

Review

Incineration Fly Ash and Its Treatment to Possible Utilization: A Review

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Abstract: Incineration has gained popularity over landfill as a key solution for the reduction of massively increasing volumes of municipal solid waste (MSW) generation worldwide and in particular China. However, it is not the end solution. The disposal of the incineration residues, which are enriched with a wide range of heavy metals and soluble salts, has become a challenge for the environmental managers. The aim of this study was to review the increasing urbanization and its repercussion on waste generation in China; waste management options were compared for possible environmentally friendly considerations. Treatment techniques of incineration fly ash were discussed to determine the effectiveness of obtaining environmentally stable material, and, finally, possible applications of incineration fly ash for utilization were discussed based on identifying the processing suitability, performance and environmental impact of incineration fly ash for its applications.

Keywords: municipal solid waste; incineration; fly ash; urbanization; heavy metals; treatment techniques

1. Introduction

In 2016, around 2.01 billion tons of solid waste was generated worldwide. With increasing population, urban expansion and industrialization, the annual waste generation is expected to increase by 70% in the coming two decades [1]. Chinese urban expansion is increasing year by year and so is the generation of municipal solid waste. With around 18.5% of the world's total population, China ranks on top for Municipal Solid Waste (MSW) generation. Landfill, being the most widely used solid waste disposal option, is confronting numerous challenges attributed to land restriction, contaminations and legislations worldwide. How to dispose of millions of tons of MSW is becoming the biggest issue for environmental managers [2,3]; alternatively, municipal solid waste disposal is expected to make developments in improved landfill bioreactors, composting and thermal processes. Amongst these, incineration is widely accepted because of its small land footprint, huge mass and volume reduction, and, additionally, energy can be recovered in the form of heat and electricity [4–6]. MSW incineration helps to achieve a 70–90% reduction in mass and volume [7,8]. In comparison to sanitary landfills, incineration is more acceptable and valued in developed and under developing countries such as China because of its huge population, increasing land prices and increasing environmental awareness in people.

However, MSW incineration confronts the problem of how to deal with the considerable amount of incineration residues (~30% by mass), which mainly consist of bottom ash and fly ash. According to the International Ash Working Group (IAWG), fly ash is defined as the ash that is discharged from the combustion chamber without the addition of any type of sorbents, such as the ash from a super heater, economizer and air preheater. In the study of municipal solid waste incineration (MSWI), fly

ash is referred to the air pollution control (APC) residues that include the ash trapped by electrostatic precipitators and bag filters. It is estimated that the amount of fly ash produced accounts for approximately 3–10% of incineration waste [9]. The fly ash possesses a higher amount of heavy metal concentrations than that of bottom ash, which is resulted by the chemical partitioning of heavy metals accelerated by the temperature and the presence of chlorine, originated from the types of wastes incinerated (food, PVC, etc.) [10]. Therefore, various countries, such as the USA, France, Canada, Japan, Italy, Germany, Sweden, etc., have introduced policies for the proper control and management of MSWI fly ash. In 2016, the Chinese government also included MSWI fly ash in the National Hazardous Waste List. The law issued by the National Environmental Protection Standards of the People's Republic of China (HJ 1134—2020) in August-2020 will prevent and control environmental pollution, improve the quality of the ecological environment, standardize and guide the environmental management of MSW incineration fly ash. According to this law, the regulations specify the overall requirements for the pollution control of MSW incineration fly ash, collection, storage, transportation, treatment and disposal. Which means fly ash in China cannot be disposed of without prior treatment [11–14].

Therefore, various treatment techniques are exercised to eliminate the hazardous and toxic content in MSWI fly ash. The characteristics of MSW incineration fly ash may serve as a key tool to choose the best option for treatment [11,12,15]. At present, there are lots of studies carried out on the stabilization/removal of heavy metals from MSW incineration fly ash. The treatment techniques can be classified as separation techniques, stabilization/solidification and thermal treatments. The treatment techniques are differentiated mainly on the characteristics of process product than that of the process [16]. The separation techniques firstly separate the heavy metals from fly ash using a chemical reagent, biological reagent or electrodialysis method. The cement solidification refers to the use of fly ash as an additive to the concrete production process. During the production process, the heavy metal components in fly ash are fixed in the concrete product. This technique not only realizes fly ash as a resource material for the production of cement, but also facilitates the harmless treatment of fly ash, whereas chemical stabilization involves different organic chelating reagents or inorganic chemicals to reduce the leaching of heavy metals in fly ash to facilitate subsequent disposal. The thermal treatment is further divided into sintering, melting, vitrification and thermochemical treatment. The treated fly ash is then required to be either disposed of or utilized in possible applications such as construction material, geotechnical applications, agriculture, etc. In each application for fly ash reuse, recycle and recover comes advantages, for example, conservation of natural resources, elimination of waste and availability of zero-cost raw materials.

To date, several research studies and reviews have been published for MSW generation and management [11–13,17–20], treatment techniques of incineration residues [21–27], social factors [25–28], demographics [28–31], economics [28,30,32,33] and on policies [34]. However, little work has been done to understand and investigate the rapid urbanization and its consequences on waste management. The occurrences of heavy metals, treatment techniques and existing practices related to the reuse of MSW incineration fly ash to identify potential application in China. Therefore, the aim of this study was to review the increasing urbanization and its repercussion on waste generation in China; waste management options were compared for possible environmentally friendly considerations. Treatment techniques of incineration fly ash were investigated to determine effectiveness in obtaining environmentally stable material, and, finally, possible applications of incineration fly ash for utilization were discussed based on identifying the processing suitability, performance and environmental impact of incineration fly ash for its applications.

2. Urbanization and Waste Management in China

2.1. Situation in China

China is a developing country. Over the past three decades, the world has witnessed unparalleled growth in Chinese urban settlement and economy. China's tremendous development has not only brought economic incline, stability and improved living standards but has also attracted

huge environmental concerns for the generation of uncontrolled waste. China is in fact the only country in the world that has mass migration within the country. The urban population proportion in 1996 was 207.4 million, making up to 17% of total population, but, according to 2019 statistics, the urban population in China reached around 874.4 million, which holds 60.4 % of total population of China. In China urban areas are highly correlated with solid waste generation of all cities with respect to their area and amount of waste generated. Because of this rapid urbanization and increasing life standards, China ranks top in the growth of MSW generation. Thus, uncontrolled increase in the volume of solid waste generation is becoming a bottleneck issue. A number of cities in China are struggling to cope with the serious crisis of solid waste generation and its management because of the increasing land prices and scarcity of land available for landfilling. The statistics presented during the year 2017 MSW generation in China surpassed 220.6 million tons per year, and the collected and disposed amount was 198 million tons. The increase in waste generation is predicted to increase 3.6%, and the disposal and collection rate is expected to increase 8.8% per year. It is estimated that the urban population in China will become 947 million and it will account for 65.9% of total population in 2025, and MSW generation will reach 325 million and 480 million tons in 2020 and 2025, respectively [35–40]. Therefore, research studies should be carried out to quantify the impact of urbanization on the composition of municipal solid waste, which has so far received little attention in China.

2.2. Waste Management

In order to handle and create awareness for waste management Chinese government has initiated some measures to encourage the development of safe MSW disposal. For example, the introduction of “eco-cities” or “environmental protection model cities” has gathered huge positive turnout [12]. Every city wants to win this title, but, to secure the title, cities need to achieve 85% to 90% safe MSW disposal. The growth of MSW management industry is gaining continuous support and financial assistance under the national environmental investment projects: USD 115 billion was allocated for the 12th five-year plan (from 2011 to 2015) and around USD 150 billion was invested for the 13th five-year plan (from 2016 to 2020) [41–43]. Such measures not only attract safe disposal and waste minimization but also play a positive role in environmental awareness. Although waste management in China is improving, it is still a challenging task, and to achieve required results needs more time and innovation in waste management [44,45]. According to [15], in Germany, all landfill sites for untreated municipal solid waste were completely shut down during 2005; however, in China, more than 57% of waste disposal is still accounted for by landfill. MSW management in China is also increasingly focusing on the recycling of wastes to avoid environmental pollution caused by the growing amounts of MSW. The Chinese government has also set a target of disposing nearly a third of the country’s waste with waste-to-energy plants by 2030.

2.3. Recycling Valuable Waste Fractions

With the continuous expansion of urban areas and industrialization, China is confronting several challenges, including resource depletion and environmental pollution. Furthermore, the growing amounts of municipal solid waste are becoming problematic owing to the limited availability of land for new landfill sites. Thus, its necessary to encourage the utilization of recyclable waste, which can simultaneously cover the issues of limited land for landfill sites, natural resource depletion and environmental pollution. Recyclable wastes are wastes generated from domestic sources and industries, but they can be recycled after adequate processing and sorting. The main examples involve metal waste, paper waste, glass waste, plastic waste, tires, electrical and electronic equipment waste and home appliances. The process of recyclable waste management is still in its infancy in China compared to other developed countries, such as Japan [46,47], which has achieved a great deal in establishing an urban waste recycling system. In this regard, the Chinese government is effectively promoting the recycling of recyclable wastes, multiple documents have been released for promoting and encouraging the urban waste recycling and source separation in China [48–53]. In 2006, the Ministry of Commerce (MOC) of China introduced a plan of recyclable waste recycling in 26 cities on a pilot scale, which then expanded from 29 cities to 35 cities from 2009 to 2012 [49–51].

Most of the pilot projects of recyclable waste recycling systems are developed in eastern parts of China [54]. The aim of developing pilot projects was to create awareness at a local government level to gain experiences from recyclable waste recycling pilot projects and then implement it on a local level based on the local realities.

In last decade, considerable progress has been seen in recycling valuable waste fractions in China. According to the statistics presented in 2016 by [55], recyclable wastes reached around 246 million tons. The recycling of most common wastes, such as plastic waste, paper waste, automobile waste, steel waste and electrical and electronic equipment waste has been improved. For example, in China, waste paper is regarded as the most common secondary resource, and its recycling rate increased from 27.5% in 2001 to 46.7% in 2015 [55]. In comparison, the recycling rate of plastic waste was lower, increasing from 20% (2001) to 30% (2015), and around 19.7% was observed for electrical and electronic equipment waste in 2015 [55].

Although the recycling of waste is increasing day by day in China, there are still some challenges to face, for example, insufficient municipal solid waste separation, MSW being the dominant waste, which has a high proportion of organic components, and the separation of domestic waste is also low [56]. These problems can be well handled by (1) the creation of proper recycling platforms, (2) encouraging sufficient MSW separation, (3) establishing market with effective government administration and (4) the introduction of new policies which can encourage and educate the common man about waste and its importance.

2.4. Waste Disposal Options

The waste disposal options in China are mainly landfill and incineration [57–60]. The other disposal options account for a very small percentage, as can be seen in Table 1, which gives an overview of MSW generation and disposal in China between 2003 and 2016. An obvious increase in the amount of waste incinerated can be seen from 2.49% to 35.25% from 2003 to 2016.

Old disposal options seem easy to handle but they carry huge problems. The MSWI residues can cause leachate enriched with salts and heavy metals, which is the main drawback for the utilization of MSWI residues. Therefore, to compare and determine waste disposal options, a comparison of disposal options can be seen in Table 2, which defines the role of each disposal option based on factors such as scale, site feasibility and its dependence on technology, the status of final product and its marketing, the main causes of pollution (air, soil and ground water), the measures taken to protect the environment and finally the current statuses of development of each disposal options, which were presented and analyzed based on the environmentally friendly considerations. Based on the comparison of waste disposal options, it can be stated that these waste options are less likely to be adopted in a modern world where increasing environmental awareness and stringent legislation rejects any such step, which endangers the environment. The Chinese government is struggling to seek out more effective ways to dispose of MSW. As in Singapore and Japan, the ratio of waste incineration is more than 90%, which is not only minimizing waste burden but also generating energy [60,61]. In China, compared to other waste management and disposal options, incineration serves the management of MSW very well because it helps in the reduction of mass, volume and generation of energy.

Table 1. MSW generation and disposal in China between 2003 and 2016 (data from National Bureau of Statistics (NBS, 2017).

Year	Collected & Transported MSW (M/T)	Number of MSW Disposal Facilities				MSW Disposed (Tons)				Share of Incineration in MSW Disposal (%)
		Total Number	Landfills	Incineration Facilities	Others	Total Volume	Landfilled	Incinerated	Other Means	
2003	148.59	575	457	47	NA	7544.7	6404	369.9	NA	2.49
2004	155.17	559	444	54	NA	8088.7	6888.9	449	NA	2.90
2005	155.82	471	356	67	NA	8051.1	6857.1	791	NA	5.08
2006	148.44	419	324	69	NA	7872.6	6408.2	1137.6	NA	7.67
2007	152.16	460	366	66	NA	9437.7	7632.7	1435.1	NA	9.43
2008	154.45	509	407	74	NA	10,306.6	8424	1569.7	NA	10.17
2009	157.39	567	447	93	NA	11,232.3	8898.6	2022	NA	12.85
2010	158.06	628	498	104	NA	12,317.8	9598.3	2316.7	NA	14.66
2011	164.06	677	547	109	21	13,089.6	10,063.7	2599.3	NA	15.85
2012	170.82	701	540	138	23	14,489.5	10,512.5	3584.1	393	20.98
2013	172.45	765	580	166	19	15,394	10,492.7	4633.7	267.6	26.88
2014	178.65	818	604	188	26	16,393.7	10,744.3	5329.9	319.6	29.84
2015	191.46	890	640	220	30	18,013	11,483	6175.5	354.4	32.26
2016	203.65	940	657	249	34	19,673.8	11,866.4	7178.4	428.9	35.25
Total	2310.5	8979	6867	1644	153	171,905.1	126,274.4	39,591.9	1763.5	17.14

Table 2. Comparison of waste disposal options.

Consideration	Incineration	Landfill	Composting
Scale	150 to 1200 t/d	Large scale	100–200 for static and 250–500 for dynamic
Site feasibility	Siting is sensitive, transportation distance should short, possibility of siting it near to downtown	Very hard, high land prices, away from urban areas, transportation within 30 km of distance.	Easy, away from populated areas, short distance
Dependability on technology	Dependable, accepted worldwide, still growing	Old technique, dependable	Acceptable
Product and its market	Generation of energy	Generation of biogas, source for power generation	Single product with low market acceptability
Final product	Demands treatment prior to disposal or reuse	Disposal option itself	Demands landfill disposal
Air pollution	Strictly demands to control flow gases, heavy metals, salts and dioxins	Chance of air pollution if left uncovered	Odor, requires odor prevention
Soil pollution	Prior site selection for ash pile /stacking	Restricted to landfill site	pH and content of heavy metals should be controlled
Ground water pollution	Dependent on construction of landfill	Leaked bottom can pollute within 3 km radius of landfill sites	Low chances
Environmental protection measures	Residue disposal, waste mass loss, No noise pollution, odor control and prevention.	Bottom leak-proof but it requires huge investments	Requires high production cost
Development and current status	Increasing and gaining popularity	Numbers are getting lower and lower. Restricted in many countries	Limited

2.5. Waste Incineration

In China, MSW incineration technology was initiated during the 1980s and underwent a rapid development and acceptance in the 1990s. According to statistics, the share of MSW incineration has gone bigger from 2.49% in 2003 to 35.25% in 2016. The MSW generation and disposal in selected provinces of China can be seen in Table 3, which discloses the operational MSW incineration plants in China. It is reported that during 2016 China had 299 incineration plants, which handled 280,000 tons of waste per day in different towns and cities. It is predicted that by the end of the “13th 5-year plan” in 2020, the proportion of waste incineration will increase and the number of incineration plants will reach to 400. So, the majority of MSW incinerators are distributed in eastern and coastal parts of China. Some developed economies and higher population density provinces, such as Guangdong, Zhejiang and Jiangsu, share nearly half of the incinerators in China [10], as shown in Table 3. Guangdong, being the most populous province, collects and transports around 23 million tons, of which around 7.86 million tons are incinerated, whereas, with a huge number of incinerators, up to 60% of MSW is incinerated in Jiangsu and Zhejiang province alone. In central and western parts of

China, incinerators are found only in the provincial capitals, probably because of lower economies than those eastern and coastal parts of China.

With the steady increase in combustion in China, it is also important to ensure that the environment is protected from direct emissions into the atmosphere. Since the incineration residues consist of particulate matter collected through different air pollution control devices, mostly characterized by higher concentrations of salts, heavy metals, etc., these characteristics hinder the use of subsequent applications and demand treatment to make it environmentally friendly.

Table 3. MSW generation and disposal in selected provinces of China in 2016 (data from National Bureau of Statistics (NBS, 2017).

Province	Collected and Transported MSW (Million Tons)	No. of Incinerators	MSW Landfilled (Million Tons)	MSW Incinerated (Million Tons)	Landfill Capacity (Tons/d)	Incineration Capacity (Tons/d)	Share of Incineration in MSW Disposal (%)
Beijing	8.73	7	4.73	2.73	9.14×10^{-3}	1.04×10^{-4}	31.23
Shanghai	6.29	7	3.30	2.73	1.12×10^{-4}	1.13×10^{-4}	43.36
Jiangsu	15.6	30	4.52	11.1	2.23×10^{-4}	3.31×10^{-4}	71.00
Zhejiang	14.3	35	5.99	8.35	1.36×10^{-4}	3.45×10^{-4}	58.24
Shandong	14.7	24	7.05	7.08	2.01×10^{-4}	2.06×10^{-4}	48.25
Hubei	8.80	12	4.45	3.75	1.13×10^{-4}	1.25×10^{-4}	42.56
Guangdong	23.9	24	14.8	7.86	4.00×10^{-4}	3.00×10^{-4}	32.87
Chongqing	4.64	3	2.99	1.95	7.35×10^{-3}	4.40×10^{-3}	41.93
Sichuan	8.87	14	5.12	3.62	1.24×10^{-4}	1.21×10^{-4}	40.81
Guizhou	2.94	4	2.38	0.404	6.89×10^{-3}	2.40×10^{-3}	13.74

3. Occurrence of Heavy Metals in MSW Incineration Ash

The characterization and composition of incineration ash varies over time and from region to region and country to country due to the difference in waste management/treatment practices [62]. In China, composition of MSW is extremely non-homogenous and variations may be because of living conditions, geographical distance, industrialization and urbanization [63–65]. Incineration of MSW in China faces serious technical challenges owing to the low calorific values. MSW in developed countries such as the USA, Japan and South Korea has higher heating values, ranging from 8400 to 17,000 kJ/kg, due to the presence of more combustible components and lower moisture contents. According to [64–66], MSW in China is dominated by food waste, and its moisture content is near to 50%. In contrast to this, the moisture content is around 20 to 30% in Europe and North America. In China, the waste management is lacking in implementing an appropriate policy for the recovery of resources from waste, for example, rubbish collectors collect the majority of combustible materials such as plastic and paper. Therefore, to handle this issue of low calorific values, the uses of supplementary fuels are often observed in China, hence the incineration residues emerge with more challenges, as supplementary fuels also contain heavy metals.

The heavy metal contents in fly ash are determined by the source characteristics of MSW, incineration conditions and air pollution control (APC) devices installed. The similar combustion technology and varying APC devices and difference in the source of MSW generated in different parts of China may also be the cause of variations in the content of heavy metals [10]. Fly ash is typically collected in various types of cyclones, filters and electrostatic precipitator. The wastes carried over by flu gases from combustion chamber are categorized in different groups, as shown in Figure 1 by [16,25], such as fly ash and APC residues.

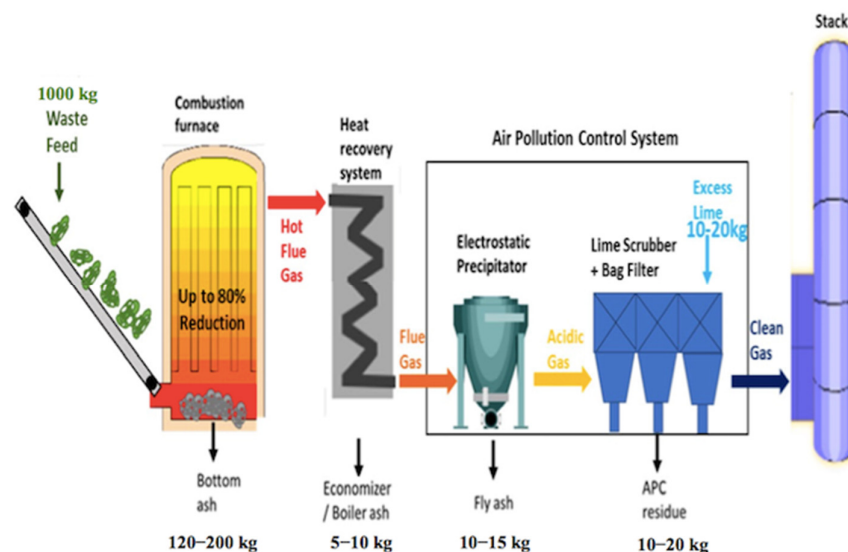


Figure 1. The mass fraction of residues in MSW incineration.

According to [16,67], these residues may differ in their composition and leaching behavior. Some authors [25,68,69] have considered fly ash (FA) and APC as a similar waste with similar waste content. Contrastingly, [16,67] have shown the probability of managing FA and APC individually. Until now, there has been such consensus on the management of these residues.

The emission of heavy metals (HMs) during the incineration of MSW is affected because of several reasons, such as the type of incinerator, sources of MSW, combustion temperature and mainly the composition of MSW [70,71]. Later on, the distribution of HMs in BA and FA are mutually resolved on the basis of physiochemical properties of HMs and their compounds.

The content of heavy metals is found higher in fly ash than that of bottom ash, as can be seen in Tables 4 and 5, which enlist the amount of heavy metals for fly ash and bottom ash. The composition of incineration ash depends strongly upon the kind of waste and the share of other waste (e.g., paper, plastic, metals, food waste) present in the waste. Paper and plastic are the source of generating Cd, Cr and Pb in southern parts of China; the abundance of paper and plastic in MSW is in higher quantities compared to other parts of China.

Table 4. Heavy metal content (mg/kg) in MSWI fly ash (FA).

Metals Ref:	Fly Ash [72]	Fly Ash [73]	Fly Ash [74]	Fly Ash [75]
Ag	31–95	ND–700	N/A	N/A
As	31–95	15–751	93	N/A
Ba	920–1800	88–9001	4300	539
Cd	250–450	5–2211	470	95
Co	29–69	2.3–1671	N/A	14
Cr	140–530	21–1901	863	72
Cu	860–1400	187–2381	1300	570
Hg	0.8–7	0.9–73	N/A	N/A
Mn	0.8–1.7	171–8500	1600	309
Ni	95–240	10–1970	124	22
Pb	7400–19,000	200–2600	10,900	2000
Se	6.1–31	0.48–16	41	N/A
Zn	19,000–41,000	2800–152,000	25,800	6288
Sn	1400–1900	N/A	N/A	N/A
Sr	80–250	N/A	433	151
V	32–150	N/A	37	N/A

Table 5. Heavy metal content (mg/kg) MSWI bottom ash (BA).

Metals Ref:	Bottom Ash [76]	Bottom Ash [73]	Bottom Ash [77]	Bottom Ash [78]
Ag	4.1–14	2–38	8.5–10.7	N/A
As	19–80	1.3–45	209–227	13
Ba	900–2700	47–2000	1104–1166	N/A
Cd	1.4–40	0.3–61	6.8–7.8	3
Co	<10–40	22–706	49.6–53.1	N/A
Cr	230–600	13–1400	323–439	900
Cu	900–4800	80–10,700	4139–4474	500
Hg	<0.01–3	0.003–2	N/A	2.6
Mn	<0.7–1.7	50–3100	869–894	280
Ni	60–190	9–430	216–242	180
Pb	1300–5400	98–6500	2474–2807	2700
Se	0.6–8	ND–3.4	230–265	N/A
Zn	1800–5400	200–12,400	4261–4535	600
Sn	<100–1300	N/A	N/A	960
Sr	170–350	N/A	N/A	N/A
V	36–90	N/A	N/A	N/A

The waste from paper may carry higher amounts of fillers such as lime and kaolin. Waste from the food may contain a considerable quantity of NaCl. In some of the big cities of China, the composition of waste is similar to that in Western Europe and comprises a higher share of paper and plastic. China's waste stream is growing faster in paper, plastics and multi-laminates [38]. However, the composition of MSW in China is far different from that of the western industrialized world. The case study by [18] explains the difference on the basis of heavy metal content in FA, BA and APC, and, according to [21], the content of Zn and Pb in China is lower than other countries, which can be seen from the Table 6. This difference may be because of operational and geographical conditions but until now there has been no such relevant information available to justify the differences. However, [38] elaborates that the high content of Zn and Pb may be because of the abundance of high waste fractions (batteries, colored containers, plastics, ceramics, etc.).

Table 6. Heavy metal Pb and Zn content (mg/kg) in MSWI fly ash (FA) and MSWI bottom ash (BA).

Country	Pb (FA)	Zn (FA)	Ref:
China	782.6–9901	2088–14,129	[79,80]
Japan	1878	8347	[81]
Korea	910–3600	5200–8640	[82,83]
Denmark	7400–19,000	19,000–41,000	[76]
Germany	13,600	49,100	[84]
Spain	5461	9382	[85]
Netherlands	5300–26,000	9000–70,000	[86]
Country	Pb (BA)	Zn (BA)	Ref:
China	647–2223	3378–3979	[87,88]
Japan	140–2700	1500–5000	[82,89]
Korea	750–1000	3100–4405	[83,90]
Denmark	1300–5400	1800–6200	[76]
Italy	1115–1473	4516–21,344	[91]
Spain	2700	600	[78]
Netherlands	100–13,700	1000–7000	[86]

Several researchers have studied and reviewed the BA, FA and APC's data on the basis of their chemical composition. The research on the occurrence of heavy metals in MSW incineration ash is rarely investigated in China; the literature available shows the difference in content because of MSW composition and generation in different parts or regions. The temporal and spatial trends in heavy metal contents of fly ash studied by [10] also agrees with the fact that variations in the content of heavy metals are because of socioeconomic conditions across China. Furthermore, after depicting the temporal trends in the contents of heavy metals in MSW incineration fly ash over the past 15 years, it was concluded that the contents of heavy metals in MSW incineration fly ash could be considered as roughly constant between 2003 and 2017 [10]. The heavy metal content determines not only the choice of treatment option, but also the possible behavior of the ash during landfill or reuse.

4. Treatment of Incineration Fly Ash

The presence of heavy metals and soluble salts classifies fly ash as a hazardous material, which requires treatment prior to disposal or utilization. The focus of recent treatment techniques/technologies is to reduce the environmental threat posed by the hazardous content of fly ash. The treatment of incineration fly ash would not only ensure the environmentally friendly applications and economical consideration of incineration fly ash but also serves as the alternate to the existing scarce resources. This section summarizes the treatment techniques of fly ash developed

in recent years. The treatment techniques/methods and principles of treatment of incineration ash are discussed below in Figure 2.

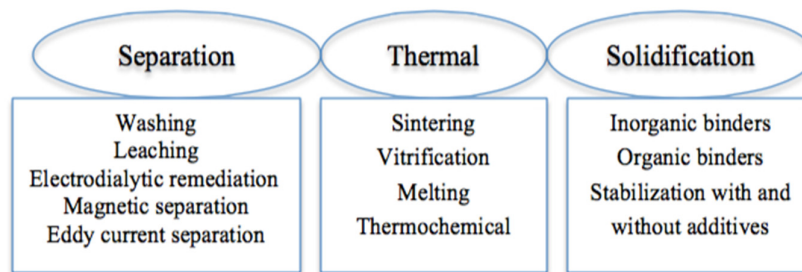


Figure 2. Shows the treatment techniques of incineration ashes.

4.1. Separation Process

Separation processes are mainly used to separate particles with similar chemical and physical properties from the bulk materials.

4.1.1. Washing

Washing treatment is aimed to efficiently reduce and eliminate chlorides, alkaline, soluble salts and heavy metals from incineration ash by exercising a liquid solution, commonly water or acid [21,22]. Water washing processes can be carried out in one step or several sequential steps. Several studies have shown that the removal of chloride is very rigorous during the first step and according to [92] lower removal efficiencies were obtained in later steps. In context to that [93–98] supported involvement of water for effective removal of chlorides during washing methods. As per the literature review by [16], water washing can remove up to 90–95% of the chlorides and the final product is improved in quality for further utilization. Strong concerns were raised by [98] for water washing because of its disadvantage to possible release of heavy metals (Cu, Pb, and Zn) in the leachates. According to [99], involvement of chemical additives or pH control may overcome this issue. Acid washing involving sulphuric acid and phosphoric acid has shown good results for removal of heavy metals like Pb, Zn and Cu [99]. Contrastingly, according to [100], H_3PO_4 is more effective than sulphuric acid. According to [95,96,100], operational parameters such as liquid-to-solid ratio, stirring speed, temperature and time can play important role in reduction of chlorine content in incineration ash during washing method. For example, higher liquid-to-solid ratios will show better results. So far, no consensus on washing parameters has been found in studies. In general, [21,22,94–96,99–101] have pointed out the drawbacks of washing treatment because larger volumes of produced water need to be treated prior to discharge because of contamination. However, recent legislation issued by the National Environmental Protection Standards of the People's Republic of China (HJ 1134–2020) in August-2020 demands a proper treatment prior to discharging the contaminated water under section GB 8978, which says the wastewater generated during the fly ash treatment process should be returned to the process first for recycling or comprehensive utilization. The wastewater generated during the treatment, if discharged directly to the environment, shall meet the requirements mentioned under section GB 8978-Integrated Wastewater Discharge Standard.

4.1.2. Leaching

The leaching process is meant to extract heavy metals from incineration ash and later recover them from the leachate solutions. According to [16,22,102–104], the heavy metal leaching process depends strongly on the kind of extraction solvent, pH and liquid-to-solid ratio. In a leaching solution, an increase in pH may hinder the leachability of heavy metals possibly because of the formation of insoluble hydroxides. Whereas chelating agents are independent of pH [105–107], the study by [105] shows the recovery of heavy metals such as Zn, Pb, Cr and Cu by using chelating

agents. The other investigation by [107,108] compared the efficiencies of EDTA, HClO_4 , HCl , NH_4Cl , NH_4NO_3 and various organic acids with usually used mineral acids and water. According to their investigations, strong mineral acids were effective in the leaching of heavy metals compared to organic acids. In particular, Cu and Pb were effectively removed by using EDTA, whereas NH_4NO_3 was seen as effective for Cu. The question is how these metals can be recovered during the process. The authors of [21] give an overview of how the treatment reagent affects the leaching behavior.

The biological reagents used for heavy metal extraction are acid-resisting and heavy-metal-resisting strains. After proper culturing, they can extract heavy metals from fly ash within the suitable conditions [109]. There are several strains related to this, such as autotrophic thiobacillus species [110] and heterotrophic *Aspergillus niger* [111]. While investigating the biological leaching of fly ash by *Aspergillus niger*, [112] discovered that calcium oxalate precipitate affects bioleaching through the weakening of molecule combination in fly ash, thus facilitating the release of heavy metals. The study conducted by [113] using thiobacillus species showed that the effective bioleaching was achieved in the presence of up to 8% fly ash, within a pH value less than 4. The pH, the amount of fly ash and the sulphide content can influence the efficiency of heavy metal extraction from fly ash by bioleaching with thiobacillus species. Considering chemical leaching and bioleaching, [114] showed that both methods could effectively leach Mg and Zn (leaching rate > 90%), Al and Mn (leaching rate > 85%), and Cr (>65%), Ga (>60%), and Ce (>50%) from fly ash. The use of chemical reagents led to more effective leaching of Cu, Fe, and Ni, whereas biological reagents promoted the leaching of Nd, Pb, and Co. Compared to chemical leaching, bioleaching can reduce the H₂SO₄ dosage by half, but requires more processing time.

4.1.3. FLUWA and FLUREC Process

Municipal solid waste incineration fly ash contains huge potential for recyclable metals such as Zn, Pb, Cu and Cd. The latest “Environmental Protection Law of the People’s Republic of China” and “Law of the People’s Republic of China on the Prevention and Control of Environmental Pollution by Solid Waste” under section GB/T 30810 urge proper treatment of fly ash to remove and recover precious metals. Such an initiative for the effective separation and recovery of metals from fly ash was taken in Switzerland in 1997, acidic fly ash leaching (FLUWA process) in 1997, as described in detail by [115] is shown in Figure 3.

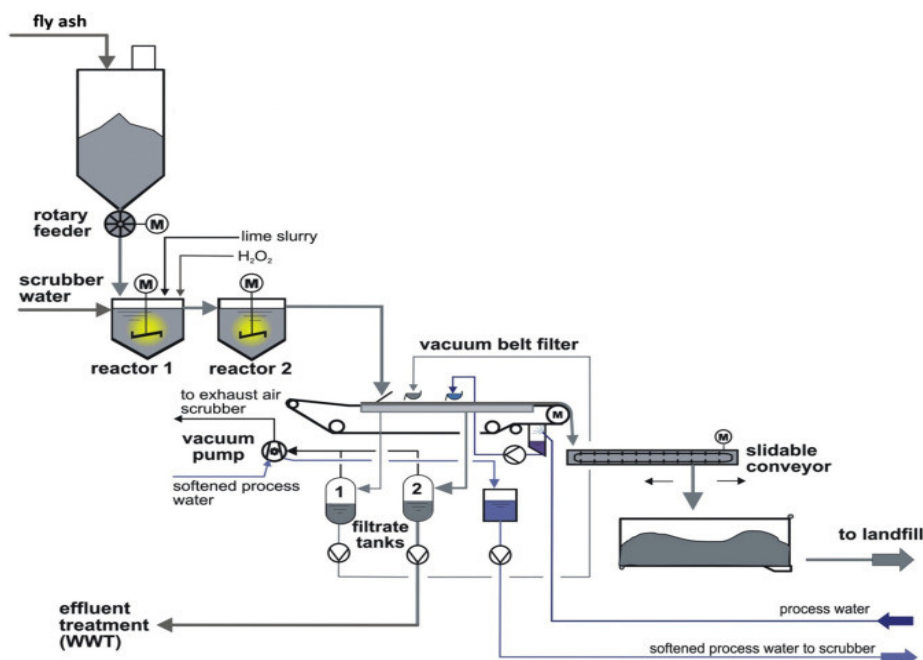


Figure 3. Basic concept of acidic fly ash leaching (FLUWA)

To date, more than 60% of fly ash in Switzerland is treated according to the FLUWA process. According to [116], during the FLUWA process, fly ash is leached using both acidic and neutral scrub water in a multistage cascade as shown in figure. Prior to fly ash leaching, mercury dissolved in the acidic and neutral scrub water is separated by a selective ion exchanger. According to [117], the extractability of metals such as Zn, Pb, Cu and Cd is mainly dependent on the characteristics of the FA, acidity of the scrub water, L/S ratio, redox-potential, temperature and leaching time. Furthermore, [117] stated that, depending on these parameters, 60–80% Zn, 80–95% Cd and 50–85% Pb and Cu can be extracted by the FLUWA process. During the FLUWA process, the addition of hydrogen peroxide helps the redox-sensitive metals (Cu, Pb and Cd) to stay in solution. Likewise, Fe^{2+} is converted into Fe^{3+} , which precipitates as Fe-hydroxide and is accumulated in the remaining filter cake. After sufficient extraction time, the suspension is separated by vacuum belt filtration into a metal depleted filter cake and a metalliferous filtrate.

The FLUWA process provides the basis for extended methods such as the FLUREC process. This process allows for the recovery of high-purity zinc ($\text{Zn} > 99.995\%$) from the heavy-metal-enriched filtrate. The FLUREC process was implemented at MSWI plant Zuchwil, Switzerland, in 2012, where about 300 tons of Zn can be recovered annually. The FLUREC process can be seen in Figure 4. For further knowledge and research work about FLUWA and FLUREC, the research work of [115–118] can be more useful.

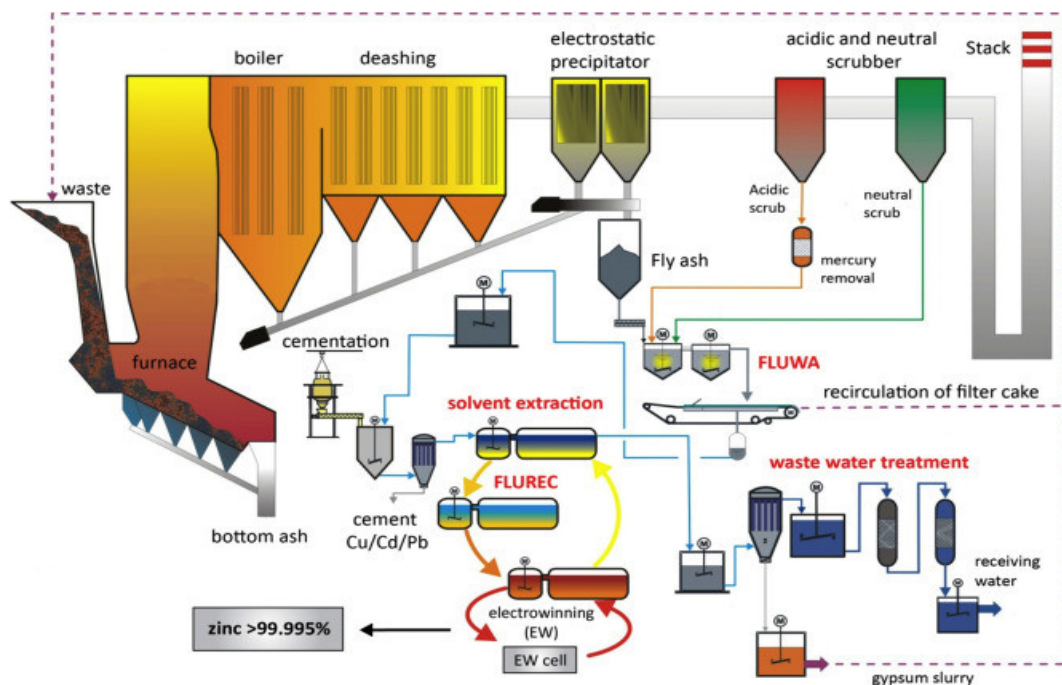


Figure 4. Process diagram of waste incineration, fly ash leaching (FLUWA) and recovery of recyclable materials (FLUREC).

4.1.4. Electrodialytic Remediation

Electrodialytic remediation is a technique that combines electro dialysis with electro migration of ions for the removal of heavy metals from fly ash. The basic principle of electrodialytic remediation technique involves suspension of incineration fly ash in a solution placed in electrodialytic cells. An electric current is then applied across the cells, which enables the metal ions to migrate towards the electrodes based on their charge, as shown in Figure 5.

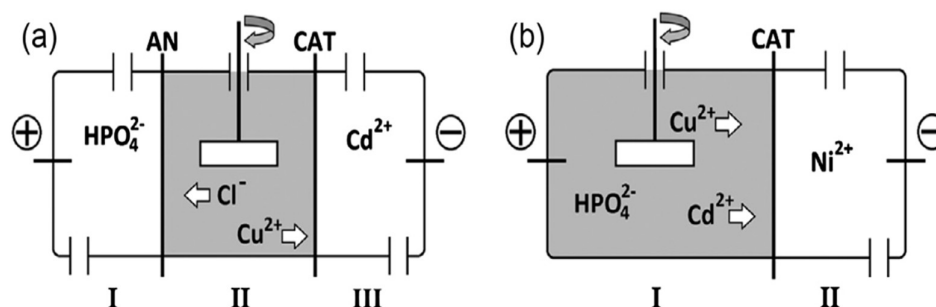


Figure 5. The (a) three-compartment and (b) two-compartment setup for the electrochemical remediation of ash.

The ion-exchange membranes placed between two cells prevent the metal ions from reaching the electrodes. Instead, they are accumulated in “concentration” compartments and are subsequently removed. There are several studies carried out by [119–122] to extract heavy metals with and without assistance of assisting agents (deionized water, 2.5% NH_3 solution, 0.25 M citric acid and 0.25 M ammonium citrate). The study by [119] used multiple experiments through three-compartment electrochemical cells and two-compartment electrochemical cells to remove the heavy metals, and chlorides from incineration residues according to the results of the study removal of heavy metals were generally low. The operational parameters may help to achieve somehow better results; like with low pH, higher amounts of heavy metals can be removed. According to [119–122], removal of heavy metals was shown to be dependent on current, density and the type of incineration residue rather than type of electrochemical cells. Although [119,120] have shown optimized removal of heavy metals with the support of ancillary agents but it leaves a liquid waste which is even harder to handle because that liquid waste further necessities treatment prior to dispose or reuse [123]. Furthermore, research studies should be carried out to minimize the time consumption, which can lower the demand of continuous energy.

4.1.5. Magnetic Separation

Magnetic separation techniques are one of the standard frameworks to separate materials based on their magnetic properties. The magnetic separation technique includes drum magnetic separator Figure 6 and cross belt separator Figure 7 techniques. The working principle of the drum separator is that the pulp flows into cell body from the pulp box, and, under the water flow of the feeding spray pipe, the mineral grain will go into the feeding area of the cell body in the loose condition. In the magnetic field, the magnetic ore particles will have a magnetic gathering and form a magnet group or magnet chain, which is influenced by the magnetic force inside the pulp and moves to the magnetic pole and finally is absorbed onto the cylinder. The non-magnetic and weak magnetic minerals are kept in the pump and are discharged from the chute with the pulp to become tailing.

In the case of a cross belt magnetic separator, the materials are uniformly fed to the belt. When materials go through the permanent magnetic drum, the non-magnetic and low-magnetic particles will leave the belt surface because of the centrifugal force and gravitation. Meanwhile, strong magnetic particles are absorbed to the belt by magnetic force and then fall into the magnetic trough when the belt leaves the magnetic pulley [124].

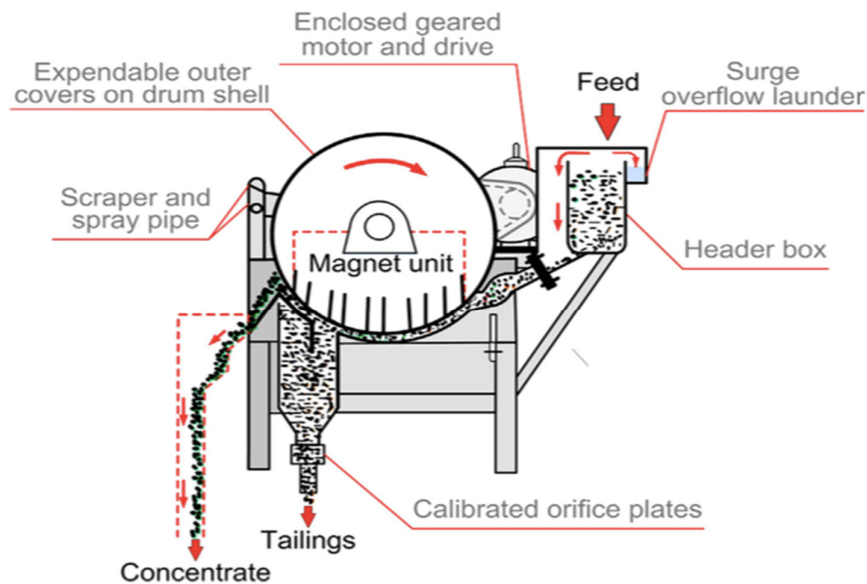


Figure 6. Shows a typical drum separator.

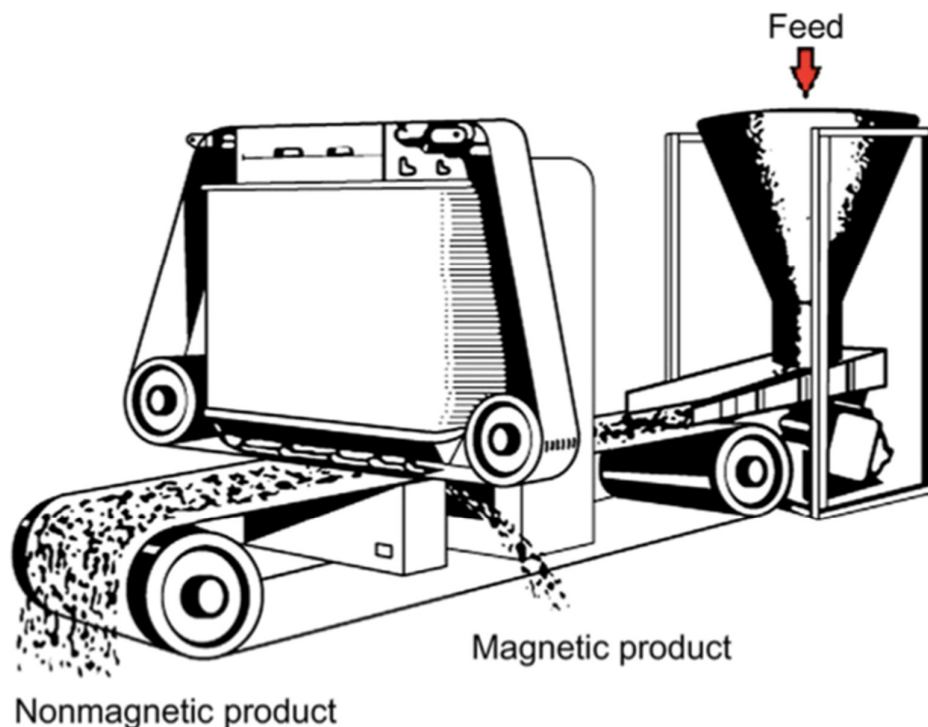


Figure 7. Cross belt separator.

The magnetic density separation (MDS) works based on the difference in density between aluminum and other non-ferrous metals in a magnetic gradient field [125]. The processes developed for the treatment of MSW incineration residues for basic physical separation techniques and specifically magnetic separation are not generally investigated for the material recovery from MSW incineration residues. Only two patents were found using magnetic separation for the extraction of ferrous materials from MSW incineration residues [126]: the first patent by [127] applied separation at the end of the treatment; the second patent by [128] described the magnetic separation steps used in a mixture of water and fly ashes. Magnetic separation may lead to the recovery of a valuable

ferrous fraction, as in the case of bottom ash, and, ideally, to a relative decontamination of the non-magnetic fraction. The effectiveness of magnetic separation reported by [129] were up to 83% for ferrous metals. Other than the recovery of ferrous material, the magnetic separation was also used for the characterization of residues by [130]. According to [130], the fly ash was first washed, then sieved and finally separated according to their density. The magnetic separations on boiler fly ashes were studied by [126], and the dry magnetic separation and wet magnetic separation were used in their study. The main drawback of such a wet separation lies in the partial dissolution of the residues, especially the heavy metal compounds leaching, once they are in contact with water. This drawback may become an advantage if the chlorides' dissolution is considered, since chlorides generally have to be removed from MSWI fly ashes. Magnetic separation could then stand for a washing step. In any case, the solution resulting from this separation should be treated before its discharge. From an economical point of view according to [126], magnetic separation appears to be beneficial only for big treatment plants. It could thus be interesting to reduce the residues to be landfilled by employing another recovery step or by improving the ash treatment.

4.1.6. Eddy Current Separation

The eddy's current separation (ECS) technology is a method of separation of nonferrous fraction. However, existing ECS designs are limited by particle size [131]. The limitations and principles of ECS Figure 8 are well explained by [132]. ECS demands an accurate calibration, keeping in mind the size of materials to be separated, and, when ECS is used on coarse and fine particles, the different rotation speed of rotors may give better results. For example, for particles larger than 5 mm, a rotation of 2000–3000 rpm, typical for standard ECS, is appropriate. For non-ferrous metals in ashes, fine fractions <5 mm may achieve high recovery efficiency.

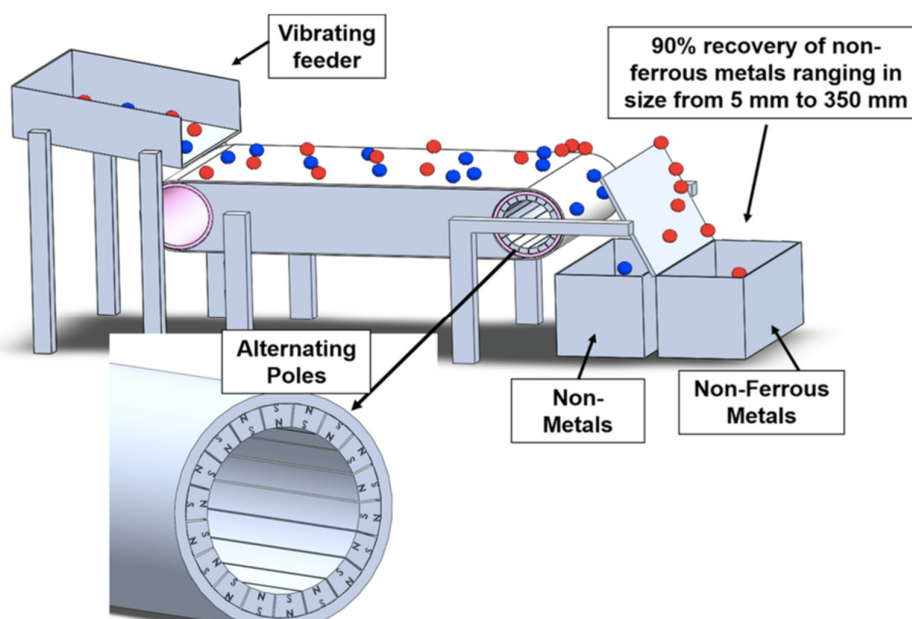


Figure 8. Description of a typical belt-driven rotary drum design of an eddy current separator.

There are other techniques present in literature but those are not developed at a full scale. Such is the case with magnus ECS, backward operating ECS and with wet eddy current separators [133–135]. The Magnus ECS is based on the “Magnus effect”: a spinning particle moving through a fluid experiences a force perpendicular both to its direction of motion and to its axis of rotation. This effect can be used to recover small NFe metal particles from the bulk stream (both wet and dry) [133]. A backward operating ECS is a standard separator whose magnetic drum rotates “backwards”.

According to [134], if it becomes difficult to separate small metal particles from the non-metal stream when the magnetic drum rotates in “forward” mode, the yield improves drastically when it rotates in the opposite direction. In a wet ECS, the water makes it possible to glue all the particles to the belt surface. For small particles, this adhesive force has the same order of magnitude as gravity. Without the action of the rotor, therefore, virtually all particles would stick to the belt and end up in the non-metal fraction. The available literature on the subject is still in development. Reviews on the technical applications of eddy current separators have been published by [136,137] and more recently by [138,139]. However, the number of extensive and critical reviews on this topic is limited to date.

4.1.7. Shaking Table

A shaking table is a common piece of equipment in a mineral processing plant. It is widely used for gold, silver, zinc, lead, tin, tungsten, iron, manganese, and coal separating, etc. It has features of a high enrichment ratio, high separation efficiency and an easy operation. The shaking tables or wet tables are considered density separation techniques that enable the separation of heavy metals particles from other lightweight particles [80].

The structure of shaking tables contains a sloping plank with ribs on the surface. The slurry flows perpendicular to the ribs and the table swings parallel to the ribs. Mineral particles were fed from the table feeding slot, at the same time, feeding water tank fed water crossly. Under vibration wave, mineral particles move breadthwise and lengthways along the surface. They would be separated in different layers and discharged from different positions as concentrate, middlings and gangue, as shown in Figure 9. It has been successfully employed on an industrial scale for the separation of metals such as tin, copper, gold, lead, zinc, tungsten, etc., of a size of 50 μm –2 mm [96,97]. However, there is little research work or literature available on shaking tables.

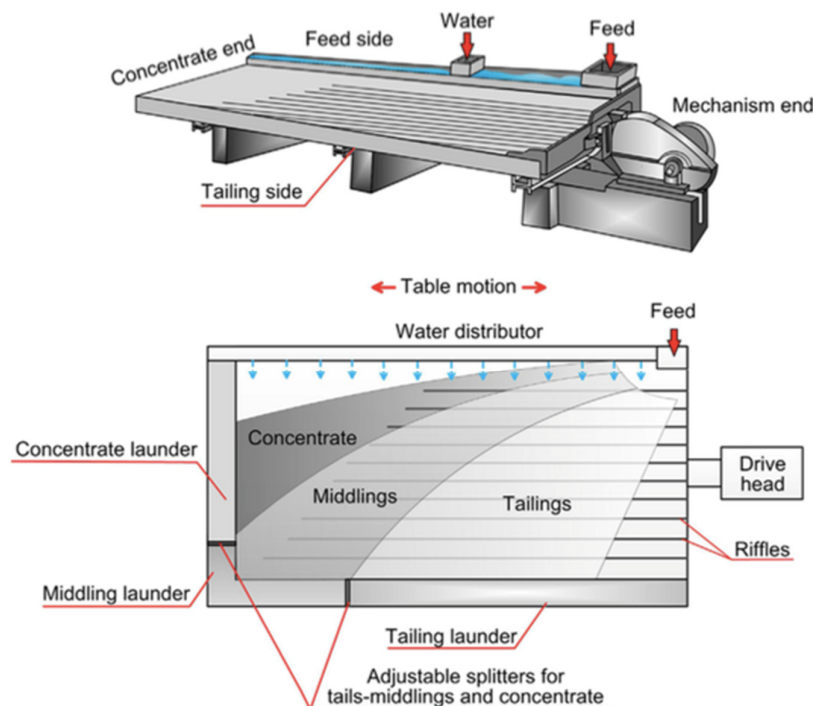


Figure 9. Shaking table.

4.2. Stabilization/Solidification (S/S)

Stabilization/solidification are widely employed processes for MSWI residues. S/S processes tend to deal with waste residues whose properties (physical, chemical and mechanical) favor reducing the leachability of contaminants from waste [23]. The solidification/stabilization indicates

the processes, which involve additives or binders to either physically or chemically immobilize the hazardous content in waste [22]. The goal of the stabilization process is to decrease the toxicity and solubility of contaminants to make it less soluble and less toxic with or without solidification. Contrastingly, in solidification in order to lessen the leachability and to immobilize contaminants, binders such as cement are usually added. This process reduces the mobility of contaminants in the treated material through encapsulation. The best approach to solidification/stabilization in practice is to initially involve chemical stabilization and then solidification of waste [25]. The addition of inorganic binders such as Portland cement for solidification/stabilization has been in practice for a long time in several parts of the world. Numerous research studies have shown that cement solidification can fix the heavy metals [140–142]. However, heavy metals in cement-solidified products are not stable in the long term and also the exposed environment may increase the leaching ability of heavy metals. Considering the long-term leaching characteristics of solidified products, an effect on heavy metals is shown by aging or carbonation [143]. The study carried out by [144] reported that supercritical carbonation can enhance/accelerate the aging characteristics of solidified products. The study conducted by [145,146] used supercritical carbonated cement product to study the heavy metals' leaching characteristics and found that supercritical carbonation promoted the leaching of Cu and Pb. Furthermore, not all heavy metals can be fixed through solidification such as Hg. In the study of [147], it was shown that Hg released by the solidified products reaches 8.51 ng/g–18.48 ng/g, resembling that of raw fly ash. Additionally, the increase in temperature and water content also promoted the release of Hg. According to [25], this process has a drawback, which makes it ineffective for the treatment of soluble salts and in return long-standing leaching will bring an environmental issue. During this process, the significant increase in volume of waste may occur, which may affect shipping and the landfill costs [21].

The introduction of new stabilization technique by [148] is shown to be capable of destroying the toxic content, reducing the heavy metal reactivity and solidifying the hazardous waste beyond the leaching limit. This four-stage technique involved: elimination of alkali chlorides by solidification; addition of phosphoric acid; calcination and solidification with cement. [22]. The combined treatments, such as washing-immobilization treatments, can possibly remove noteworthy quantities of sulfate and mainly chloride from incineration fly ash. This treatment would encourage the development of the hydrate phase, which will change the heavy metals into less reactive forms. Afterward, the water remained as waste after washing treatment could be treated as explained by [149]. This treatment can not only help to treat the incineration fly ash and stabilize it in cement with any issue of heavy metals leaching but can also reduce the cost to make this treatment economical.

4.3. Thermal Treatment

The thermal treatments are aimed to acquire stabilization in the residue, which is resistant to leaching and creating a residue for utilization. Thermal treatment thus ensures the maximum detoxification of residue. In contrast to this, [149] reported the threat by the presence of alkali chlorides, sulphates and volatile compounds during the thermal treatments. Therefore, these processes involve high temperatures, which increases the cost but creates a possibility for the removal of pollutants and toxic materials [21,149]. The research contribution by [150] reported an option to dodge heavy metal volatilization, achieving immobilization in a specific atmosphere, use of additives, temperature and pretreatment. As thermal treatment undergoes an air control system, [151] reports to take measures for cooling methods because it may influence the characteristics and leaching behavior of residues. The major thermal treatments are grouped as sintering, vitrification, melting and thermochemical treatment.

4.3.1. Sintering

The sintering can be defined as the temperature-induced coalescence where densification of porous solid materials occurs below the melting point. The treatment temperatures involved in the sintering process are generally between 700 and 1200 °C [33,45,151–154]. Sintering involves heating the fly ash to the point at which particle bonding occurs and chemical phases in the fly ash reorganize,

then the solid phase transformation and recrystallization occurs, which results in a product with lower porosity and with higher density and strength. The sintered product with lower porosity tends to eliminate the chances of leachability from the final product.

Several studies have been performed to investigate the effects of sintering on incineration ash, incineration fly ash, incineration bottom ash and mixture of FA and BA [104,107–109,152–156]. According to the research study carried out by [157], it was found that the sintered heavy metals (Zn, Cr, Pb, and Cu) were mainly present in an oxidized form. Except Pb, the leaching concentrations of most of the heavy metals were within the limits of the national standard. No significant increase was seen in the leaching concentrations of heavy metals even after 30 days of aging of sintered products. The operational conditions, such as sintering time and treatment temperature, may have an influence on the solidification of heavy metals. Therefore, to establish an overall pollution toxicity index (OPTI), a study was conducted by [158] for the evaluation of heavy metal pollution. According to experimental results, higher sintering temperature led to more volatilization of heavy metals, and the lowest value for the overall pollution toxicity index (OPTI) was at 1040 °C. The authors of [159] conclude that, at lower temperatures and shorter times, higher integrated control efficiency conditions of heavy metals were achieved. Some suggested various pretreatments such as water washing, ball milling, sieving and pelletizing of the ash [154,160–163]. These pretreatments are comparable to traditional ceramic processing as the glass-ceramics produced by the sintering process often undergoes pre-melting [164]. The study on untreated fly ash by [155] proved to be ineffective for the production of concrete aggregates, because of the adverse chemical characteristics of these fly ashes in terms of chloride, sulfate, and vitrifying oxide contents. Contrastingly, to counter the presence of soluble salts, water-washing treatment can enhance the chemical properties of sintered products, which, according to [152], satisfies the basic requirements for concrete aggregates. Water washing treatment is not only effective for the removal of metal chlorides but also serves as the effective tool for reducing the sintering temperature and treatment time. Although water washing lowers the power consumption during the sintering, it would require further treatment for wastewater generated.

4.3.2. Melting/Vitrification

The melting/vitrification is derived from the metallurgical industry. The melting and vitrification of fly ash involves higher temperature ranges from 1000 to 1600 °C. In the melting process, the organic part of the MSWI fly ash is decomposed, whereas the inorganic part is transformed into a glass slag via crystal phase transformation [21,23]. In contrast to this, vitrification is a process of melting residues with glass-forming additives or other waste solids to create a homogenous liquid phase, and then cooled to form an amorphous, homogenous single-phase glass. In the melting process, the temperature required for the melting process depends upon the composition of fly ash [21–26]. The melting/vitrification process generally involves fossil fuel and electric heating. The fuel burning melting/vitrification involves fuel oil, coke and gas. In this regard, a detailed study is presented by [82], which summarizes the properties of fuel burning and electric melting/vitrification furnaces and systems. The volatilization of heavy metals during melting/vitrification processes is studied by several researchers [165–169]. The low volatile heavy metals are transformed and solidified in the dense lattice structure of melted slag, thereby reducing their leaching rate. However, the easily and semi-volatile heavy metals in fly ash exist in the secondary fly ash during the melting process. The volatilization of these heavy metals is related to the high content of Cl in the raw fly ash. According to the research study of [167], the volatilization of Cl in a fly ash melting furnace is directly related to the molar ratio of $\text{Cl}/(\text{Na} + \text{K})$ in the raw fly ash sample. A higher $\text{Cl}/(\text{Na} + \text{K})$ molar ratio promoted the volatilization of Cl, leading to the detection of more PbCl_2 in the secondary fly ash. Similarly, the finding of [168] shows that low volatile heavy metals (Cr, Ni) were better solidified in the melting slag. Contrastingly, the easily and semi-volatile heavy metals such as Hg, Cd, Pb, and Zn are difficult to stabilize. Plasma is currently an area of in electric melting/vitrification. The high-energy-density and high-temperature of a plasma flame can promote the melting/vitrification reactions and reduce the residence time of MSWI fly ash. Related

research [169–172] has shown, after plasma melting/vitrification treatment, the volume reduction of MSWI fly ash. The volatilization and leaching characteristics of heavy metals before and after plasma melting/vitrification [169–171,173] determined that most heavy metals sealed in vitreous slag are Cr, and Ni, which are low volatile heavy metals. In comparison, Cd and Pb exhibit a strong volatile property. The leaching of other heavy metals in melting/vitrification slag such as Zn and Cu is significantly lower than that of the original fly ash, which proves that vitreous slag can seal the heavy metals from fly ash.

4.3.3. Thermochemical Treatment

China is increasingly shifting towards incineration because of its small land footprint, huge waste volume reduction and energy recovery. However, incineration residues such as fly ash and bottom ash contain toxic substances, heavy metals, which become problematic if disposed/reused without treatment. There are two main options to handle fly ash in terms of heavy metals: either reducing the heavy metals' leachability by stabilization or the recovery of heavy metals from fly ash. Thermal treatment of fly ash causes stabilization and the recovery of heavy metals depending on the treatment temperature. Thermal treatment contains three major ways (vitrification, fusion/melting, and sintering) to stabilize the heavy metals. The heavy metals stabilization/removal through vitrification, sintering and melting seems to be energy consuming compared to the removal of heavy metals by thermal treatment with the addition of chlorinating agents. Heavy metal chlorides exhibit a higher vapor pressure; thus, involvement of Cl helps to delay the condensation of these metal compounds during thermal treatment. This situation quickens the removal of heavy metals. Accordingly, for the effective removal of heavy metals from MSWI fly ash, the addition of a certain amount of chlorinating agents with MSWI fly ash can lead to an effective removal of heavy metals. Typically, the metals of interest are Zn, Cu, Pb and Cd. The reaction mechanism of chlorinating agents described by [174–177] has shown promoting effects on the removal of heavy metals. The volatility of trace elements is divided into several categories by research studies according to their nature of volatility as easily volatile (Cd, Pb), semi-volatile (Cu, Zn) and low volatile (Cr, Ni). Cadmium and lead tend to be volatilized during the combustion process. Table 7 shows an overview of previous studies and the experimental parameters used for the removal of heavy metals from fly ash. According to [178], the part of heavy metal removal can be improved by wetting the mixture of fly ash and adding the chlorinating agents with water and later drying it to improve the contact area. In contrast to this, [179,180] states that pelletizing the fly ash plus adding the chlorinating agents mixture prior to treatment may decrease the entrained particles, which can result in a low removal rate of copper. The availability of chlorine and the formation of metal chlorides may strongly increase the volatility of metals. Semi-volatile metals like Zn may act contrarily depending on the conditions of the waste to energy (WtE) plant. If the chlorine is available, the Zn can volatilize as atomic Zn at reducing conditions or as $ZnCl_2$, whereas ZnO is easily formed under oxidizing conditions and has very low volatility. Therefore, it's essential to use chlorinating agents to increase the volatilization of these trace metals from fly ash. There are several chlorine additives used as can be seen in Table 7 to achieve the required vaporization. Amongst all the chlorine additives, $MgCl_2$ has shown effective results. The removal of Cu, Zn, Pb, and Cd by different studies using different chlorinating agents is 80–95%, 75–97%, 99% and 99%, respectively.

Table 7. Overview of previous studies and their experimental parameters for the removal of heavy metals from fly ash.

Incineration Ash Type	Chlorinating Agents	Heating Source	Temperature	Metals Studied	Time (Minutes)	Reference
Fly ash	MgCl ₂	Muffle furnace	1100 °C	Cd, As, Pb, Zn, Cu, Cr, and Ni)		[177]
Fly ash	CaCl ₂	Rotary reactor	950, 1000 and 1050 °C	Cd, Pb, Cu and Zn	10, 15 and 25	[181]
Fly ash	HCl gas, CaCl ₂ ·2H ₂ O, MgCl ₂ ·6H ₂ O, MgCO ₃ + HCl	Quartz glass reactor, laboratory scale gas-tight rotary furnace	1000 °C	Cd, As, Pb, Zn, Cu, Cr, Mo and Ni)	30	[182]
Fly ash	CaCl ₂	Rotary kiln and FBR	800, 850 and 900 °C	Cd, Pb, Cu and Zn		[180]
Fly ash	MgCl ₂ ·H ₂ O	Tube furnace	900–1000 °C	Cu, Zn		
Fly ash	CaCl ₂	Quartz tube furnace	950 °C	Zn	240	[183]
Fly ash	CaCl ₂	Laboratory-scale rotary kiln	900–1000 °C	Cd, As, Pb, Zn, Cu, Cr, and Ni)	30	[184]
Fly ash	MgCl ₂	Pilot scale indirectly heated rotary kiln	900–1100 °C		40	[185]
Fly ash	Mixture of chlorinating agents	Directly heated rotary kiln	900–1100 °C	Cu, Zn	30	[186]
Fly ash	MgCl ₂ ·6H ₂ O	Tube furnace	900–1000 °C	Cu, Zn	30	[157]

There are several factors, which can influence the removal of heavy metals. The higher treatment temperature and longer residence time can help to achieve a better removal of heavy metals. However, the temperatures exceeding the melting range may affect the evaporation. The lower temperatures affect the residence time greatly [174–180]. There are some processes commercially available such as the ASH DEC process for the removal of heavy metals from MSWI fly ash and sewage sludge ash. Similarly, [174–176,179–181,183–186] have formed the basis for the ASH DEC process, as it already existed at a commercial level for the recovery of phosphorus from sewage sludge ash. In this model, ash is well mixed with water and chlorinating agents (MgCl₂ or CaCl₂, KCl). At first, the ash is treated at 300 °C for about 30 min and then at 900–1100 °C for a maximum of 60 min. During this process, the metal chlorides formed are captured in the flue gas cleaning system and later recover with the following treatment.

5. Possible Application of Incineration Fly Ash

The emergence of recycling philosophy has encouraged the recovery, recycle and reuse of the incineration ash. Thus, the use of landfills for incineration ash disposal is decreasing year by year in many countries. Incineration ash is rich in heavy metals and soluble salts, which may appear as an

alternative resource to existing scarce and precious natural resources. Literature unveils the availability of huge information on the applications of waste [12,17,21–23,25]. Each application for fly ash reuse, recycle and recover comes with advantages, for example, the conservation of natural resources, elimination of waste and availability of zero-cost raw materials.

In this section, we mainly focused on identifying the processing suitability, performance, and environmental impact of incineration fly ash for its applications and finally, at the end of this section, Figure 10 summarizes the utilization sources, development stage and the limitations faced.



Figure 10. Use of fly ash, technical status and the limitations faced.

5.1. Construction Material

5.1.1. Cement

Cement Process/Production

The availability of lime, silicates and aluminosilicates in incineration ash matches the elemental composition of cement, thus making it possible substitute for cement raw materials [19–25]. The cement processing and production requires a huge amount of energy; therefore, cement production is also regarded as an energy-intensive process. The decarbonization of calcium carbonate into quicklime is the main energy consumer in cement processing, and each ton of clinker that is produced emits a similar quantity of carbon dioxide into the atmosphere. The incineration ash contains lime,

which can help in the reduction of carbon dioxide. However, there are a few technical issues that discourage the use of this application, such as higher chloride content, which can possibly affect the products' quality, and cycling effect in the cement kilns may become the basis of clogging and corrosion inside the heat exchangers. The presence of heavy metals in higher concentrations may also affect the chemical composition of cement and afterward creates an environmental burden. Therefore, pretreatment procedures are required to treat and remove the chloride and heavy metal content. Taking into account the hydration behavior of cement clinker, it is observed that the content of alkali metals increases the hydration and that the content of Zn, Cd and Pb impedes the rate of hydration of cement. The content of alkali metals in MSW incineration ash is reduced after washing pretreatment, and the hydration rate of treated MSW incineration ash clinker is lower than that of untreated MSW incineration ash clinker [187–191]. The use of MSW incineration fly ash may also serve as a material for lightweight concrete aggregate by handling through pellets. This could be used in non-structural forms, for example, as interior walls for insulating purposes [192]. Cement-solidified MSW incineration fly ash has been reported to be useful for safety reuse as artificial aggregate in Portland cement mortars, and the study conducted by [193] has shown low leaching levels of heavy metals, having a high compressive strength without any delay in mechanical strength improvement for the mortars incorporated into aggregates. Even though leaching is regarded as the main drawback or environmental concern for the application of MSW incineration ash, in Japan, a brand named Ecocement has made an attempt at involving large amounts of ash in cement clinker [188]. Furthermore, to expand and involve applications of incineration, ash requires pretreatment, and the quantity of incineration ash required to add to the chosen process must be sensibly controlled to achieve and ensure the required product quality and process safety.

5.1.2. Glass Ceramics

The ceramics industry produces pottery, porcelain and building materials (bricks, tiles, stoneware, etc.). The ceramic products are made of malleable and earthy materials such as clay, and higher temperatures play an important role in making them harder. To help retain shape and face the higher temperatures, inert materials such as quartz are used to confer the cohesion, plasticity and structural support of ceramic product. The silica is also used with a combination of melting agents and stiffeners for glazing ceramic bodies. According to [194], when a glaze layer is applied on a ceramic product and then fired, the glaze components are melted into a vitreous material. The ceramic industry utilizes a huge share of natural raw materials, and the incineration residues such as bottom ash (BA) and fly ash (FA) hold higher amounts of SiO_2 , Al_2O_3 and CaO , which enables them to substitute part of the natural raw materials for the ceramic production without pretreatment. However, fly ash (FA) contains high quantities of heavy metals and iron oxide, which may adversely impact the ceramic product. However, [195] suggests that this problem could be resolved by choosing an appropriate amount of incineration fly ash. The studies by [21–23,25] have presented a detailed review on ceramics.

5.2. Geotechnical Applications

5.2.1. Road Construction

Increasing urbanization and industrialization demands a transportation network of all-weather hard-surfaced roads/routes. The construction of roads requires lots of materials, one of which is sand or cement bases. Incineration ash is technically feasible for road construction and can serve as an alternative to sand and cement bases and sub-base [196], but the presence of heavy metals poses a threat to soil and underground water. The use of incineration ash as a raw material for the construction of roads serves two goals: firstly, it reduces the consumption of natural resources; secondly, it is a successful step towards “zero landfill” [197]. Usually, the main problem with using incineration ash is its leaching potential, which contaminates the environment, soil and water system. Several research studies ([198–202]) have been carried out to characterize the long-term consequences of leachate and its impact on the environment and reuse. According to [198], the leachate pH and

concentrations in major elements (Ca, Na and Cl) as well as in Al and heavy metals (Cu, Pb and Zn) quickly drop during the first 2 years to asymptotically reach a set of minimum values over 10 years. The authors of [198] propose continuous field experimentation to assess the environmental impact in different times and seasons that would allow for a better recycling strategy of incineration ash. According to their results, the Cu and Zn of the test road show a geochemical anomaly compared to the surrounding area. The Zn has low initial leaching, which will increase with time and will in long term exceed that of the test road and surroundings. In contrast to this, the leaching of Cu from the test road is initially high, but decreases over time and is, in the long term, below the values of natural weathering. Similarly, the study conducted by [185] shows an increase in Zn over time with the use MSWI ash. To avoid this problem, pretreatment and use of additives can play a crucial role.

5.2.2. Embankments

Embankments are primarily constructed of soil and stones to resist water for land reclamation and retaining walls. It has been a common practice to stabilize the soil with lime and cement to achieve geotechnical properties. This stabilization increases the shear strength and reduces soil compressibility. Incineration ash possesses pozzolanic characteristics, which becomes the advantage for reusing it in the construction of embankments. The incineration ash alone can be mixed with soil for its possible applications [203]. The main issue with constructing embankments with incineration ash is the same as using it in the construction of roads—the possible contamination of groundwater and soil. The studies by [204] reported the comparison of incineration ash stabilized with lime and cement. According to [204], the study carried out by [203] limited their focus only on the leaching of incineration ash, but they did not consider the aftermath for the soil system; this could have shown a precise indication of leaching behavior from embankments constructed with a mixture of incineration ash and soil. Pretreatment is suggested by [205] as a possible solution for leaching.

5.3. Adsorbent

With the economical stability and demands of people, an increase is seen in industries such as textile, leather, food, cosmetics, paper and printing to meet people's demands. However, these industries use dyes to color their products, which result in the generation of colored wastewater [206–208]. The adsorption technique is a broadly accepted and low-cost alternative to activated carbon to eliminate pollutants from dye. The treatment for the removal of dyes can be categorized as chemical, physical and biological treatments. A research study by [206–208] showed that adsorption is the most effective physico-chemical process to remove dyes. According to [202], the adsorption mechanism can be influenced by pH, particle size, adsorption dosage, and temperature and contact time. Although, MSWI fly ash has a lower content of SiO_2 and Al_2O_3 (15–30%), but different types of zeolites have been synthesized by utilizing the MSWI fly ash by fusion or hydrothermal process [209,210]. The study carried out by [211] involved converting MSWI ash to zeolitic-type adsorbent materials by using hydrothermal conversion. The converted MSWI ash has shown encouraging results for application as a sorbent towards several heavy metals. The performance was found to be much better than clinoptilolite natural zeolite; around an 80% reduction in heavy metal concentration was achieved through converted MSWI ash. The results of experiments conducted on MSWI fly ash reported by [212] from landfill leachate showed that that, upon increasing the dosage of MSWI fly ash, the rate of heavy metals removal increased in the leachate.

5.4. Agriculture

Enriching soil and fostering the plant growth application of fertilizers are commonly adopted worldwide. The main composition of fertilizers contains three main elements—nitrogen, potassium and phosphorus. Fly ash is regarded as the second largest source of potassium and phosphorus. This certifies that incineration fly ash can replace the commercial fertilizers and that there is hope for scarce natural resources.

Soil Amendment

The use of incineration ash as a potential fertilizer studied by [213] showed that plant growth in amended soils was twice the time of that in non-amended soils. In addition, incineration ash can substitute lime to reduce soil acidity. The authors of [214] determined the efficacy of incineration ash in agriculture using a liming agent. The ash characterization and chemical composition holds a decisive role in providing the required results. Incineration fly ash serves as a potential fertilizer, but it has also serious environmental issues. Regarding the impact of heavy metals, several research studies have pointed out their concerns [21–26]. The use of incineration ash increases the risk of salinity. Excess salinity in soil water can decrease plant available water and cause plant stress. Salinity becomes a problem when enough salts accumulate in the root zone to negatively affect plant growth. Excess salts in the root zone hinder plant roots from withdrawing water from the surrounding soil. The increase in salinity has resulted in a decrease in crop productivity [215]. To avoid the restriction of incineration fly ash as fertilizers, an appropriate pretreatment can play vital role in reducing the salt content and to some extent the heavy metal content.

5.5. Sludge Conditioner

Sludge contains up to 95% of water content, which requires dewatering treatment to achieve a reduced volume. The disposal of sludge and incineration ash is not only expensive but also demands a huge area of land, which erupts with environmental problems if left untreated. From the viewpoint of solid waste management, the best way to reduce the amount of sludge and incineration ash to be disposed is to reuse them by possibly combining sludge and fly ash mixture. However, both wastes contain a certain level of pollutants, especially heavy metals, depending on the characteristics of both wastes. The main concern for the solid waste management is that the leachates from both the wastes or their products should not pose any issues regarding contamination. Several studies have been carried out to investigate the possibilities of combining both wastes and reusing them. The study carried out by [216] says that the presence of oil in small amounts makes it harder to dewater the sludge. To avoid this issue, sludge can be conditioned with filter aids. The addition of MSWI fly ash decreased the specific resistances and capillary suction times of oily sludge with up to the 3% dosage. The total suspended solids of filtrate decreased with fly ash dosage, but the toxic concentrations of heavy metals increased considerably [216]. Based on the studies conducted, it is observed that the variations in the characteristics of fly ashes and the sludge are based on their operational parameters, which suggests that, to draw conclusive and beneficial results, further testing and studies are required before establishing a best approach for sludge conditioning with fly ash.

6. Conclusions

Urban population proportion is increasing year by year in China, and it results in the generation of massive amounts of waste. In China, urban areas are highly correlated with municipal solid waste generation. Municipal solid waste management in China is increasingly shifting towards incineration. Increasing reliance on incineration as a quick disposal option reduces mass and volume considerably, but it generates incineration fly ash. The threat posed by the hazardous content (heavy metals) in improperly handled fly ash is due to the lack of technical and economical feasibility of disposal. However, fly ash can serve as an alternative to natural resources used in cement, ceramics, embankments, road construction, etc. To achieve a safe disposal and reuse of fly ash, cost-effective resource recovery and reuse technologies need to be implemented. The treatment techniques mostly used are thermal treatment, solidification/stabilization and separation techniques. The thermal treatment and solidification/stabilization can be found at full scale (industrial, pilot and lab scales), whereas separation techniques fit in pretreatment. Based on the review of recent literature, it can be concluded that thermochemical treatment with the involvement of chlorinating agents can remove maximum amounts of heavy metals from fly ash. The solidification/stabilization method is also capable of fixing the heavy metal in fly ash, but the solidification/stabilization effect is poor under acidic conditions or after a long period of aging. The separation method can selectively recover heavy

metal, but a large amount of contaminated wastewater is generated after the treatment, which increases the difficulty of subsequent harmless disposal. This method is still on a laboratory scale, and further research is required. Therefore, it is necessary to comprehensively consider the advantages and disadvantages of different treatment methods and to identify appropriate methods to achieve a harmless disposal of MSWI fly ash. In order to avoid the landfilling of fly ash, potential applications for the utilization of treated fly ash need to be supported by the government and private sectors. The stringent legislations and defiance monitoring are compulsory to manage and reduce the occurrence of heavy metal abundance caused by mismanagement prior to incineration and after the improper disposal.

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