REVIEW ARTICLE

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Characteristics of incineration ash for sustainable treatment and reutilization

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Abstract

Municipal solid waste incineration (MSWI) generates bottom ash, fly ash (FA), and air pollution control (APC) residues as byproducts. FA and APC residues are considered hazardous due to the presence of soluble salts and a high concentration of heavy metals, and they should be appropriately treated before disposal. Physicochemical characterization using inductively coupled plasma mass spectroscopy (ICP-MS), X-ray diffraction (XRD), and X-ray fluorescence (XRF) have shown that FA and APC have potential for reuse after treatment as these contain CaO, SiO₂, and Al₂O₃. Studies conducted on treatment of FA and APC are categorized into three groups: (i) separation processes, (ii) solidification/stabilization (S/S) processes, and (iii) thermal processes. Separation processes such as washing, leaching, and electrochemical treatment improve the quality and homogeneity of the ash. S/S processes such as chemical stabilization, accelerate carbonation, and cement solidification modify hazardous species into less toxic constituents. Thermal processes such as sintering, vitrification, and melting are effective at reducing volume and producing a more stable product. In this review paper, the treatment processes are analyzed in relation to ash characteristics. Issues concerning mixing FA and APC residues before treatment, true treatment costs, and challenges are also discussed to provide further insights on the implications and possibilities of utilizing FA and APC as secondary materials.

Keywords Municipal solid waste · Incineration · Fly ash · Air pollution control residues · Characterization · Treatment processes

Introduction

Municipal solid waste incineration (MSWI) is a common waste management technique in many countries due to its

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capacity for high volume reduction and possibility of energy recovery (Kothari et al. 2010; Leckner 2015). However, the resulting bottom ashes and flue gas contain heavy metals and other hazardous pollutants, and controlling their emission is necessary. Typically, regulatory limits on incinerator emissions are strict, leading to many developments in air pollution control systems (Liu et al. 2015).

With the development of efficient air pollution control systems, the focus has now shifted from emission control to the treatment and disposal of fly ash (FA) and air pollution control (APC) residues collected by these systems. The fate of FA and APC residues depends principally on national policies. In most developed countries, treatment and reuse of these residues is possible because of environmental policies and possibly subsidies (Huang and Chuieh 2015), while in other countries, these ash residues are buried in landfills (Sun et al. 2016).

Despite the relatively high concentration of heavy metals, salts, and organic recalcitrant, which may limit utilization possibilities, the best strategy is still treatment followed by utilization as secondary resources. Proper characterization has to be conducted before treatment as the properties of these residues may differ greatly depending on the source of the municipal solid waste (MSW) (Quina et al. 2008a, 2008b; Ahmaruzzaman 2010; Fruergaard et al. 2010). Although extraction procedures are used to provide heavy metal speciation and leachability from metal-bearing ash particles, there are many arguments about their suitability in characterization of metal phases. Therefore, comprehensive characterization by analytical techniques has been implemented to reveal both qualitative and quantitative characterizations of FA and APC residues.

In a modern MSWI plant, there is the combustion chamber where the waste is burnt for volume reduction, the boiler system to recover energy, and the air pollution control system to reduce emissions. The air pollution control system removes fine particulates (with devices such as the electrostatic precipitator, fabric filter, or cyclone) and regulates harmful gases (with devices such as dry lime scrubbing or wet lime scrubbing).

It should be noted that different residues are generated at various stages in the MSWI plant (Fig. 1). In this review, only fly ash and APC residues collected at the air pollution control systems are considered. The definition of fly ash by the International Ash Working Group is "the particulate matter carried over from the combustion chamber and removed from the flue gas stream prior to addition of any type of sorbent material" (Chandler et al. 1997). Therefore, in this review, the ashes collected prior to the scrubbing process are classified as fly ash, while those generated during the scrubbing process are termed as air pollution control residues. In some incineration plants, there was no prior removal of the FA before the scrubbing process, and both FA and APC residues were collected together, so it is considered as a form of APC in this review paper by definition. It is important to classify the FA and APC residues properly as these have different properties, and inappropriate use of terms could cause misinterpretation (Quina et al. 2008a, 2008b). In addition,

available research based on mixed APC and FA is not taken into account due to the large variance in the reported data.

Due to the presence of a substantial amount of chlorine in MSW, the utilization of FA and APC residues is difficult due to the accumulation of metal chlorides (Joseph et al. 2018). Despite the difficulties, many countries with land constraints have put in effort to reutilize FA and APC residues. With a combination of well-developed standards, strong enforcement of environmental regulations, and government support, most FA and APC residues can be reused as secondary raw materials in cement industry, in road pavement and soil amendment, and as a sludge-conditioning agent (Ahmaruzzaman 2010).

In case utilization standards are unavailable, countries have avoided the use of FA and APC residues but have instead enforced the treatment of these FA and APC residues before disposal into landfills. In less-developed countries without strict enforcement or disposal regulations, the FA and APC residues are usually mixed with bottom ash for dilution before disposal into a landfill in order to save cost.

The best management practice of FA and APC residues is no doubt reutilization followed by treatment before disposal. These materials should never be disposed without treatment. In addition, as MSWI is becoming a prevailing method for waste management, the amount of MSWI FA and APC residues is expected to increase. Therefore, there is a need for proper treatment so that these materials can be utilized as a resource instead of being wasted.

In this review paper, different treatment processes for FA and APC residues are evaluated in order to reutilize these ashes safely and economically. Due to the importance of proper characterization, this paper also reviewed the common characterization methods used for FA and APC. In addition, the paper provides new insights with regard to mixing FA and APC, true treatment cost, and treatment challenges.





Physicochemical characterization of FA and APC residues

The physicochemical properties of FA and APC residues are affected by factors such as MSW feedstock, incinerator design, and type of air pollution control units (Song et al. 2004; Moon et al. 2016). Therefore, characterization is important in order to determine the best strategy for subsequent ash management (Quina et al. 2008a, 2008b).

Physical properties such as particle size distribution, permeability, moisture content, and density generally affect heavy metal leaching and determine the environmental impacts after disposal (Fig. 2). The particle size distribution which is estimated by sieving is one of the most important physical properties as ash with smaller particle size tends to retain a higher concentration of heavy metals (Wan et al. 2018; Shi and Kan 2009) due to the larger surface area for condensation of volatile heavy metals. Other than dry sieving, the laser diffraction size analyzer has also been used to measure the size of particles in FA and APC residues (Aubert et al. 2006). Density is another important physical property of FA as this affects the uniformity and porosity of the ash particles (Chandler et al. 1997).

FA is generally very heterogeneous in nature. The total elemental composition via acid digestion followed by inductively coupled plasma mass spectroscopy (ICP-MS) is one of the most documented methods used to identify the amount of each element in the FA (Chang et al. 2009). In addition, other than elemental composition, the mineralogy and types of oxides affect the efficacy of treatment methods. Mineralogy and oxide phases have been studied using techniques such as X-ray diffraction (XRD) and X-ray fluorescence (XRF) (Wan et al. 2006; Haiying et al. 2010).

Other than inorganic elements, municipal solid waste incineration ash also contains toxic organic pollutants. Chlorinated organic compounds such as chlorobenzene, polychlorinated biphenyls (PCBs), polychlorinated dibenzodioxins (PCDDs), and polychlorinated dibenzofurans (PCDFs) have been detected (Tabata et al. 2013). The presence of various forms of organic and inorganic chlorides results in the formation of PCDDs and PCDFs during the cooling process of the flue gas at around 450 °C (Kawabata et al. 2003). The toxicity of different PCDDs and PCDFs varies, and it is evaluated by their toxicity equivalence quantity (TEQ).

Particle size comparison

The particle size of FA and APC residues is in the micron scale. Size is influenced by the MSW feedstock and location that the particulates are collected. The average size in terms of % volume passing D_{50} of FA and APC is between 10–166.41 µm and 0.5–48.71 µm, respectively (Table 1). Other information such as the D_{10} and D_{90} indicating the smallest 10% and largest 10% fraction can also be used to illustrate the size distribution. Figure 3 shows the typical particle size distribution of FA and APC. FA is, in general, larger in size than APC and has a much larger spread in terms of size variation due to the difference in the point of collection and nature heterogeneity.

The particle size of FA is largely dependent on operating conditions of the incinerator such as flue gas velocity and temperature in the heat recovery system (boiler). In addition, among the different types of FA, the more downstream the point of collection is, the smaller the particle sizes are, as the larger FA particles are captured by the upstream systems (boiler, economizer).

The main constituent of APC is the lime additive added to neutralize the acidic flue gases. Therefore, the variations in particle size between different studies are due to the different types of scrubbing processes employed. In general, APC particles are smaller than their FA counterpart as most of the larger particulate matters are already being collected prior to the scrubbing process.

Comparison of elemental composition

The elemental composition of FA and APC is often studied by digesting the samples and followed by ICP-MS and/or inductively coupled plasma optical emission spectroscopy (ICP-OES). The outcome is classified as major elements (> 10,000 mg/kg), minor elements (< 10,000 mg/kg and > 1000 mg/kg), and trace elements (< 1000 mg/kg).

Fig. 2 Common analytical methods for FA and APC



Ash type	Volume (%) particle size passi	ing	Reference
	D_{10}	D_{50}	D_{90}	
FA (ESP)	0.2	55	300	(De Boom and Degrez 2012)
FA	2.15	10.02	31.03	(Li et al. 2015)
FA	5	30.34	77	(Mangialardi 2001)
FA (cyclone)	12.5	40	72	(Karkfeldt and Steenari 2007)
FA	13.32	70.72	256	(Florea et al. 2016)
FA (ESP)	104	166.41	332.08	(Lin et al. 2018)
APC (semi-dry)	0.15	0.5	50	(De Boom and Degrez 2012)
APC	5.2	35.1	159.8	(Li et al. 2018)
APC (mixed)	7	15.08	80	(Jin et al. 2016)
APC	10.76	36.31	87.34	(Florea et al. 2016)
APC (mixed)	17.51	48.71	146.1	(Li et al. 2018)

In Table 2, it can be seen that APC residues comprise mainly Ca, Cl, Si, and Al, while other major elements include K, Na, and S. The minor elements are usually Ba, Cu, Mn, Zn, Pb, Fe, and Mg. Trace elements include Hg, Ni, Cr, Ag, and As. Despite the trace elements are being a relatively small component, they have been studied extensively due to their high toxicity.

Although FA also consists of Ca, Cl, Si, and Al, these are in very different proportions compared to APC residues (Table 3). APC contains much higher Ca and Cl content, while FA has higher Si and Al. For air pollution control (scrubber unit), lime is added to the gas stream to neutralize the acidic gases such as HCl to form CaCl₂. Another difference is that Mg, Ti, and Fe are considered major elements in FA, but minor in APC. This difference is due to the condensation or solidification of the less volatile metals and metal chlorides onto the FA during heat recovery where temperature is much lower than that in the combustion chamber. The amount of trace elements is relatively consistent in both APC and FA in most studies except for Hg. Since Hg has a relatively low boiling point, it exists in the gaseous state in the flue gas until finally captured in the APC unit.

Both APC and FA also contain a significant amount of Zn and Pb, which can easily leach out and harm the environment (if not managed properly). However, the total elemental composition is not fully representative of the actual leaching potential of each heavy metal due to the mineral speciation and other factors. Therefore, other characterizations such as XRF and XRD can provide more insights to the ecotoxicity of both APC and FA.

X-ray fluorescence

XRF is used to determine the elemental oxide composition in FA and APC residues, especially in preparation for ceramic and concrete applications. Although there are variations in the

oxide composition reported by different studies, there is agreement that both FA and APC residues have high alkali content in the form of CaO.

In Table 4, it can be seen that APC contains a large amount of CaO (as high as 58.65%) and other oxides such as SiO₂, Al₂O₃, Na₂O, K₂O, and SO₃. High alkalinity is detected due to the calcium hydroxide added in excess to remove the acidic flue gases. Consequently, the pH and acid-neutralizing capacity (ANC) are usually very high. With pH around 12, APC residues can cause problems such as leaching of amphoteric metals (Zn and Pb), while the high ANC results in difficulty in lowering the pH to reduce leaching effect.

On the other hand, although FA contains a significant amount of alkaline oxides such as CaO, it has also a high amount of acidic SiO₂ (Table 4). Because of that, there are differences in the chemical properties between FA and APC residues in terms of pH and ANC. Normally, FA has enough alkaline material to establish an alkaline pH of around 10 to 11 in water, but with low ANC. In some extreme cases that the alkalinity is not enough, FA results in a neutral or even slightly acidic pH.

Figure 4 presents a ternary diagram of CaO, Al_2O_3 , and SiO₂ for FA and APC as compared to ordinary Portland cement (OPC) and coal fly ash which are materials commonly used for construction. APC has composition very close to that of limestone and OPC since the major component is the lime additive, while FA also consists of relatively high Ca content. Some of the APC residues are, in fact, a mixture of FA and scrubbing residues due to the lack of prior filtering process; therefore, some of the APC samples tend to exhibit compositions relatively similar to FA.

X-ray powder diffraction

The mineral phases in FA and APC have also been studied using XRD. XRD analysis measures the intensity of different





crystal peaks from each unique compound to estimate relative amounts within the sample. However, both FA and APC contain many different phases, which can cause overlapping and difficulties in identifying the phases. This problem is usually overcome by the use of software programs to simplify the matching process.

Due to the nature of FA and APC, XRD only provides information on the different types of minerals found without proper quantification (Tian et al. 2018). In Table 5, it can be seen that the main minerals in APC are CaClOH, NaCl, KCl, CaSO₄, CaCO₃, and Ca(OH)₂, while those in FA consisted of many different heterogeneous phases such as calcium aluminosilicates, SiO₂, and other more complex metallic compounds. In comparison, the composition of APC is more consistent than that of FA. A large proportion of APC is the reaction product from the acidic flue gas and the lime injected, while the content of FA consists of many different entrained metallic silicate particles carried over from the combustion chamber.

Treatment technologies

Due to the hazardous nature of FA and APC residues, treatment is required before their disposal into a landfill or reutilization. Currently, most of the treatment technologies are focused on reducing the environmental impacts, e.g., by separation to reduce the total pollutant concentration or stabilization to control the rate of leaching. The treatments are classified into three main groups: (i) separation processes, (ii) solidification/stabilization (S/S) processes, and (iii) thermal processes. Figure 5 presents the treatment process roadmap for FA and APC residues.

Depending on various factors such as disposal regulations, potential for utilization, and cost of treatment, different treatment strategies are adopted. Despite the variations in details, treatment typically comprises separation (e.g., washing) (Lam et al. 2010) to reduce the soluble fraction followed by either thermal treatment (Chou et al. 2009a, 2009b) or S/S (Mangialardi 2003; Zheng et al. 2011). A summary of such treatments has been compiled by Van der Sloot et al. (2001), while Sakai et al. (1996) have emphasized the importance of treatment cost.

In the assessment of the most appropriate treatment methods, both technical and economic factors are considered. Technical considerations include treatment outcomes and scaling up before proceeding to economic feasibility. The latter includes the operational cost and the value and amount of the final products generated. For instance, the costly thermal treatment usually generates products that can be utilized to reduce the overall cost, while S/S usually generates a larger volume of waste that has to be constantly monitored in a hazardous landfill and can increase the overall cost of treatment. Therefore, a complete analysis in terms of both technical and economic factors should be taken into consideration with the local context kept in sight.

Separation

Separation methods make use of the different properties in the ash components to separate them using water washing, solvent leaching, thermal evaporation, and electrodialysis (Table 6). The main objective of these methods is to improve the quality and homogeneity of the product for further treatment or utilization, but some methods can also extract valuable metals (De Boom and Degrez 2015; Nagib and Inoue 2000).

Washing processes

The washing process exploits the differences in solubility of salts in water. Water washing is commonly used as a pretreatment step prior to other treatment methods as the high salt content might be detrimental to the stability and quality of

 Table 2
 Elemental composition of APC residues

	FA (mg/kg) (Chandler	FA ESP (mg/kg) (Le Forestier and T ihoured 1008)	FA (mg/kg) (Eighmy et al.	FA (mg/kg) (Yan et al.	FA ESP (mg/kg) (De Boom and	FA (mg/kg) (Carignan	FA (mg/kg) (Loginova	FA grate fired (mg/kg) (Tang and Stannori 2015)	FA (mg/kg) (Jung et al.	Typical v of FA (m	'alues ıg/kg)	
	(1661 - 1997)		(0661	61107	105 m 2017)	U al. 2000)	VI 41. 2017)		(1007	Min	Лах 🤌	verage
Са	74,000–130,000	168,500	46,000	. 1	172,000–322,000	. 1	. 1	180,000	- 1	46,000	22,000 1	56,071
\mathbf{Si}	95,000–210,000	132,200	38,000	Ι	15,000-52,000	I	I	67,000	I	15,000 2	10,000 8	7,029
\mathbf{AI}	49,000–90,000	93,400	21,000	I	6100-36,000	I	I	36,000	34,127	6100 9	3,400 4	5,703
C	29,000-210,000	69,100	232,000	I	129,000–250,000	55,000	I	1	I	29,000 2	50,000 1	39,157
К	22,000–62,000	28,000	109,000	I	30,000-54,000	I	I	21,000	I	21,000	09,000 4	6,571
Na	15,000-57,000	24,900	84,000	I	56,000-84,000		I	26,000	I	15,000 8	4,000 4	9,557
Mg	11,000–19,000	18,100	1100	Ι	5400-10,000	I	I	18,000	I	1100 1	9,000 1	1,800
S	11,000-45,000	12,400	I	Ι	38,000-69,000	31,000	I	I	I	11,000 (9,000 3	4,400
\mathbf{Fe}	12,000–44,000	12,100	1600	Ι	4800 - 11,000	I	I	20,000	6682	1600 4	4,000 1	4,023
Ξ.	6800 - 14,000	10,600	6000	Ι	3400-1000	I	I	14,000	I	1000	4,000 7	971
Zn	9000-70,000	7339	104,000	3149	9500-10,000	1048	19,000	17,000	8347	1048	04,000 2	3,489
Ь	4800–9600	6200	Ι	I	2200-13,000	I	I	I	I	2200	3,000 7	160
\mathbf{Pb}	5300-26,000	2611	27,000	819	3720-4940	316	3500	3000	1878	316 2	7,000 7	189
Ba	330–3100	1482	Ι	1037	450-1270	96	1500	1	I	96	100 1	158
Sn	550-2000	863	5900	I	870-1260	106	630	1	106	106 5	900 1	365
Mn	800-1900	800	400	564	Ι	I	I	1100	I	400	900	27
Cu	600–3200	741	2220	427	650-730	37	1500	840	520	37	200 1	042
\mathbf{Sb}	260-1100	720	Ι	202	370-690	I	1500	I	81	81 1	500 6	15
\mathbf{C}	140–1100	549	495	541	170-450	26	290	450	61	26 1	100 3	88
\mathbf{Sr}	40-640	388	Ι	Ι	360-550	I	I	Ι	I	40 (40 3	96
Cd	50-450	166	1660	I	210-270	20	300	83	98	20	660 3	31
ïŻ	60–260	96	70	57.6	100 - 150	6	130	I	Ι	6	1 09	04
>	29–150	40	25	37.3	I	I	60	1	I	25	50 5	7
Мо	15-150	36	47	6.2	I	I	30	I	Ι	6.2	50 4	2
\mathbf{As}	37–320	28	096	28.1	Ι	2	60	I	4.9	5	60 1	80
Co	13-87	28	13	8.0	Ι	1	22	I	I	1 8	1 2	5
Hg	0.7 - 30	19	9.8	24.5	Ι	5	I	I	I	0.7	0 1	5
Ag	2.3-100	I	192	Ι	I	I	I	I	Ι	2.3	92 9	8
Se	0.4–31	I	17	4.7	I	I	15	I	81	0.4	11 2	5

Table 3Elemental composition of FA

t)		
FA (Auth et al. 2004	25:22 20.7 10.0 2.7 2.7 1.4 1.4 1.4 1.4 1.7 11.6 13.6	
FA (De De Boom et al.) 2014)	22.2 7.6 3.7 0.89 0.89 1.1 1.3 1.3 1.3 1.3 1.9 0.04	0.4
FA . (Haiying et al. 2007	35.8 20.5 3.2 3.7 4.0	
FA fluidized bed (Hu et al 2016)	20.46 31.7 5.87 5.87 2.87 2.72 2.56 3.98 	
FA (Florea et al. 2016)	30.21 11.13 5.29 5.12 1.49 1.49 1.28	
I FA (Hartmann et al. 2015)	20.23 10.93 14.02 2.34 0.84 15.46 6.79 	
APC (Hu et al. b) 2016)	34.72 4.12 0.85 1.04 8.11 8.11 8.11 5.45	
APC (Florea et al. 2016	35.93 0.78 0.19 0.19 0.32 - - - - - -	
APC mix (Huang et al. 2019)	60.27 8.31 4.54 2.23 2.77 2.99 0.66 3.43 3.43 - 102	70.1
APC mix (Jin et al. 2016)	38.58 10.22 4.18 - 8.07 6.02 5.26 4.95	
APC (Chimenos et al. 2005)	30.14 10.6 5.14 0.87 2.08 6.22 6.52 0.83 0.83 - 1.47	
APC (Quina et al. 2014)	41.9 14.1 14.1 1.0 3.7 3.2 3.2 	
APC (Heuss- Aßbichler et al. 2010)	58.65 6.01 1.72 2.29 2.05 0.33 0.21 0.33 0.58 0.33 0.58 0.06	
APC (del Valle- Zermeño et al. 2013)	38.74 6.19 3.64 0.75 1.26 6.57 5.34 5.34 7.1 1.06	10.1
Relative (%) in oxides	CaO SiO2 Al2O3 Mg2O Mg2O Mg2O Na2O P2O3 P2O5 7nO ZnO	

XRF of FA and APC residues

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the final product (Mangialardi 2003; Chimenos et al. 2005; Chen et al. 2017). In the literature, various conditions such as the liquid-to-solid (L/S) ratio and duration have been studied and it is found that water washing can remove up to 90-95% of the chlorides and the final product is improved in quality for further utilization (Chen et al. 2016). Mangialardi (2003) recommended an L/S ratio of 25 with a 15-min duration, while Wang et al. (2009) used only an L/S ratio of 2 with a 1-h duration to reduce the amount of effluent.

However, some researchers have noted that the washing effluent contains a high content of heavy metals (Pb and Zn) (Wang et al. 2001) and possibly high pH (pH 12). Chimenos et al. (2005) varied the L/S ratio of 1 to 10 using a two-step washing process and found that the optimal L/S ratio is around 3. They also found that the use of MgSO₄ to reduce the pH from 12.5 to 11 can reduce the leaching of Pb and Zn. Others have suggested chemical additives such as phosphate to reduce the solubility of heavy metals (Vavva et al. 2017; Ecke 2003; Shi and Kan 2009).

The extraction efficiency and leaching of undesirable metals are highly dependent on the pH of the ash, the L/S ratio, and the metal of concern. FA with pH lower than 11 and low alkalinity is suitable for water washing as a pretreatment step since the leaching of harmful amphoteric metals is limited, while for APC, the pH is too high and more precautions have to be taken due to the possibility of Pb, Zn, and Cr leaching into the effluent.

The selective recovery of heavy metals and salts based on particle size before washing has also been demonstrated (Funari et al. 2017); however, their recovery can only be beneficial of subsequent use. Another approach towards washing is the development of better agitation equipment (Ko et al. 2013) or recirculation of wastewater in various sequential washing steps to reduce the amount of wastewater generated (Chimenos et al. 2005).

Leaching

The leaching process aims to extract heavy metals from ashes via the use of an aggressive leaching agent. Both acidic and alkaline leaching agents for FA are proven to be effective in improving the extraction of heavy metals, while alkaline leaching agents are more suitable for APC residues because of the high acid-neutralizing capacity.

Nagib and Inoue (2000) applied NaOH and HCl leaching on FA at high temperature (90 °C) and long duration (up to 100 min) and found that it increases the extraction of Pb (45.7%) and Zn (29%), while Chiang et al. (2008) managed to extract 89% Cd, 60% Zn, and 32.5% Cu from APC by using a high L/S ratio of 40 and a duration of 2 h with 1 N HCl. In other studies, multiple leaching steps and different chelating agents have been deployed (Jiao et al. 2016), while Van der Bruggen et al. (1998) showed that Pb leaching could



Fig. 4 Ternary diagram comparison of FA and APC bulk composition

be enhanced by the use of EDTA and predicted the leaching by simulation using MINTEQ.

Other than physicochemical leaching, researchers have also explored the potential in making use of bacteria to enhance the leaching process of FA (Yang et al. 2009a; Fedje et al. 2010). Wang et al. (2009) used *Aspergillus niger* with 4% (w/v) FA in 100 mL sucrose to extract up to 96% Cd, 91% Mn, 73% Pb, 68% Zn, 35% Cr, and 30% Fe.

Despite the continuous growth in the use of bioleaching as an environmentally friendly option, chemical leaching is still more efficient and consistent in selective leaching and stabilization of heavy metals in FA and APC (Funari et al. 2017). Commercially available chemicals such as LIX860M-I and Cyanex 923 are found to have high selectivity and low extractability of other metals, while the extraction efficiency can be further enhanced with dilution in organic solvent such as kerosene (Tang and Steenari 2015).

Electrochemical processes

The electrochemical process applies a potential difference to separate and recover the heavy metals via redox reactions at the interface of the electrodes. The advantage of this method is that no additional chemicals are required and no residues are generated. Jensen et al. (2015) have investigated the potential of electrodialysis on three different types of fly ash. However, a major limitation is the low efficiency when a concentration of the metals becomes low. To improve the low efficiencies, Viader et al. (2017) have tried electrodialytic separation assisted by cationic membranes and achieved good removal efficiencies for Cr, Cu, Ni, Pb, and Zn. In another study, pre-treatment steps (water washing and sieving) were used to improve the electrodialytic process (Chen et al. 2018).

Jensen et al. (2010) worked on the electrochemical extraction of APC and showed that the conductivity of the suspension decreases from 52 mS/cm to between 1.7 and 5 mS/cm indicating the removal of most charged species, while Kirkelund et al. (2010) also proved that despite negligible change in the pH after treatment, most metals in the final product meet the requirement except for Pb and Zn. Important operating parameters include the current density, pH, distance between electrodes, and dosage of assisting agents.

In another study, ultrasound-enhanced electrokinetic remediation was applied for the removal of Zn, Pb, Cu, and Cd from FA. It was demonstrated the synergetic effects of acidification and ultrasonication for enhancing the remedial efficiencies of heavy metals in the further electrokinetic technology. It was also demonstrated that the combined system reduced the environmental toxicity for the FA to the maximum extent (Huang et al. 2018).

Thermal evaporation

Thermal evaporation is a separation process that makes use of high temperature to separate volatile metals from the bulk ash (Yu et al. 2012). The bulk ash is heated to a certain temperature to vaporize volatile metals (Pb and Cd), and these are then recovered through condensation. Hg, which has low boiling point, is not condensed but remains in the gas phase, while other less volatile metals remain in the solid phase.

Important operating factors reported include duration, temperature, and additives. Jakob et al. (1995) have reported that higher temperature and longer duration generally could improve the removal efficiencies for FA, but they could be decreased if the operating temperature exceeded the melting range of the target species. Due to this limitation on the operating temperature, the removal efficiencies for other less volatile metals (Zn and Cu) are lower, and the most effective temperature for evaporating Zn, Pb, Cu, and Cd is 1300 °C in argon atmosphere.

To overcome the high-temperature conditions, many researchers investigated the use of chlorine additives to induce a reducing condition in order to increase the removal of other less volatile trace metals. Nowak et al. (2012) investigated the use of various chlorides such as MgCl₂, CaCl₂, and NaCl and found that MgCl₂ was the best chloride additive for removal of up to 95% Pb and Cd at 1000 °C. The mechanism behind this was the formation of volatile metal chlorides, whereas another mechanism was that the presence of chlorides prevents the incorporation of heavy metal into the melted matrix. In general, this method is mostly carried out on FA due to the nature of the ash composition to form a more stable product for other utilization purpose.

Thermal evaporation process is well developed, and various lab-scale and commercial projects are available such as CT-Fluapur® (Jakob and Mörgeli 1999) and ASH DEC (Nowak et al. 2010). However, the treatment cost is still relatively high and has to be combined within the waste incinerator to reduce the cost.

Ash type	CaClOH	CaCl ₂	Calcite CaCO ₃	Lime CaO	Portlandite Ca(OH) ₂	Anhydrite CaSO ₄	Sylvite KCl	Halite NaCl	Quartz SiO ₂	Other minerals	Reference
FA			1		\checkmark	1	1	1	1		(Zhan et al. 2018)
FA			1			\checkmark	1	1	1	(Zn,Fe)S Zn_2SiO_4 KAISi O N2AISi O C2AI Si O	(Loginova et al. 2019)
FA			1	1		1	1	√		$\begin{array}{c} \text{Ca}_{2}\text{Al}(\text{Si},\text{Al})_{2}\text{O}_{7}\\ \text{Ca}_{2}\text{SiO}_{4}\\ \text{MgO}\\ 3\text{Ca}\text{O}\cdot\text{Al}_{2}\text{O}_{3} \end{array}$	(Hartmann et al. 2015)
FA	\checkmark		√			1	√	1		2 5	(Bontempi et al. 2010)
FA	\checkmark						√	1	1	KNO ₃ PbO ₂	(Liu et al. 2009)
FA	√	1			1	~		1		$MgAl_{2}(PO_{4})_{2}(OH)_{2}$ $Mg(CIO_{2})_{2}\cdot 6H_{2}O$ $Na_{2}H_{2}P_{2}O_{7}\cdot 6H_{2}O$ $CaAl_{2}Si_{2}O_{8}$ $AIO(OH)$ $FeAl_{2}(PO_{4})_{2}(OH)_{2}$	(Chou et al. 2009a, 2009b)
FA			1			\checkmark	\checkmark			SiCl	(Liu et al. 2018a, 2018b)
FA			\checkmark			\checkmark	1	\checkmark			(González et al. 2017)
APC	\checkmark		1		\checkmark	\checkmark	1	1	1		(Jin et al. 2016)
APC			1		1		\checkmark	1		K.F $e^{2}+2Al(Al_{2}Si_{2})O10(F,OH)$ C $a_{2}Al(OH)_{6.5}Cl_{0.5}\cdot3(H_{2}O)$ K(Mg,F e)3AlSi_{3}O_{10}(F,OH)_{2}	(Huang et al. 2019)
APC	\checkmark		\checkmark			\checkmark	\checkmark	\checkmark		SiCl ₄	(Li et al. 2018)
APC			\checkmark			1	1	1	1	$Ca_2Al_2SiO_7$ (AlSi_3O_8)Na, K	(Bertos et al. 2004)
APC	1				√			√		Ca(ClO) ₂ ·4H ₂ O Pb ₂ Sb ₂ O ₇ NaAsO ₂ CaTiSiO	(Eighmy et al. 1993)
APC	\checkmark		1	~			√		√	MgO KCaCl ₃	(Sun et al. 2008)
APC	\checkmark		√		\checkmark	\checkmark	1	\checkmark			(Baciocchi et al. 2009)
APC	\checkmark	1	\checkmark	~			1	\checkmark	1		(Deng et al. 2018)
APC			\checkmark			\checkmark	\checkmark	\checkmark	\checkmark	Fe ₂ O ₃	(Yu et al. 2015)
APC	\checkmark				1	\checkmark	1				(González et al. 2017)

Solidification and stabilization

S/S is a well-proven technology to immobilize heavy metals via chemical and physical modifications (Malviya and Chaudhary 2006). In general, solidification reduces the mobility of hazardous species by improving physical properties such as permeability and surface area, while

stabilization chemically modifies the hazardous species by converting them into less soluble or less toxic constituents. Currently, S/S is focused on process optimization to improve the efficiencies and economic viability (Galiano et al. 2011). S/S includes chemical stabilization, accelerated carbonation, and Portland cement solidification (Table 7).

Table 5XRD of FA and APC residues

Fig. 5 Treatment technologies for FA and APC residues



Chemical stabilization

Chemical stabilization makes use of chemical additives to produce thermodynamically favored solid phases in order to reduce the total availability for leaching. This treatment method consists of (i) dissolution of the waste residues, (ii) addition of chemicals for stabilization and pH adjustment, and (iii) filtration to separate the high salt filtrate from the stabilized residues.

The technology is quite established, and various additives such as soluble phosphates, sulfides, and activated carbon have been used at full scale (Nzihou and Sharrock 2002). Eighmy et al. (1997) demonstrated that 1.2 mol of H_3PO_4 per kg APC with an L/S ratio of 0.4 could reduce Pb leaching by 99.5%, while the pH of the final product was not affected by the H_3PO_4 added due to high alkalinity. Quina et al. (2010) suggested that H_3PO_4 could affect the neutralizing capacity since both Zn and Pb were stabilized, and there was increased leaching of Cr(VI). Recently, Wang et al. (2018) have utilized phosphate and other chelate agents to stabilize FA, while leaching effect was examined using citric acid.

Other additives such as ferrous sulfate, colloidal silica, Bayer red mud, and alumina oxides have also been investigated with good immobilizing potential (Hu 2005; Park 2009; Liu et al. 2018a). In general, APC is commonly treated via chemical stabilization due to the high alkalinity and high acidneutralizing capacity.

Accelerated carbonation

Carbonation is a natural weathering process due to the dissolution of CO_2 in water, which lowers the pH and eventually results in the formation of calcite and other new minerals capable of entrapping heavy metals. Ecke (2003) has reported decreased leaching of APC at 2 orders of magnitude for amphoteric metals such as Zn and Pb after carbonation but also highlighted increased release of other metals such as Cd, Cr, Ni, and Sb. Li et al. (2007) studied the effect of L/S ratio between 0.1 and 0.8 using accelerated carbonation and suggested the optimum L/S ratio is 0.3. The main factors affecting the rate of carbonation are diffusivity and reactivity of carbon dioxide, and these can be controlled by adjusting the pH, L/S ratio, humidity, temperature, and pressure (McCarthy et al. 2018).

Solidification and chemical fixation with cement binder

Solidification involves the mixing of FA or APC with a binder such as cement to form a more stable monolithic solid after the hydration reaction. The enhanced properties of the final product such as lower hydraulic conductivity and lower porosity limit the leachability of the heavy metals by reducing contact, while the increase in structural strength and durability ensures long-term stability and structural integrity (Bie et al. 2016). In addition, the formation of a crystalline structure during the pozzolanic reaction is capable of incorporating heavy metals into the matrix to provide potential long-term immobilization (Yakubu et al. 2018).

Although FA seems to delay the hydration process, the final strength is acceptable with high stabilization of Pb, Zn, Cr, Cd, Ni, and Hg. Aubert et al. (2007) pretreated the FA with a series of washing solutions and, afterwards, calcination at 600 °C, attaining pozzolanic effects which made it suitable to replace cement partially, while the formation of ettringite was able to entrap various heavy metals.

Solidification is generally carried out on pretreated FA to remove chlorides due to its pozzolanic properties, which enables it to partially replace cement, while APC is usually not

	initial you	separation inenious		
Separation	Residue type	Methodology	Results and findings	References
Washing	APC	Washing of APC with varying L/S ratios from 1 to 10 with the addition of MoSO.	Optimal L/S ratio of 3 with 2-step washing of 1-h duration each The use of MoSO . to lower the nH from 12 5 to 11 also reduces the leachshility of Ph and Zn	(Chimenos et al.
Washing	FA	L/S ratio of 25 with a 15-min washing duration	Removal of chlorides without releasing heavy metals Improvement in FA quality and reduction of retardation of setting time during stabilization	Mangialardi 2001; Mangialardi
Washing Washing	FA FA	L/S ratio of 2 stirred for 60 min at 30 r/min agitation L/S ratio from 2 to 100 stirred for 60 min at 30 r/min	Incaution of the wastewated at pr 0.5 to 7.5 streedive in removing include Extraction of 50.6% of K, 41.1% of Na, 5.2% of Ca, and 1% of Cr during the pretreatment Removal of 95% of CL, 50% of major elements (Na, K, and Ca), and 13% of Cr Worlded only arithtic increation to enclude achieves in and has been been and been and been achieved achieves and been been achieved achieves and been been achieved achieves and been achieves and been been achieved achieves and been achieves and been achieves and been achieves and been achieves achieves achieves and been achieves active	(Wang et al. 2009) (Wang et al. 2001)
Washing	FA	Wet sieving followed by washing using the L/S ratio of 2 for 3 times	Fine particles lower than 38 µm contain a higher amount of Pb and Zn. Washing reduces the amount of soluble salts (Pb and Zn contains a higher amount of Pb and Zn. Washing reduces the amount of washing appears potential in metal extraction from fine factions while reusing as cement material for the correst factions.	(Funari et al. 2017)
Leaching	APC	L/S ratio of 40 with a duration of 2 h bH was controlled from 3 to 7.5 using 1 N HCl	At pH 3 to 4, the extraction of Cd, Zn, and Cu was 89%, 60%, and 32.5%, respectively. The amount of bydroxide mecinitates that is recordable is about 1% of the APC mass	(Chiang et al. 2008)
Leaching	APC	L/S ratio of 20 extraction with LIX860M-I and Cyanex 923 diluted in kerosene	LIX860N-1 showed high selectivity for Zn with a small amount of Fe co-extracted. Cyanex 923 extracted Zn effectively, but Cd, Fe, and Pb were also co-extracted. The increase in the extractant in organic phase leads to the increased extraction of the selective metals but also increased the level of co-extracted of	(Tang and Steenari 2015)
Leaching	FA	Leaching using NaOH and HCl under different temperatures, durations, and concentrations	The use of NaOH is effective for extracting Pb (45.7%), but not so for Zn (29%). The use of HCl is more effective in extraction for Pb and Zn but other metals such as Fe, Mg, and Al. Higher temperature (up to 00° C) and house duration (up to 100° mod by the house of 100 min) also had to higher extraction.	(Nagib and Inoue 2000)
Leaching	FA	4% (<i>w/v</i>) FA in 100 mL sucrose with the addition of	The 2-step bioleaching extracted 96% Cd, 91% Mn, 73% Pb, 68% Zn, 35% Cr, and 30% Fe	(Wang et al. 2009)
Leaching	FA	csperguas musc Leaching with an L/S ratio of 10 using HCl from 0 to 1.8 M for 3 h with shaking	Leaching of Pb is enhanced by the use of a chelating agent such as EDTA. The effects of leaching can be predicted by the simulation using MINTEOA2	(Van der Bruggen et al. 1998)
Leaching	FA	Use of a chelating agent with an L/S ratio of 2 in a 3-step leaching process using EDTA followed by Na-Gl then	Gluconate is 10–200 times more efficient than water to elute Cd. Cu, and Zn, while EDTA is most efficient for Cd. Changes of the surface particles after leaching increase the amount of leaching due to higher	(Loginova et al. 2019)

methods
separation
of
Summary
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Ta

(Jakob et al. 1995)

Higher L/S ratio and lesser duration are favored for extraction of Pb while lower L/S ratio and long duration (Yang and Yang show better extraction for Cd. pH below 5 is preferred for all the metals. Up to 96% Pb and 93% Cu can 1998)

Ammonium citrate shows great potential in the removal of heavy metals. After 70 days, 86% Cd, 20% Pb, (Pedersen et al. 62% Zn, 81% Cu, and 44% Cr were removed

LS ratio of 10 with a current of 5 A on raw and carbonated pH changes in APC are negligible after the treatment. Leaching of Pb and Zn in the final product does not (Kirkelund et al.

Zn decreases by more than 1 order of magnitude

meet the requirement, but the reduction in leaching of other metals is evident

LS ratio of 10 with the constant current 5 mA/cm² up to a Conductivity of the suspension decreases from 52 mS/cm to between 1.7 and 5 mS/cm, indicating a removal (Jensen et al. maximum of 15 V of the APC undergoes dissolution, and leaching of Cu, Pb, Cd, and 2010)

Jse of Sporosarcina pasteurii and Myxococcus xanthus for Sporosarcina pasteurii results in higher mobility of Cd, Pb, Zh, and Cu. Bacterial treatment results in higher (González et al.

surface area

compaction and lower porosity

15 days

Electrochemical APC

water

FA

Leaching

The chlorides can lead to the formation of volatile metal chlorides which can increase the rate of removal. (Jakob et al. 1996)

Another plausible mechanism is the presence of chlorides which prevents the incorporation of the heavy

metal into the matrix

Temperature of 750 °C, 840 °C, and 920 °C with chloride

additives for 1 h to 6 h

0 g to 200 g of CaCl₂, MgCl₂, and NaCl is being added to Removal of up to 95% for Cd and Pb, more than 80% for Cu and Zn. MgCl2 is found to be the best chloride (Nowak et al. 1 kg ash and heated at 1000 °C for 60 min additives in terms of Zn removal but decreases the removal of Ni due to possible formation of spinel. 2012)

The most effective temperature for evaporating Zn, Pb, Cd, and Cu occurs around $1300 \, ^{\circ}\text{C}$

be recovered

HNO₃, H₂SO₄, HOAc, and NaOAc as an extracting

Temperature from 670 to 1300 °C in air and in argon

atmosphere

agent

FA FA FA

Fhermal Thermal Thermal

volatilization volatilization

volatilization

L/S ratio of 6.5 using 0.25 M ammonium citrate as an J/S ratio of 20 to 40 for 60 min to 90 min using HCl,

APC

FA FA

Electrochemical Electrochemical

Electrochemical APC

assisting agent and 0.8 mA/cm²

favored as it contains excessively high Ca content (Shi et al. 2018). However, solidification using a binder results in an increase in the mass of the secondary waste which has to be disposed of. A new trend to resolve the increase in mass of the secondary product is to make use the combination of various wastes such as APC, FA, coal FA, and solidifying agents to increase the proportion of waste-to-binder ratio (Wang et al. 2015).

Thermal methods

Thermal processes generally aim to reduce the volume of the bulk ash and produce a more stable product, which can be disposed of or utilized. Based on the different operating conditions such as temperature and additives, thermal treatments can be classified into three types: (i) sintering, (ii) vitrification, and (iii) melting/fusion (Table 8) (Lindberg et al. 2015). Although thermal vaporization is considered a separation method in this review, it can also occur concurrently in all three of the thermal methods mentioned. The main disadvantage of thermal methods is usually associated with high treatment cost, but due to the advancement in technology, such methods are becoming increasingly commercially acceptable in countries such as Japan, China, and the USA.

Sintering

Sintering is the coalescence and densification of powdered substances into larger particles at elevated temperatures below their melting point. This process usually takes place at temperatures between 900 and 1100 °C and so is the lowest among all the thermal treatment processes, resulting to lower porosity and higher strength and density. Sintering is effective in reducing leaching of volatile metals (Cd, Pb, and Hg) due to vaporization during the heating process and of other heavy metals due to a reduction in the porosity. Sintering treatment mainly focuses on FA only due to the formation of the amorphous matrix to stabilize the different heavy metals.

The main parameters affecting sintering include pressure, temperature, and duration. Others have investigated the production of ceramics and construction materials using sintering of FA (Mangialardi 2001; Polettini et al. 2004; Wey et al. 2006; Haiying et al. 2007; Wang et al. 2017). In order to reduce treatment cost and improve final product quality, Wey et al. (2006) and Mangialardi (2001) have reported the use of water washing process prior to sintering. Above 960 °C, it is shown that sintering can produce ceramic tiles with low leachability and high mechanical properties (Haiying et al. 2007).

Co-sintering with various additives such as bottom ash and slag to achieve the optimal proportion of CaO and SiO_2 ratio for better mechanical properties has been demonstrated (Hu et al. 2016). It is also found that Ca aluminosilicate formed

during co-sintering is capable of suppressing ash agglomeration and favoring metal chloride recovery via volatilization.

Vitrification

Vitrification is the melting of ash residues with other glassforming additives to produce a homogenous liquid phase between 1100 and 1500 °C followed by rapid cooling to form a rigid and amorphous glassy matrix without crystallization (Quina et al. 2008a, 2008b). Vitrified products are generally more consistent and chemically stable due to the incorporation of heavy metals into the glass matrix to form a homogenous solid phase and also the encapsulation effects provided by the outermost glassy layer (Ghouleh and Shao 2018).

Although vitrification is a technically feasible solution, the high energy consumption during the process increases the treatment cost and this has limited its potential due to economic issues. Therefore, the research has focused on using other additives such as biomass (Alhadj-Mallah et al. 2015) or make use of co-treatment with other wastes (Huber et al. 2016) to reduce the energy demand and make this process more economically viable. However, due to the heterogeneous nature of the FA and APC, the quality of the final product is affected, and the subsequent utilization can be limited.

Melting

Melting process operates at similar temperature as vitrification, but the main difference is that during melting, no additives are added. Also, the final product formed is homogenous and contains various major crystalline phases. As compared to vitrification, melting is more preferable as no additives are required and there is no need for rapid cooling of the final product. Melting systems are reported to be used on a commercial scale to treat ash especially in Japan where disposal at landfills is costly (Ecke et al. 2000). According to Sakai and Hiraoka (2000), the two main systems are (i) fuel-burning melting system and (ii) electric melting system. In general, the fuel-burning system is generally used at smaller incinerators with no power generation, while the electric melting system is preferred at larger incinerators with power generation (Carnogurska et al. 2015; Quina et al. 2008a, 2008b).

Hydrothermal

Hydrothermal process is a form of thermochemical of subcritical or supercritical water at relatively low temperature with the advantage of self-generated pressure. In the recent years, investigations of hydrothermal treatment on FA have become more popular due to the destruction of organic contaminants and improved stability of heavy metal leaching (Hu et al. 2015). It is found out that hydrothermal treatment under alkaline condition can result in crystallization of FA into more

Table 7 Summ	ary of stabi	lization and solidification (S/S) methods		
S/S	Residue type	Methodology	Lesults and findings	References
Chemical stabilization	APC	1.2 mol H_3PO_4 per kg ash with an L/S ratio of 0.4 mixed at 108 ν /min for 10 min	teduced leaching on Ca (6.8%), Cd (37.5%), Cu (57.9%), Pb (99.5%), and Zn (28.2%) Aost of the Pb is in the form of Pb ₅ (PO ₄) ₃ Cl which is stable. The amount of H ₃ PO ₄ does not affect the nH due to high huffering canacity but might beh in initial solubilization of the mineral phases	Eighmy et al. 1997)
Chemical stabilization	APC	Stabilization using a chelating agent, phosphate treatment, and ferrite treatment	hosphate treatment is most effective in reducing the availability of Pb. The redox potential test also (reveals that treatment from a chelating agent causes oxidation in the final material due to the increases in EH or constant by	Mizutani et al. 2000)
Chemical stabilization	APC	Chemical additives such as H ₃ PO ₄ , Na ₂ O·SiO ₂ , Na ₂ CO ₃ , sodium diethyl-dithiocarbamate trihydrate, and sodium hydrosulfide hydrate	tabilization of Zn and Db is achieved, but increased leaching of $Cr(VI)$ is observed. Also the tabilization of H_3PO_4 decreases the acid-neutralizing capacity and is not desirable. Soluble factions such as K, Na, and Cl are not affected much by these chemical additives, and other strategies can be used to remove them	Quina et al. 2010)
Chemical stabilization	FA	Different chemicals such as NaOH, EDTA, sodium sulfide, and thiourea are added to 10 g of fly ash and stirred at 110 ν /min for 8 h	AgoH is effective in reducing the total Pb in the treated residue while EDTA is effective for both Pb (and Ci. Sodium sulfide and thiourea are effective in forming insoluble forms	Youcai et al. 2002)
Accelerated carbonation	APC	Temperature of 20 °C to 60 °C, 0.03% to 50% CO ₂ concentration, for a duration of 4 days to 40 days	$2\Omega_2$ concentration is the most important parameter. Zn and Pb concentration reduces by 2 orders of (magnitude. Cd increases in mobility due to a drop in pH and should be controlled via adding silicates	Ecke et al. 2003)
Accelerated carbonation Accelerated carbonation	FA APC FA	33 L CO ₂ /kg/day for 25 days (APC) and 50 days (FA) L/S ratio of 0.1 to 0.8, 100% CO ₂ atmosphere, pressure of 3 bar, a duration of 3 h	The use of high CO ₂ content during leaching can demobilize Cr by maintaining a reducing condition. (Leaching of Pb and Zn remains high, and the effects of carbonation on APC are low /S ratio of 0.3 is found to be optimum under this condition, and pH after carbonation is decreased (from 12.5 to 7.	Todorovic and Ecke 2006) Li et al. 2007)
Solidification	APC	Stabilization by the addition of polymer PVC, PE, PS, and cement The W/C ratio of 0.4 to 0.48 cured at room temperature for 28 days	The combination of pelletized MSW1-polymer solidification in a hydraulic binder is effective in treducing the release of Pb, Zn, and Na	Massardier et al. 1997)
Solidification	FA	Washing at an L/S ratio of 10 for 1 h with 15% by mass phos- photic acid followed by calcination at 600 °C 75% cement and 25% fly ash with a W/C ratio of 0.5 and 28-day curing duration	The treated fly ash shows pozzolanic effects and can be used to replace cement. The formation of the tringite is able to fix various heavy metals, but the leaching of Cr is still exceeding the limits	Aubert et al. 2006, 2007)
Solidification	FA	Water content of 0.3 to 0.45 with 10% to 80% fly ash replacement Curing at ambient temperature with relative humidity > 90% for nu to 90 days	The amount of hydrated phase which reduces with increasing FA replacement might lead to lowering (compressive strength at short curing age due to cement dilution. For 80% replacement, a SUCS value of 1.05 is obtained at the 90-day curing duration	Polettini et al. 2001)
Solidification	FA	100 g FA added to 100 mL deionized water with the respective dosage of SGA, TBA, SDD, and Na,S	Novel solidifying agents (SGA and TBA) achieve better binding and stabilization due to the multiple (hydrosulfide groups as compared to SDD and Na,S	Wang et al. 2015)
Solidification	FA	Complex silicate cement as a binder for co-disposal of FA and electrolytic manganese residue at an L/S ratio of 2 with NaOH additives	uccessful demonstration of MSWI FA co-disposal with electrolytic manganese residue with 7.5 M (NaOH as the optimal concentration b, Cu, and Mn are well immobilized due to geopolymetric encapsulation. A 25 wt% EMR-FA geopolymer exhibits better leaching results than those of 25% CSC binder	Zhan et al. 2018)

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stable aluminosilicate (Murayama et al. 2002) which immobilize the leaching of metal ions. The silicon-aluminum additives were also found that can assist the hydrothermal process by the formation of aluminosilicate minerals (zeolites) and stabilization of heavy metals (Shi et al. 2017a, 2017b). One of the main products is the tobermorite which is capable of being an ion exchange for heavy metals such as Pb, Cd, and Zn (Coleman and Brassington 2003). Most of the studies revolves for coal fly ash mainly and not so much for FA and APC due to the high amount of Ca and lower amount of Si/Al, which is deemed to be the most important factor for tobermorite synthesis (Shi et al. 2017a, 2017b). However, the newly developed method microwave-assisted hydrothermal treatment with the Na₂HPO₄ additive has demonstrated the high efficiency for solidifying heavy metals in FA, the great energy savings, and low leaching toxicity (Qiu et al. 2017).

Comparison of treatment technologies

Table 9 summarizes the different treatment methods for FA and APC residues. Each treatment method is evaluated based the following factors:

- Technical requirement
- Treatment cost
- Technology development
- Volume change
- Secondary pollutants
- Destruction of organic pollutants

Technical requirement refers to the capital cost of the facilities and the skilled personnel required operating the facilities, which is directly related to the treatment cost. A simple treatment such as water washing and cement encapsulation tends to have low cost ($45 \in t^{-1}$), while simplified equipment is required. Leaching, chemical stabilization, and accelerated carbonation require a certain amount of chemical additives (cost $61-92 \in t^{-1}$). Lastly, treatments such as electrodialysis and thermal processing tend to be rather costly (> $367 \in t^{-1}$) because of the high energy demand and expensive start-up cost of the facilities (Ecke et al. 2000; Quina et al. 2008a, 2008b).

Technology development refers to the stage of the technology (lab, pilot, or industrial scale), and it is an important factor to consider when deciding on a treatment strategy. In general, most established technologies such as chemical stabilization and solidification have lower risks of implementation, but lesser room for research and development. Washing treatment is considered a pretreatment step in combination with other processes. New technologies such as electrodialysis and hydrothermal treatment are at the stage of lab scale to pilot scale due to scalability and economic issues. Thermal processes are more developed (especially in Japan and China) and exist in pilot to industrial scale, but not fully economically viable yet due to the high cost and energy demand (Lindberg et al. 2015). Leaching process such as FLUWA is an industrial-scale technology (Schlumberger et al. 2007) for recovery of metals via chemical leaching. Chemical stabilization is another widely studied and developed technology for many years with industrial-scale projects such as COSMOS (Bontempi et al. 2010), Ferrox (Bontempi et al. 2010), and WesPhix (Eighmy et al. 1997). Cement stabilization has also carried out in industrial scale for FA and APC residues and handles up to 15,000 tons of FA from WtE each year.

Volume change is another important factor when considering the treatment technology. Methods that make use of separation tend to experience a decrease in volume of the final product, but they generate a secondary effluent which is more concentrated and toxic and requires further treatment. Separation methods such as washing, leaching, and electrodialysis generate wastewater that contains soluble salts and heavy metals. High-temperature treatment such as vitrification produces a concentrated fly ash with highly volatile metal chlorides of Pb and Zn. On the other hand, sintering, cement binder, and chemical stabilization focus on the encapsulation and stabilization of the heavy metals and they do not produce secondary pollutants, but the volume of the final product tends to increase due to the various additives added to stabilize FA.

Lastly, the fate of organic pollutants such as PCDD/F emitted from waste incineration is of a major concern over the past few decades due to their high toxicity. These organic pollutants are usually found in boiler and filter ash (Vehlow et al. 2006). Although their leaching can be reduced via encapsulation and chemical fixation, the optimal solution is to destroy them using the thermal process. Thermal methods such as vitrification and melting which use extremely high temperatures (over 1000 °C) have shown over 99.9% destruction of organic compounds (Sakai and Hiraoka 2000; Nishida et al. 2001).

Utilization of FA and APC residues

The main objectives of waste incineration are volume reduction and energy recovery, whereby the residual ashes are considered as a form of secondary waste to be landfilled. In the recent years, due to the increasing amount of FA and APC generated, there is much controversy on the proper management of these ashes and residues. Even after proper treatment, the final product faces some limitations preventing utilization in many applications (Lam et al. 2010). Table 10 provides a summary of the utilization methods categorized by the ash types, development stage, and the limitations faced.

Table 8 Summary of thermal methods

Thermal	Residue type	Methodology	Results and findings	References
Sintering	APC	0% to 30% (<i>w</i> / <i>w</i>) APC added to clay compressed at 20 MPa before sintering at 1000 °C for 5 min	Water adsorption and shrinkage of final ceramic products are at 9% and < 9%, respectively, and can be used as second-class ceramic tiles. TCLP leaching is below limits. Biotoxicity tested against <i>Escherichia coli</i> , <i>Staphylococcus aureus</i> , and MG63 shows that the APC is effectively detoxicated via the sintering.	(Deng et al. 2018)
Sintering	FA	Compacted at 15 N/mm ² to 28 N/mm ² with 8% moisture Followed by sintering at 1140 °C for 15 min to 120 min	High Cl content in ash leads to higher mass loss and contributes to lower compressive strength after sintering. Higher compact pressure also results in higher compressive strength of the sintered products. Significant strength increases as duration increases un till a 60-min duration	(Mangialardi 2001)
Sintering	FA	Compacted at 28.3 MPa, sintered at 1100–1150 °C for 30 min and 60 min	Densification of the sintered product is shown via $0.5-12\%$ shrinkage and density increase of $1.5-2$ g/cm ³ . Leaching is reduced due to metal encapsulation and bonding mechanism within the sintered product. Volatile metals are being released (56% Zn. 88% Pb. and 63% Cr)	(Polettini et al. 2004)
Sintering	FA	Washed at an L/S ratio of 20 and dried at 105 °C for 24 h. Sintered at 700–900 °C for 1 h to 2 h	Sintering at 900 °C results in grain coalescence in fly ash to produce a smoother surface under SEM micrographs. Energy demand is reduced if the ash is pretreated with water washing. Most metals are stabilized with the exception of Cr which increases after sintering	(Wey et al. 2006)
Sintering	FA	Combined disc palletization of fixed ash followed by thermal treatment at 450 °C to 1050 °C	Pellet with 0.15 kg/kg water content shows high mechanical strength and is possible for further processing. 450 °C is enough to reduce the leaching to a safe limit while higher temperate improves the separation of volatile metals like Cd and Pb into the secondary fly ash. 650 °C is also necessary to reduce the TEQ of PCDD/F and DL-PCB. A duration of 10 min is enough for the thermal treatment	(Huber et al. 2018a, 2018b)
Vitrification	APC	Vitrification at 1450 °C for 1.5 h with different amounts of bottom ash additives	The increase in bottom ash added results in lower basicity and formation of a more amorphous matrix. Leaching of Pb, Cd, and Cr is reduced while Zn and Fe increase. 50% volume reduction is achieved after vitrification	(Li et al. 2003)
Vitrification	FA	1200–1500 °C with silica sand (glass forming), sodium carbonate, borax (fluxing agent), and titanium oxide (nucleation agent)	With proper composition of additives, the temperature of vitrification can be lowered from 1500 to 1200 °C	(Yang et al. 2009a, 2009b)
Vitrification	FA	FA mixed with biomass in the ratios 3:1, 2:1, and 1:1 vitrified at around 1100–1500 °C	Biomass such as wood and rice husk ashes can be used to reduce the energy cost during melting. Wood ash that contains higher alkaline contents is capable of reducing the melt flow temperature from 1426 to 1179 °C due to its neutralizing effects to form polycrystalline products. The melting heat of 1900 kJ/kg for the melting mixture is 700 kJ/kg lesser than that of pure FA	(Alhadj-Mallah et al. 2015)
Melting	FA	1260 °C, 1320 °C, and 1350 °C melting temperatures	Higher melting temperature increases the density and volume reduction. Vaporization of Pb is up to 99.9% at 1350 °C while more than 80% of Zn and 95% of Cr remain in the melted residue. The dioxin level is 0.053 ng TEQ/m ³ after treatment	(Wang et al. 2008)
Melting	FA	1450 °C melting for 10 min under 100 kW DC plasma arc followed by air cooling or water quenching	Concentration of CaO, SiO ₂ , Al ₂ O ₃ , and MgO increases while that of Cl and SO ₃ decreases sharply after treatment. Water quenching shows lower leaching results when compared to air cooling due to less time for crystallization and formation of a more amorphous glassy product	(Ni et al. 2012)
Hydrothermal	FA	CFA, bentonite, and kaolin are being added to MSWI FA at 1:9 and 3:7 ratios for hydrothermal treatment at 150–200 °C	Formation of aluminosilicate zeolites such as tobermorite is observed after hydrothermal treatment. Pb, Zn, Cd, Cr, and Cu leaching have significantly reduced, but Pb still exceeds the limit. It is indicated that the heavy metals are stabilized within the zeolites rather than transferred to the liquid phase during the hydrothermal process. Higher temperate results in higher tobermorite formation but also increases the amount of leaching	(Shi et al. 2017a, 2017b)

	Separation	n		Solidification	and stabilization		Thermal		
	Washing	Leaching	Electrodialysis	Chemical stabilization	Accelerated carbonation	Cement encapsulation	Sintering	Vitrification	Hydrothermal
Technical requirement	Low	Low	High	Low	Mid	Low	High	High	High
Treatment cost	\$	\$\$	\$\$\$	\$\$	\$\$	\$	\$\$\$	\$\$\$	\$\$\$
Technology development	Lab	Industrial	Lab	Industrial	Pilot	Industrial	Pilot	Pilot	Lab
Volume change	Decrease	Decrease	Decrease	Increase	No change	Increase	No change	Decrease	Decrease
Secondary pollutant	Yes	Yes	Yes	No	No	No	No	Yes	Yes
Destruction of organic pollutants	No	No	No	No	No	No	Yes	Yes	Yes
Suitable for FA or APC	FA	FA	FA + APC	APC	FA + APC	FA + APC	FA	FA + APC	FA

Table 9 Comparison of FA and APC residue treatment methods

Metals and salt recovery

Recovery of metals and salt from FA and APC has been investigated widely, and the process is well established in large commercial scale (Schlumberger et al. 2007). The recovery process usually involves chemical leaching such as acid (Funari et al. 2015), commercial chelating agent with metal selectivity (Tang and Steenari 2015), or bioleaching for a more environmentally friendly and low-cost process (Ramanathan and Ting 2016).

Typically, volatile heavy metals, such as Pb, Zn, Cd, and other rare earth metals, in FA are being separated from the rest of the residues for detoxification and recovery, while APC does not undergo such treatment due to high acidneutralizing capacity and low metallic contents. However, the recovery of rare earth metals is not economical due to the low amount of those metals in FA (Funari et al. 2015). After the selective removal of metals, a large amount of soluble salt remains within the extracting liquid. These salts are usually not recovered, and instead, the extracting liquid is required for further treatment before disposal (Colangelo et al. 2012). An exception is the HALOSEP® process developed to recover chloride brine from FA and APC, which can be used in winter to de-ice the road (Quina et al. 2018).

Cement production and cement replacement

Cement is the most commonly used building material in the world, and as FA and APC have very similar composition, they are used as partial replacement to reduce the amount of energy required and CO₂ generated during cement production. There is huge interest in using FA and APC in both production of cement and as a cement replacement (Siddique 2010).

The application of FA in various studies has shown positive results in the hydration behavior for up to 30% replacement as alkali metal enhances the hydration, while Zn, Pb, and Cd retard the hydration (Lederer et al. 2017). The presence of high chlorides and sulfate content can also be detrimental to the early strength due to slowing hydration. Although chlorides and sulfate can be easily removed by a simple washing method, the washed FA experiences a loss of pozzolanic activity after washing (Keppert et al. 2012). Also, the metallic Al and Zn present in FA generates a significant amount of H₂ which results in expansion and voids within the concrete and affects the compressive strength (Aubert et al. 2004). Despite these limitations, FA as a cement replacement is still a viable option as it has the added advantage of stabilizing the heavy metals, while it also possesses pozzolanic properties that can help improve the long-term strength of the concrete (Joseph et al. 2018).

APC, on the other hand, is not directly used as a cement replacement due to the high Ca content (Shi et al. 2018), but it is commonly co-processed as a raw material in cement production in rotary kiln (Joseph et al. 2018). Major advantage of using APC is that it contains lesser carbon phases than limestone, and thus, it helps to reduce the emissions during cement production. In addition, the high temperature of the rotary kiln can concurrently destroy the organic pollutants in APC (Bogush et al. 2015). It is suggested that the use of APC in clinker production results in a cement with lower workability and lesser setting time, so gypsum should be added to delay the setting (Kikuchi 2001). The presence of high chloride content in APC is also a concern as it might cause clogging

Table 10 Utilization andlimitations of FA and APCresidues

Utilization	Ash type	Technology development	Limitations
Metal and salt recovery Cement production	FA FA/APC	Industrial scale Industrial scale	Economical valuesSecondary emission of metal chlorides
Cement replacement	FA	Pilot scale	High cost and energy demandProduct quality
			• Limited amount of utilization
			Delayed ettringite formation
Pavement and embankment	FA	Pilot scale	Expansion due to metallic Al/ZnLeaching of heavy metals
			Low durability
Zeolite synthesis	FA	Lab scale	Leaching of chloridesLeaching of heavy metals
Lightweight aggregate	FA/APC	Lab scale	 Low-quality zeolite Leaching of heavy metals
			• Risk of ASR
Glass ceramic	FA	Lab scale	Product qualityEconomical values
			High cost and energy demand
			 Leaching of heavy metals

and corrosion problems. As such, it is of high importance that the APC undergoes proper pretreatment before being used as raw material in cement production (Saikia et al. 2007).

Use as pavement and embankment

The construction of roads and embankments tends to use a large amount of natural aggregates. The use of FA in replacement of these natural aggregates can help to conserve the natural aggregates for better applications (Francois and Pierson 2009). The majority of the studies make use of bottom ash as the base layer for road construction, while lesser studies use FA and APC due to the higher leaching and lower durability.

The leaching issue, however, can be resolved by washing pretreatment of FA and APC to meet the leaching standards before utilization (Åberg et al. 2006). A FA melting plant for stone production in Japan also successfully demonstrated a commercially viable use of FA pavement blocks in a park (Nishida et al. 2001).

Zeolite synthesis

Zeolites are crystalline aluminosilicates that can be used as sorbents, catalysts, and ion exchange applications. Despite the low Al and Si content in FA and APC due to the significant portion of Ca, the hydrothermal process under alkaline conditions has proven to be effective in synthesizing zeolites (Wałek et al. 2008). Although the typical cation exchange capacity (CEC) of FA and APC zeolites is only 90 mEq/100 g as compared to commercial zeolites at 200–300 mEq/100 g, the feasibility of using waste-derived zeolite is still of high interest. Research on the modification of the FA-derived zeolite improved the CEC to

be comparable to other sorbents (Miyake et al. 2002). To deliver calcium-containing aluminosilicate zeolites such as tobermorite, the addition of a 30% mass of composite additive (including CFA and diatomite) and a 3% mass of tobermorite seed (reaction temperature 200 °C) was applied for seed-assisted hydrothermal solidification of MSWI fly ash (Shi et al. 2016). The developed technology increased the kinetic formation of tobermorite, while inhibited the hibschite generated from tobermorite.

Glass ceramics and lightweight aggregates

FA and APC contain a high amount of SiO₂, Al₂O₃, and CaO and are considered suitable to replace clay for the production of glass ceramics after thermal treatment such as sintering and vitrification (Andreola et al. 2001). Despite the use of raw FA which resulted in detrimental effects on the mechanical properties of the ceramic due to excessive chloride content, Haiying et al. (2007) managed to achieve high-strength ceramic tiles with low water adsorption rate with 20% FA. Successful synthesis of lightweight aggregate was achieved by heating APC with clay at an elevated temperature of 1170 °C (Quina et al. 2014). Such lightweight aggregates have dense ceramic outer shell with good mechanical properties with up to 50% lower density (due to honeycombed internal non-interconnected pores).

Perspectives

Mixing of FA and APC

In some countries, FA and APC are usually collected as a combined waste (del Valle-Zermeño et al. 2013). However,

16991

others suggested that FA and APC should be managed separately due to their differences in the composition and, hence, chemical properties (De Boom and Degrez 2012).

In this review, APC and FA are compared in terms of physicochemical characteristics and treatment methods, and the literature has suggested separate collection of APC and FA, and treatment as two different materials. The compositions of FA and APC are sufficiently different, and their properties could be used in different applications. For example, FA which has a higher content of Al₂O₃ and SiO₂ is favorable for zeolite production (Yoo and Jo 2015), while APC which contains excess lime content is useful for cement clinker production (Stegemann 2014). When the two are mixed, the resultant waste is less favorable for both applications.

Mixing of FA and APC could also result in some undesirable outcomes such as explosion due to the release of hydrogen gas (Mizutani et al. 2000). The release and buildup of hydrogen gas can be due to the presence of aluminum under high pH conditions (incompatible waste) (Zhang et al. 2009; Hu et al. 2011). The authors argue separation of FA and APC is desirable to prevent FA (which has a high aluminum content) contacting APC (which has high alkalinity) because it can cause production of hydrogen gas.

Lastly, when FA and APC are mixed, the pH of the combined residue is still high (10 to 12) and this might result in the enhanced leaching of amphoteric metals from the FA as reported by Zhang et al. (2016). In addition to the increase in pH, the ANC of the combined waste is also relatively high, and this would make it harder to adjust the pH for other processes.

True cost of treatment

Treatment cost has always been a concern. However, treatment cost should not be evaluated on the basis of only the treatment process. In addition to the cost of the treatment process, there are also the cost of risks and environmental impacts of the resulting material, the value of the final products, and the cost of maintenance/monitoring after disposal.

As mentioned previously, different treatment methods yield different types of treated products with various qualities. Therefore, it is important to consider the cost in a more holistic manner. Low-cost treatment methods such as stabilization and solidification usually generate a larger volume of final product, which might not be suitable for utilization, and it requires to be sent to the landfill for disposal. In this case, the cost of maintaining the landfill for a larger amount of residue should be added on to the true cost of treatment, whereas in using the thermal treatment, the final product is usually of higher quality and can be utilized in various applications. In this case, the value of these final products can be used to offset the higher cost of treatment (Luo et al. 2018).

One important aspect that has been undervalued is the risk of monetization by evaluation of environmental risks into dollar terms for quantification. It is very difficult to translate the value of long life, good health, and clean air into dollar as there is the fundamental flaw that each individual values each factor very differently. Therefore, there is a need for environmental and government agencies to evaluate each factor and incorporate subsidies and tax to promote proper treatment for FA and APC residues.

Challenges and opportunities

Despite the numerous reports in the literature, there are few case studies treating FA and APC residues at full scale for utilization purposes. The challenges and opportunities for further utilization of these secondary resources are identified in this review paper.

Firstly, one of the main issues with FA and APC is the variation in terms of mineralogy, morphology, and chemical properties due to the different operating conditions at the various locations. It is, therefore, difficult to propose a single method to treat FA and APC. Proper characterization before treatment in each case is essential before deciding on the most appropriate method to use. In addition, the heterogeneous nature of the treated products makes these less valuable and less applicable. Consequently, researchers have focused on stabilizing the FA and APC just enough for disposal in landfill. The development of a treatment method, which can convert FA and APC into a homogenous product, is a challenge.

Next, many researchers have focussed on the heavy metal content before and after treatment and have neglected the effect of treatment on organics such as dioxins due to the difficulties associated with analysis of such compounds. Development in analytical methods for such compounds is needed.

Lastly, the treated products could still create problems in the longer term because of leachable contents and such concern would reduce public acceptance on utilization of treated FA and APC. More studies on the long-term effects of treated FA and APC should be conducted to increase knowledge and public confidence in utilizing these resources.

Conclusions

As solid waste management increasingly shifts towards incineration for energy recovery, more by-products such as FA and APC residues are generated. These are considered hazardous due to the presence of heavy metals and soluble salts and must be treated before disposal. However, it is evident that FA and APC residues have potential in many applications such as zeolite production, road pavement, construction material, and ceramics. In order to utilize or safely dispose of FA and APC residues, treatment must be applied to improve quality and reduce the negative environmental implications. In general, the three treatment types are separation, thermal, and solidification/stabilization processes. Both thermal processes and S/S processes may be found at full scale with separation processes often serving as a pretreatment. Increasing amounts of FA and APC are being produced, and there is a need to explore the potential application of the treated material instead of just disposal. This is necessary for a more sustainable approach to waste management.

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