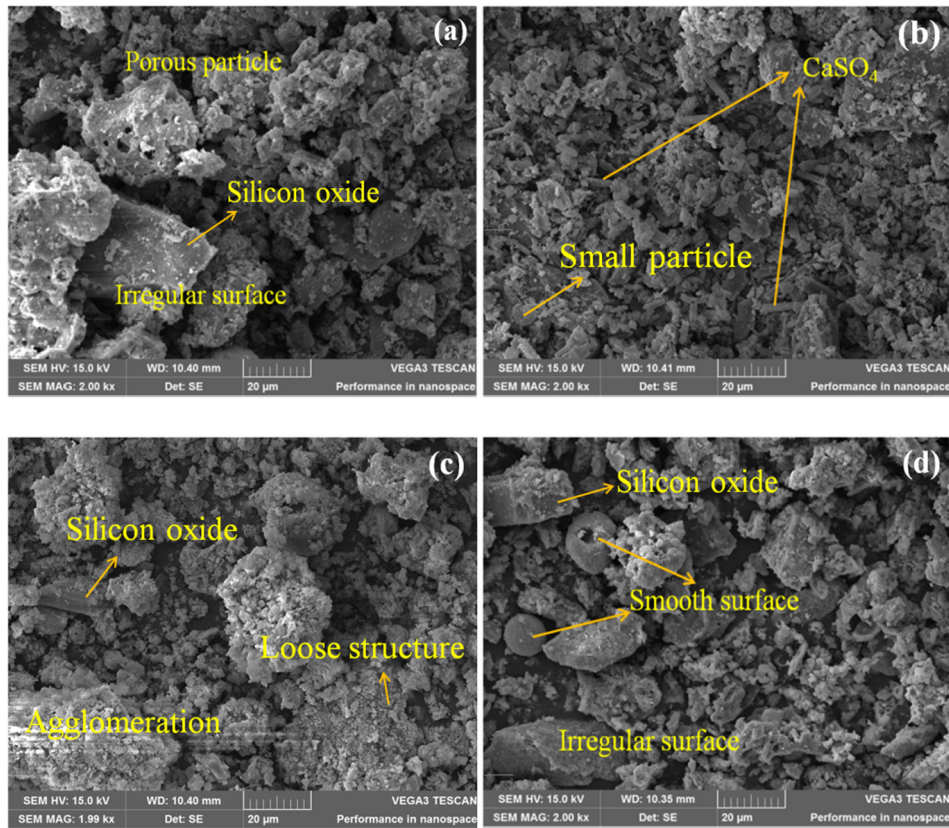


Fig. 3. SEM-EDX images of ISSA samples (magnification 1000 times) (a) original ISSA, (b) H₂SO₄, (c) Oxalic acid and (d) EDTMP.

Table 4
Chemical compositions of ISSA by EDX results.

Elements	Original Weight %	H ₂ SO ₄	OA	EDTMP
C	7.79	10.89	19.51	14.73
O	37.68	41.60	44.26	36.69
Na	3.87	0.82	0.00	2.12
Mg	1.24	0.00	0.68	1.00
Al	5.82	2.47	2.19	6.15
Si	15.09	7.99	8.51	11.82
S	0.00	8.58	0.00	0.00
P	0.81	0.00	0.00	4.14
K	1.76	1.14	0.79	1.37
Ca	5.81	10.42	12.27	3.32
Fe	18.91	16.10	10.70	18.44
Cu	0.45	0.00	0.31	0.21
Zn	0.76	0.00	0.78	0.00

Table 5
Particle size distribution and specific surface area of untreated and residual ISSA.

Extractant	<10 μm %	10–50 μm	50–100 μm	>100 μm	Specific surface area (m ² /kg)
Untreated	12.74	77.10	10.16	0.00	379.6
HNO ₃	20.98	52.00	19.14	7.88	522.4
H ₂ SO ₄	19.32	47.50	17.77	15.41	483.0
OA	19.66	48.98	18.60	12.76	526.8
CA	15.91	49.98	21.90	12.21	429.3
EDTMP	11.09	56.43	28.87	3.61	322.6
EDTA	17.09	59.60	22.02	1.29	444.6

μm . The agglomeration effect was much more pronounced for acid extractants than chelants.

The difference in particle size distribution of residual ISSA must be taken into account if it is intended to reuse the residual ISSA in construction materials due to potential effects on workability and strength (Pan et al., 2003; Lin et al., 2008b).

3.6. Changes in mineralogy due to extraction

Mineral analysis was undertaken by XRD and the results are presented in Fig. 4, where the untreated ISSA is shown to be poorly crystallized. The degree of crystallinity was 40.22%, which was similar to the result reported by Anderson (2002). Treatment with acid extractants substantially increased the degree of crystallinity in residual ISSA (OA-87.67%; CA-85.33% and H_2SO_4 -76.12%) with the exception of HNO_3 (to 41.40%). The chelant-based extractants actually decreased the degree of crystallinity in the residual ISSA (EDTMP-25.31% and EDTA -33.29%).

The main crystalline components of untreated ISSA could be clearly identified as hematite (Fe_2O_3), quartz (SiO_2), whitlockite ($\text{Ca}_9(\text{MgFe})(\text{PO}_4)_6\text{PO}_3\text{OH}$) and anorthite ($\text{CaAl}_2\text{Si}_2\text{O}_8$), plus a minor presence of berlinite (AlPO_4) and giniite ($\text{Fe}^{2+}\text{Fe}^{3+}(\text{PO}_4)_4(\text{OH})_2 \cdot 2(\text{H}_2\text{O})$). Apparently, whitlockite was dissolved by both organic acid and inorganic acids (but not by chelating agents), especially for HNO_3 , H_2SO_4 and OA, while berlinite and giniite were not significantly removed, which verified the results in XRF. In addition, the intensity of hematite and quartz was hardly changed or even strengthened in the ISSA after extraction (also reflected by the XRF result), indicating that Si and Fe in the ISSA was quite stable.

The precipitation of secondary compound crystals during extracting with chemical agents may increase the specific surface area of the extracted ISSA (Fedje et al., 2010). As shown in Fig. 4, it is likely that bassanite (CaSO_4) was formed in the H_2SO_4 -extracted ISSA and calcium oxalate (CaC_2O_4) was identified on the particle surfaces of oxalic acid treated ISSA. This result is in accordance with the SEM-EDX data in Section 3.4 and XRD data presented both here and by Ottosen et al. (2013).

The formation of bassanite must be taken into account when using the extracted ISSA in Portland cement-based materials because CaSO_4 would influence the early hydration chemistry of Portland cement, especially the Calcium Aluminate phases, and may have an influence on setting time and early strength development (Skapa, 2009).

3.7. Pozzolanic activity

The results of the SAI tests are given in Fig. 5. The control mortar strength was 37.2 MPa at 7 days and 46.9 MPa at 28 days. According to BS 3892 (1997), SAI results greater than 0.8 after 28 days are indicative of a positive pozzolanic activity. For correctness, it is pointed out that BS 3892 is intended to be used only for coal fly ash and that the substitution rate stipulated was 30% whereas in this study we use 20%.

It has been reported that the pozzolanic activity of ISSA is lower than that of some common pozzolans (Pan et al., 2003; Lin et al., 2008b). The results in Fig. 5 show that only the untreated ISSA can be regarded as a potentially useful pozzolanic material in blended cements. The best results for residual ISSA samples barely exceeded a SAI of 0.70 and were not as promising as those presented by Donatello et al. (2010a) after washing with H_2SO_4 . The SAI results for residual ISSA treated with CA or EDTMP were so low as to completely invalidate their potential reuse in Portland cement-based materials. Analysis of crushed mortars by DTA (see Fig. 6) shows that almost no C-S-H or $\text{Ca}(\text{OH})_2$ was found in the extremely low SAI mortars, which confirms that the low strength

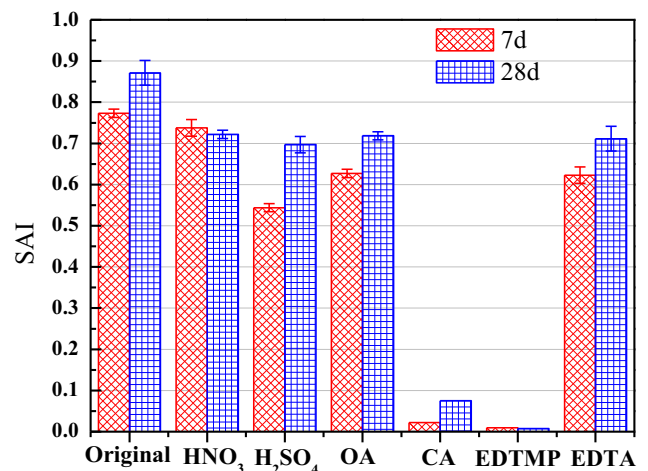


Fig. 5. Strength activity index of samples after 7 and 28 d.

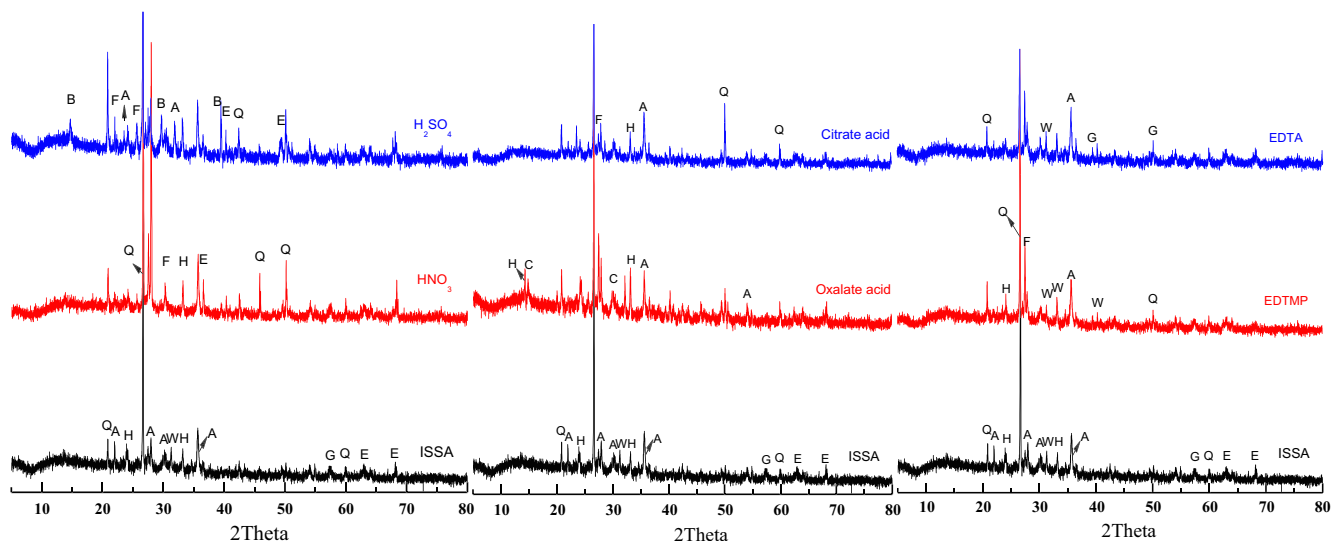


Fig. 4. XRD patterns of ISSA samples before and after chemical extraction (Q: Quartz; A: Anorthite; H: Hematite; B: Bassanite; C: Calcium oxalate; W: Whitlockite; E: Berlinite; G: Giniite).

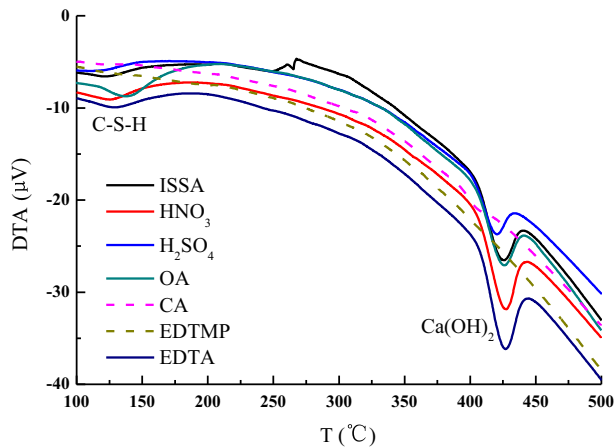


Fig. 6. DTA curves of mortar samples containing ISSA after curing for 28 d.

was caused by inhibited cement hydration. For the EDTMP extracted ISSA, the large amounts of phosphorus (see Table 3) may be partly responsible for the significant decrease in SAI (Takahashi et al., 2001; Maghsodloorad and Allahverdi, 2017). But the reason for the citric acid extracted ISSA needs to be further studied.

It is well known that the degree of pozzolanic activity of a material correlates with the contents of reactive amorphous silicate or aluminosilicate phases. One possible reason why the SAI of the residual ISSA decreased could be due to the loss of amorphous material, as evidenced by the degree of crystallinity measurements with XRD analysis. However, when assessing pozzolanic activity by a proxy measure such as strength development, other factors that influence strength, such as porosity, water content and hydration reaction kinetics should be considered. Donatello et al., (2010c) found that the pozzolanic activity of a highly water absorbing material (e.g. ISSA) might be underestimated using the SAI test if the flowability is not adequately compensated by superplasticizer and/or if extra water is added during mixing. However, noting from Fig. 5 that the SAI indices of ISSA and extracted ISSA (EDTA, H₂SO₄ and oxalic acid) marginally increased with time, it can be concluded that these samples can be potentially used as supplementary cementitious materials albeit with a low pozzolanic activity.

4. Conclusions

This work aimed to investigate the potential of different extractants on P recovery from ISSA and also on secondary effects such as the fate of heavy metals (from the perspective of co-dissolution with P and from the leachability in residual ISSA). The fate of other major elements and the potential use of residual ISSA in blended Portland cement mortars as a pozzolanic supplementary cementitious material were investigated. The following conclusions can be drawn from this study:

- (1) Acid extractants, both inorganic and organic, were more effective than chelating agents for the recovery of P but also extracted more heavy metals. This was attributed to the acid soluble nature of the ISSA. The optimal P extraction agent was sulfuric acid with a concentration of 0.5 mol/L (74% of total P recycled), but the optimal extractant in terms of selectivity for P and not heavy metals was CA with a concentration of 0.5 mol/L (72% of total P dissolved but less than 21% of total Zn and Cu). Other acid extractants achieved similar P recovery but higher co-dissolution of Zn and Cu.

- (2) The XRF results indicated that Fe and Si did not co-dissolve with P from and that Ca solubility was high, but that Ca would precipitate if the extractant contributed anions that would form insoluble Ca-precipitates (i.e. SO₄²⁻ from H₂SO₄ to form basanite and C₂O₄²⁻ from oxalic acid to form calcium oxalate).
- (3) Even though Zn and Cu was leached during P recovery, the Zn and Cu that remained in the residual ISSA was actually more soluble than the forms of Zn and Cu present in the untreated ISSA. This could be due to the possible adsorption of Zn and Cu on residual ISSA particles by weak electrostatic forces during P recovery. This effect was only noted with the acid extractants and not the chelants.
- (4) The chemi-mechanical action of the P recovery process generally caused ISSA particles to break into smaller granules (<10 μm) and increase the specific surface area. However, agglomeration of these finer particles and, in some cases precipitation of new phases, caused the fraction of larger particles (>100 μm) to increase substantially. Unlike acid extractants, chelating agents had little effect on the morphology and particle size distribution of ISSA.
- (5) According to the SAI tests, the pozzolanic activity of untreated ISSA was low to start with (0.75–0.85) but decreased further with residual ISSA after P extraction (0.55–0.75). This could be partly due to the loss of the reactive amorphous phases from ISSA (from degree of crystallinity measurements). The use of citric acid and EDTMP to extract P resulted in abnormally low SAI values (<0.10) due to a strong inhibition of Portland cement hydration. Therefore, more attentions and studies should be conducted for proper management of the residual ISSA. In short, sulfuric acid can be used as the optimal extractant from the perspective of comparatively low price and maximization recovery of ISSA (including P and residue).

Acknowledgements

The authors would like to thank the financial support of the Hong Kong Research Grants Council (PolyU 152132/14E), the Hong Kong Scholars Program (XJ2016035), the National Natural Science Foundation of China (41602315), the Youth Innovation Promotion Association CAS (2017376) and the Open Research Fund of State Key Laboratory of Geomechanics and Geotechnical Engineering, Institute of Rock and Soil Mechanics, Chinese Academy of Sciences (NO. 2015003).

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at <https://doi.org/10.1016/j.wasman.2018.01.007>.

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