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Review

Characteristics and Treatment Methods of Medical Waste Incinerator Fly Ash: A Review

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Abstract: Medical waste incinerator fly ash (MWIFA) is quite different from municipal solid waste incinerator fly ash (MSWIFA) due to its special characteristics of high levels of chlorines, dioxins, carbon constituents, and heavy metals, which may cause irreversible harm to environment and human beings if managed improperly. However, treatment of MWIFA has rarely been specifically mentioned. In this review, various treatment techniques for MSWIFA, and their merits, demerits, applicability, and limitations for MWIFA are reviewed. Natural properties of MWIFA including the high contents of chlorine and carbonaceous matter that might affect the treatment effects of MWIFA are also depicted. Finally, several commendatory and feasible technologies such as roasting, residual carbon melting, the mechanochemical technique, flotation, and microwave treatment are recommended after an overall consideration of the special characteristics of MWIFA, balancing environmental, technological, economical information.

Keywords: medical waste incinerator fly ash; characteristics; treatment methods; heavy metals; dioxins

1. Introduction

Medical waste incinerator fly ash (MWIFA) refers to a kind of incineration residue accounting for approximately 3–5 wt.% total mass of the original waste, which is collected by the bag filter of the waste incineration system [1]. MWIFA contains high levels of dioxins, leachable alkali chlorides, carbon constituents, and heavy metals. The dioxins in fly ash typically account for more than 80% of that in the incinerator [2,3]. Consequently, it has been designated as hazardous waste by many countries. MWIFA contains quite large amounts of chlorines and carbons, which makes the constituents of MWIFA more complex than MSWIFA and, thus, treatment of MWIFA is also more difficult. Suitable techniques for MWIFA treatment need to be explored [4].

Currently, several conventional methods are used to treat MSW incinerator fly ash (MSWIFA), these methods can be grouped into three classes: (1) separation; (2) solidification/stabilization (S/S); and (3) thermal (melting, roasting, sintering, and low temperature treatment) [5,6]. These methods either remove heavy metals from fly ash or stabilize heavy metals in an insoluble form. Recently, investigations on the degradation/decomposition of dioxins in MSWIFA using thermal degradation, mechanochemical treatment, hydrothermal treatment, catalytic hydro-dechlorination, and supercritical water oxidation techniques has been investigated, and detoxification of MSWIFA has gradually been realized. However, methods that are applicative to MSWIFA may not be appropriate for MWIFA treatment because of the high concentrations of chlorines and carbon constituents present in MWIFA.

Therefore, developing effective technologies for MWIFA treatment is urgent. The objectives of this paper are to compare the various treatment methods of MSWIFA and to evaluate their feasibility for MWIFA treatment referring to the special characteristics of high concentrations of chlorines, dioxins, carbon constituents, and heavy metals.

2. Special Characteristics of MWIFA

2.1. High Content of Chlorides

MW contains a high amount of chlorinated plastics. The typical chlorine level in MW is 1.1–2.1%, whereas it is 0.2–0.8% in MSW [7]. After incineration, chlorines in plastics easily migrate to MWIFA leading to the high chlorine content in MWIFA. In MW incineration, HCl is generated after a reaction between organic chlorides and H ions [8]. Then, HCl vaporizes into the flue and reacts with an alkali, such as NaOH or $Ca(OH)_2$. Finally, NaCl and $CaCl_2$ are formed, contributing to the high chlorine content in MWIFA [9,10]. In addition, disposable disinfectants (such as NaCl and KCl) may also contribute to this result [11]. Especially in China, the medical service consumption of NaCl is higher than that in other countries. Table 1 lists the chemical composition of MWIFA and MSWIFA [12–17]. The main chemical compounds in both are SiO_2 and CaO. Because a large amount of $CaO/Ca(OH)_2$ must be sprayed into the exhaust gas to reduce acid gas emission [18], a relatively high content of CaO is found in the constituents of both MWIFA and MSWIFA. However, the chlorine content is approximately 20% in MWIFA, lower than 10% in MSWIFA [19].

Table 1. Chemical composition of medical waste incinerator fly ash (MWIFA) and municipal solid waste incinerator fly ash (MSWIFA)/%.

	SiO_2	CaO	Al_2O_3	Fe_2O_3	MgO	K_2O	Na_2O	SO_3	Cl	TiO_2	F	LOI	References
MWIFA	17.13	24.42	2.85	1.78	1.80	2.80	15.20	6.37	20.43	1.34	2.59	11.10	[12]
MWIFA	9.06	5.37	10.11	1.49	3.48	1.64	22.05	1.03	17.07	-	0.75	-	[13]
MWIFA	8.00	38.50	6.90	1.10	2.30	3.30	1.60	1.60	30.70	3.20	-	-	[14]
MSWIFA	73.10	1.06	16.70	1.96	-	3.94	2.42	-	-	0.35	-	-	[15]
MSWIFA	55.37	19.39	9.20	4.93	0.41	0.43	0.24	1.53	0.44	-	-	-	[16]
MSWIFA	62.30	0.50	28.10	2.10	1.00	1.00	0.50	0.40	-	-	-	2.50	[17]

2.2. High Content of Dioxins

Dioxins exhibit resistance and have a hydrophobic nature towards metabolism. The bioaccumulation in the fatty tissues of humans and animals, and chemical persistence of dioxins in the environment has attracted considerable attention. Consequently, a better understanding of the emission theories of dioxins is necessary [20].

Three theories for dioxin emission from incinerators have been established in combustion research. (1) In de novo synthesis, dioxins are produced in the postcombustion zone through the reaction of a residue carbon or metal catalyst in the fly ash reaction [21]. During this process, solid-phase chlorides (such as metal chlorides) and gas-phase chlorines (such as HCl and Cl_2) play overriding roles in the precursor formation of dioxins [22–24]. Under a temperature range of 180–600 °C, heavy metals in the incinerator are vaporized and react with chlorines to form heavy metal chlorides in the flue (Formula (1)). These heavy metal chlorides are effective catalysts in the formation of dioxins [25,26]. With the help of formed catalysts, HCl can react with O_2 or oxidizing radicals to form chlorine radicals, indirectly promoting dioxin formation [27] (Formula (2)).

$$4HCl + O_2 = 2Cl_2 + 2H_2O \quad (1)$$

$$HCl + OH^- \rightarrow Cl^- + H_2O \quad (2)$$

(2) In precursor synthesis, dioxin levels are closely linked to other generated persistent organic pollutants (POPs). Dioxins are formed from precursors, such as chlorobenzenes, polychlorinated

diphenyl ethers, chlorophenols, as well as part of polycyclic aromatic hydrocarbons (PAHs), Polybrominated diphenyl ethers (PBDEs), and Polychlorinated biphenyls (PCBs) [28–30].

(3) In incomplete combustion formation, dioxins are generated during the incomplete combustion of chlorinated plastics and intermittent operations of MWIs [22]. In addition to the content of chlorines and metals, the formation of dioxins is also relevant to the content of SO_2, CO, O_2, and carbons in fly ash [23]. Moreover, other factors promote the formation of dioxins, such as types of incinerators or kilns, operation conditions, and types of APCDs.

In addition, there are small quantities of dioxins in the raw MW itself, which can be incompletely destroyed during incineration and transferred to MWIFA [4].

Dioxins are highly hydrophobic and strongly lipophilic possessing extremely nonpolar properties [25]. Consequently, dioxins can easily adhere to fly ash particles with similar characteristics with the help of PAC. Approximately 91 to 98% of dioxins in flue gas can be effectively captured by PAC, removed through APCDs, and transferred into MWIFA [22,31,32]. Pan reported that MWIFA has a higher concentration of dioxins by approximately 2 to 3 orders of magnitude than that of MSWIFA [32,33]. Table 2 presents the concentrations and (toxic equivalent qualities) TEQs of dioxins in MWIFA and MSWIFA from different incinerators and APCDs.

Table 2. Concentrations and toxic equivalent qualities (TEQs) of dioxins in MWIFA and MSWIFA.

Treatment Object	Total Numbers of Incinerators	Treatment Capacity	Types of Incinerators	Air Pollution Control Device	Concentrations of Dioxins (ng/g)	References
MSW	15	300~1500 t/d	Grate-type or fluidized bed incinerator	SS + AC + BF	2.8–190	[33]
MSW	16	300~1800 t/d	mass-burning	SD + AC + BF; SD	9.07–46.68, average 23.53	[34]
MSW	5	200~350 t/d 400~700 t/d	Grate-type or fluidized bed incinerator	SS + BF	19.2–236	[35]
HSW	1	10 t/d	rotary kiln + circulated fluidized bed;	SS + BF	2918	
HSW		15 t/d	rotary kiln	SS + AC + bag filter	78.79	[36]
HSW	1	10 t/d	rotary kiln fluidized bed multi-stage pyrolysis	SS + AC + bag filter	67.83, 125.3	[3]
HSW	1	20 t/d	Rotary kiln + second combustion chamber	SS + AC + BF		[37,38]

SD: semidry scrubber; AC: activated carbon injector; BF: bag filter; SS: Semidry scrubber.

2.3. High Content of Carbon Constituents

High carbon content is another crucial characteristic of MWIFA not present for MSWIFA. Carbon constituents in MWIFA mainly include PAC and UC. UC usually accounts for 3 to 10 wt.% in incinerator fly ash [39]. Because of the high absorbent efficiency and simple implementation, PAC injection followed by BF application is a standard configuration to reduce POPs, such as dioxins emitted from incinerators [40,41]. With a higher level of dioxin emission and stricter emission standard for dioxins, a larger amount of PAC must be injected upstream of the BF [42]. Usually, 105–115 ng of dioxins can be absorbed per gram of PAC [43]. Finally, approximately 92 to 99% of dioxins in the flue can be removed [44,45]. After injection of 100 mg/m^3 of PAC into an MSWI, the dioxin level in flue gas decreases to approximately 0.04 ng I-TEQ/Nm^3 from an original value of 0.3 to 0.4 ng I-TEQ/Nm^3. For an MSWI, 50–200 mg/Nm^3 of PAC is generally required for a single BF [34,46]. For an MWI, the amount of PAC injected into the rotary kiln can be up to 800 mg/Nm^3 [3]. The greater amount of PAC injected, the higher the POP removal efficiency becomes. However, waste PAC containing dioxins must be carefully treated because of dioxin toxicity.

Flue gas meets emission standards after dioxins, PAHs, PCBs, and other toxic organic pollutants are captured by the injection of PAC. A high amount of injected PAC accumulates on the surface of the filter, becoming a part of fly ash. Injected PAC and UC in solid waste are the main carbon sources of fly ash [34]. The carbon content in some MWIFA reaches 11.4–91.0% [3].

2.4. High Content of Heavy Metals

Another characteristic of MWIFA is that it is enriched with heavy metals [47]. Generally, the large amount of Zn and Cr in MW may come from syringes, waste plastics, rubber, and medical adhesive plaster [48,49]. The usage of PVC plastics in MW usually contributes to Pb- and Cd-loads in MW [46]. The plasticizers and adhesives included in the packaging of MW always contain Cu, Zn, and other heavy metals because of the frequent usage of aluminum foil, PET, PVC, and PP [22]. In addition, Cd and Hg are also found in dental clinics, broken medical facilities, and discarded batteries [50]. Heavy metals in MW usually present as metal oxides, metal elements, volatile metallic chlorides, and sulfates. In incineration, they are not destroyed; instead, a small fraction form volatile metallic vapors and enter into the flue gas, which may be emitted into the environment if not effectively removed by APCDs. Most heavy metals migrate or concentrate in the fly ash and bottom ash; depending on the formed compounds of heavy metals and their physicochemical properties during incineration [51]. The migration of heavy metals during incineration is depicted in Figure 1. A relatively high concentration of low-volatility heavy metals, such as Cu and Cr, are enriched in the bottom ash. For Pb, Zn, and Cd, the relatively high temperature and existing chlorines in MW promote chlorination and volatilization [52]. Then, the formed heavy metal chlorides are absorbed by UC and the injected PAC, leading to the enrichment of $PbCl_2$, $ZnCl_2$, and $CdCl_2$ in the final MWIFA [53]. Compared with MSWIFA, the content of heavy metals, such as Zn and Cu, in MWIFA is higher, reaching >5.24 g/kg and 1.02 g/kg, respectively (Table 3) [54]. Moreover, a high level of Cu may facilitate the formation of POPs by working as a catalyst. Toxic heavy metals in MWIFA can be easily precipitated into the soil and eventually leached into underground water if not immobilized or removed in time; this may indirectly cause harm to human beings and the environment [45].

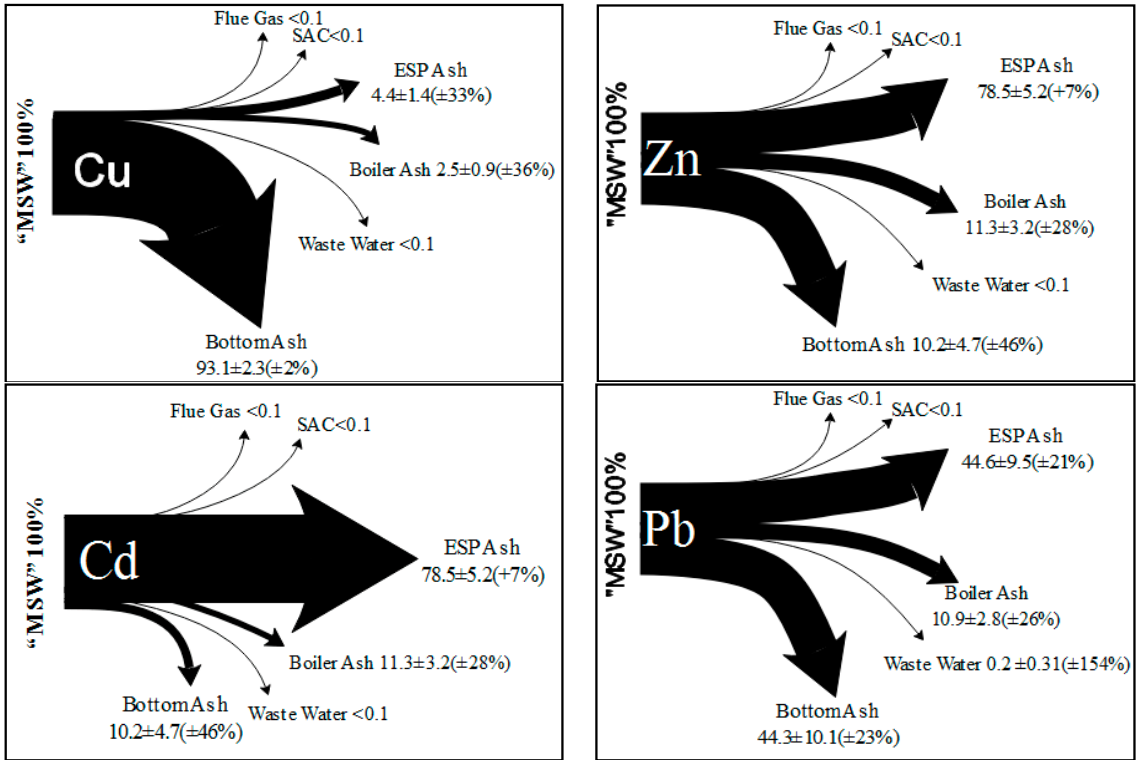

Figure 1. Partitioning of Cu, Zn, Cd, and Pb during medical waste (MW) incineration.

Table 3. Comparison of heavy metal concentrations in MWIFA and MSWIFA/mg/g.

Heavy Metals (mg/g)	MWIFA		MSWIFA	
	Range	Average	Range	Average
Cr	0.02–0.26	0.06	0.07–0.86	0.21
Ni	0.12–0.18	0.04	0.02–0.12	0.06
Zn	8.29–121.41	78.69	0.40–25.80	7.66
Cd	0.12–0.64	0.23	0.03–0.47	0.13
Ba	1.21–2.90	NF	0.54–4.30	NF
Pb	1.90–5.40	3.54	0.20–10.60	2.83
Cu	0.80–2.91	1.70	0.19–1.30	0.68
As	0.07–0.24	0.17	0.09–0.24	0.05

References: [1,48,55–58]; NF: No found.

3. Available Techniques for MWIFA Treatment

3.1. Water Washing Pretreatment (WWP)

WWP, an economical and effective dechlorination technique, is widely used for removing soluble chloride salts from incinerator fly ash with water. WWP is usually applied together with other post-treatment methods [59]. By using this technique, more than 70% of chlorine can be effectively removed [60], it also helps to mitigate potential environmental impacts and reduce treatment costs and cement consumption [61,62]. However, the generated wastewater containing released heavy metals should be subject to secondary treatment by adjusting its pH [59]. Moreover, WWP is unable to remove the insoluble chlorides and thus unable to reduce the content of chlorine lower than 0.5%. Therefore, other methods, such as thermal treatment, mechanochemical technique, and phosphoric acid stabilization need to be used after WWP.

3.2. Acid Leaching Pretreatment (ALP)

ALP can facilitate the separation and recovery of heavy metals from fly ash together with other post-treatment, such as thermal treatment, melting, solvent extraction, sulfidation precipitation, flotation, and immobilization treatment [1,61,63]. Many kinds of lixiviants have been applied in the ALP process: organic acids, such as tartaric acid, acetic acid, oxalic acid, and citric acid; inorganic acids, such as HNO_3, HCl, and H_2SO_4; chelating reagents, such as diethylenetriaminepentaacetate, ethylendiaminetetraacetate, and nitrilotriacetic acid; and alkaline lixiviants, such as sodium hydroxides and ammonium salts. Among those agents, chelating reagents present better leaching ability, however, they are more harmful to the environment. A proportion of heavy metals in fly ash are difficult to recover after ALP because they can strongly chelate with the reagents [64]. Alkaline lixiviants have little effect on the removal of this subset of heavy metals, such as Cu, Zn, and Cd, so an integrated leaching process should include acid to extract the other alkaline-insoluble metals [64]. This inevitably adds both inconvenience and extra cost [52]. In addition, fungal bioleaching, hydrothermal treatment, and subcritical water extraction are also used. However, they are rarely used because they are either expensive or need more processing time [64].

Massive experiments proved that acidic lixiviants are more feasible and effective, especially for mineral acids [52]. HCl and H_2SO_4 are the most commonly used mineral acids because they are biodegradable, relatively low-cost, and environmental friendly. Meanwhile, they present better leaching yields (>85%) and the formed water-insoluble substances (such as $PbSO_4$ or $PbCl_2$) can undergo complexation reactions with lead ions [65,66]. Tang found that approximately 92% of Cd, 90% of Pb, 68% of Cu, and 81% of Zn were solubilized and leached out with HCl [58].

Apart from lixiviant types, the acid leaching process is also influenced by ash compounds, ash morphology, pH, liquid to solid ratios, and leaching times. However, a large amount of acid is often consumed as fly ash presents high alkaline property and the final residues need further disposal, which should be overcome in future research work.

3.3. Cement Solidification Technology (CST)

CST aims to prevent heavy metals in MSWIFA from leaching out. In the solidification process, a certain amount of water, cement, and additives (such as phosphates, chelating agents, ferrite solutions, colloidal aluminate oxides, silica, sodium sulfide, and thiourea) are added into incinerator fly ash (Table 4) [66–73]. Before being disposed in landfill the unconfined uniaxial compressive strength of the solidified body and its leaching toxicity are tested. CST has been widely developed in the past two decades. It is the most widely accepted solidification and/or stabilization method prior to landfill disposal in most countries because it is relatively easy to implement and is low-cost. However, it consumes a large amount of cement. After cement solidification, both the volume and mass of cement-solidified bodies were found to increase by 40% and 30%, respectively [67]. Sometimes the proportions were as high as 50% [5]. These solidified products are nonrenewable resources and would reduce the useful lifetime of a landfill site; the long-term stability of heavy metals in solidified products cannot be guaranteed and they cannot be assured to be completely harmless [74]. Thus, long-term environmental risks are questionable. Moreover, resource recycling of heavy metals cannot be realized by CST. In addition, the cost is relatively high if the chelating agent is used in the cementation process [62].

Table 4. A literature overview about cement solidification of incinerator fly ash.

Objectives	Optimal Parameters	Findings	Reference
90% Washed ash + 10% Portland cement solidification.	liquid/solid = 0.88, washing time = 15 min, setting time = 3.30 h or 5.30 h.	50–80% of chlorides were washed out. Washing shortens the setting time of solidified body. The expense reduced by approximately 50 to 63%. After solidified body was stored for seven days at 20 °C, leachabilities of Cd, Cr, Cu, and Pb were lower than the limited values of Italy.	[66]
Cementitious stabilization	Cement proportion = 13–40 wt.%. water content = 20–30 wt.%	Cementitious body is used for transportation or landfilling. And heavy metals can be stabilized. Besides, the solid waste mass is increased by up to 40 wt.%. The cost was about 240 CNY/t.	[67]
Activators + cement stabilization	Ash/cement/$Ca(OH)_2$/Na_2SO_4 (K_2SO_4 or $CaCl_2$) = 100/25/20/5.80. Water/(fly ash + cement) = 0.35.	The addition of $Ca(OH)_2$ together with either Na_2SO_4, $CaCl_2$, or K_2SO_4 improves the hydration reaction of carbon enriched fly ash. After curving for 90 days, compressive strength reaches to 35 MPa under optimal conditions.	[68]
Four-stage waste washing pretreatment + stabilization	No found.	Four-step washing pretreatment can optimize the stability and compressive strength of fly ash and produce usable concrete aggregates.	[63]
Sand additives + blended cement Solidification.	sand/mixture = 3; water/mixture = 0.47 or 0.50. cement/fly ash = 3	Cement and furnace slag can produce modified ash, which can substitute cement in dispose of the pretreated fly ash harmlessly; but it presented a poor immobilization for Cr.	[69]
Portland cement Solidification	Fly ash/cement < 50 wt.%. L/s (w/s) = 0.40.	Fly ash contained with 16 wt.% of chlorides cannot be effectively immobilized when cement content is lower than 50 wt.%. Compressive strengths were low after 28 days.	[70]
Sulfoaluminate cement Solidification.	fly ash amount = 50%; water/binder = 0.30. curving time = 28 days.	Under the optimal conditions, compressive strength was 32.60 MPa. Leaching concentration of Zn, Pb, and Cu meet the threshold values of China. Sulfoaluminate cement was proved to be better than Portland cement in solidification.	[71]
Water washing pretreatment + Portland cement stabilization	Washing conditions: L/s = 5:1, 0.50 h; binder: sand: water = 2:6:1. Fly ash: cement = 1:1.	Washing process can remove >80% of Cl^- and $SO_3{}^-$ in the MSWIFA. Compressive strength was 11.52 MPa when mixing with 50% fly ash and curving for 28 days. Final products were used as base construction/decorative material, backfill, or patios.	[72]
Portland cement solidification to produce fly ash based geopolymer concrete.	aggregate:sand = 1:1, ash = 400 kg, cement = 15 wt.%, resting time = 30 min, temperature = 70 °C.	Compressive strength was higher than 25 MPa when curving for 7 to 28 days under optimal conditions. A 30 min of resting time was found to be more effective than 24 h.	[17]
Fly ash cenosphere modified cement pastes with nano silica (NS).	Water/binder = 0.30. Cement/ash = 9:1. nano silica = 1 wt.%.	Application of fly ash cenosphere can decrease the density of the final products without greater loss for strength (about 75 MPa). An excellent compact microstructure was obtained after the addition of nano silica.	[15]
Cement solidification of MSWIFA from India.	Cement content < 6%. Curving time = 7 days.	Cement addition improves compressive strength of solidified product due to the pozzolanic reactions. More than 6% of cement addition is not beneficial both for volume and economic. Final products can be used as lightweight filling materials.	[73]

Especially, when cement solidification is used for treatment of MWIFA, characteristics of higher concentrations of PAC, chlorides, and POPs would bring about a series of problems: high content of PAC in fly ash reduces the compressive strength of the solidified body and increases porosity, providing convenience for the entrance of leachates, and more heavy metals can easily leach out through the holes in the cement-solidified body. In order to meet landfill requirements, up to 70% of cement should be added to MWIFA, exceeding the upper limit of 40% for MSWIFA [68]; in addition, the high amount of chlorides and sulfates prolong cement solidification time and impede the cement hydration reaction [75,76]. Moreover, POPs, especially for dioxins, enriched in MWIFA cannot undergo decomposition. Thus, the potential emission risk for dioxins in solidified products is another fatal weakness for CST. High concentrations of dioxins are detected in soils around landfills, which are hundreds of times higher than those in common soils [33]. Furthermore, with the increase in population, less pollution-free land is available for landfilling of fly ash solidification bodies [77]. In conclusion, the present CST for MWIFA is just one of the expedients and should be reconsidered.

3.4. Melting Technique (MT)

Melting technique (MT) has been widely applied in MSWIFA treatment in the past two decades. It can immobilize most heavy metals and decompose POPs in MSWIFA under temperatures higher than the melting points of MSWIFA [32]. Currently, the electric melting furnace and fuel melting furnace methods have been studied [78,79]. The electric melting furnace mainly includes the arc melting furnace, plasma melting furnace, and resistance melting furnace. The fuel melting furnace mainly includes the reflector melting furnace, rotary kiln melting furnace, and residual carbon melting furnace [80]. Among these furnaces, the plasma molten furnace has been better developed since it can decompose toxic substances and reduce the volume of fly ash more efficiently [81]. After melting treatment, heavy metals are distributed into the molten fly ash, ingots, and slags; the mobility of toxic metals in slags is effectively controlled during this process [82]. Melting operation at above 1300 °C is superior with regard to dioxin decomposition. The decomposition efficiency of dioxins was higher than 99% when exposed to incinerator fly ash under 1400 °C [83] (Table 5). Furthermore, molten glasses or slag byproducts can be used as construction materials or road construction [33].

In spite of a series of merits, MT is limited in treating MSWIFA. First, MSWIFA contains a relatively high content of Na_2O and SO_3. Thus, the refractory of the furnace can easily suffer corrosion. Secondly, it operates under a high temperature and needs to consume a considerable amount of energy. Energy consumption and expense for this technique could reach approximately 1.5 kWh/kg and 3000 CNY/t, respectively [79]. In addition, melting processes which involve extensive equipment can increase cost up to 15 times compared to cement solidification. Thus, it is not suitable for most actual technologies in developing countries.

Table 5. A retrospect concerning the melting treatment of incinerator fly ash.

Feedstock and Objectives	Devices	Treatment Conditions	DDRs/Leaching or Stabilization of Heavy Metals	Re-utilization of Slags	Reference
MSWIFA Melting treatment	Residual carbon furnace	T = 1200 °C or 1400 °C	DDRs are over than 99%. Metals can be separated by evaporation or physical gravity after cooling.	Construction materials.	[67]
MSWIFA Melting treatment	Electronic arc furnace	T = 1250–1300 °C time > 5 h.	DDRs are 99.90%. Stabilization of heavy metals is not mentioned.	Roadbed materials	[81]
Water extraction + MSWIFA melting process	Electric heated furnace	L/s = 10, MT = 1000–1350 °C.	Zn (70.60%), Cu (73.90%), and Pb (58.10%) were immobilized in the water-extracted fly ash, higher than those for raw fly ash.	Byproduct can be reutilization.	[84]
Production of glassy slag with MSWIFA by vitrification.	DC double plasma torch	T = 1500–1600 °C.	DDRs are 99.32% (99.95% in TEQ). Leaching concentrations of heavy metals in slags meet the regulatory standard but visibly volatilized.	Glassy slags were safe enough to reuse.	[79]
Cullet additive + melting + sintering.	Electronic Arc Furnace	Ash/Cullet = 3 TiO_2 = 3 wt.%, 850 °C, 30 min	Leachate concentrations of Cu, Pb, Zn, Cr, and Cd for glass-ceramics toxicity identification standard of China.	Substitute nature marble, porcelain tiles and granite.	[80]

Table 5. *Cont.*

Feedstock and Objectives	Devices	Treatment Conditions	DDRs/Leaching or Stabilization of Heavy Metals	Re-utilization of Slags	Reference
Water washing + melting + sintering of MSWIFA.	Electronic Arc Furnace	L/s = 20, TiO_2 = 3 wt.%, 900 °C ash/cullet = 3, Melting/sintering time = 30 min/2 h.	Leachate concentrations of Cu, Pb, Zn, Cr, and Cd for glass-ceramics toxicity identification standard of China.	Glass-ceramics can substitute of nature materials.	[75]
Conversion MWIFA into harmless slag by Melting	DC thermal plasma torch	Melted for 15 min. working gas: argon. Flow rate = 12–14 L/min	DDRs > 99% in TEQ. Leaching concentration of heavy metals in the slag meet the Chinese regulatory standard. The volume reduction is 78% after melting.	Produced slag presents good performance in microstructure.	[33]
Production of porous materials with MSWIFA by melting process.	Thermal Plasma furnace	Bottom ash/fly ash = 1. L/s = 0.50, 1600 °C. frother = 3 wt.%. cement: slag = 1	Concentrations of heavy metals in water-quenched slag meet the TCLP criteria of Taiwan.	Products are used as architectural and decorative materials.	[85]
Zero waste treatment of MSWIFA	Electric arc furnace	T = 1630–1730 °C. 2 s residence time	DDRs = 99.999%. Heavy metals in furnace dust and slag meet the regulatory thresholds of Taiwan EPA	Zero landfill	[86]

DDRs: Dioxin decomposition rate.

When melting is used for treating MWIFA, there are more obstacles. First, factories and facilities of MWI are small-scale (treatment capacity of 15–30 t/d) and dispersive, which is far less than treatment capacity of MSW incinerators (above 500 t/d) (Table 2). Secondly, the high contents of carbon constituents in MWIFA combust easily under high temperature, which is harmful to the graphite electrode as for the electric melting furnace. Thus, only residual carbon melting furnace might be suitable for use [75,80]. Thirdly, MWIFA contains a high chlorine inventory. Heavy metal chlorides formed in the melting process are too volatile to be fixed in the vitreous products and inevitably cause secondary pollution [87]. Thus, more advanced APCDs are needed, but this would enhance extra energy consumption and costs. Therefore, from the viewpoint of economy, technology, and profits, most melting technologies are not the preferred solution for simplex MWIFA treatment, except residual carbon melting furnace.

3.5. Roasting

The roasting method is a thermal treatment process. It operates under the following conditions: 1–20 h, 670–1200 °C, and with or without addition of chlorinating agents ($CaCl_2$, NaCl, etc.) [88,89]. These chlorinating agents react with heavy metals or metal oxides. Then, the formed metal chlorides with low boiling points evaporate. By using the roasting treatment, heavy metals can be recycled, and POPs, especially for dioxins, can be effectively decomposed. After detoxification, residual ash can be safely landfill-disposed [90] (Table 6).

Table 6. Recent literature retrospect in the field of roasting treatment of incinerator fly ash.

Objectives	Treatment Conditions	Findings and Results	Reference
Thermal treatment for MSWIFA in Switzerland	670–1000 °C 750 °C, >6 h; at 840 °C 3–4 h; and at 920 °C 1.50 h.	Heavy metal oxides can transfer to metal chlorides and be completely evaporated. Metal evaporation would proceed as long as there are chlorides in fly ash. But it may be restricted by the formed metal silica/alumina constituents.	[88]
Roasting treatment for MSWIFA	Roasting for 3 h under 1000 °C. Fly ash = 3 g. $CaCl_2$ $2H_2O$ = 0.62 g (that is 0.30 g of Cl).	$CaCl_2$ was found to be an alternative chlorinating agent for heavy metals volatilizing. Volatilization efficiencies of recovered metals are proportional to standard free-energy changes values for the corresponding chlorination reactions.	[91]
Thermal treatment of MSWIFA.	Fly ash/$CaCl_2$ = 15:1 (w/w). Roasting temperature = 990 ± 10 °C for 1 h	This technology is found to be effective for removal of most heavy metals. But after ash was treated under 1000 °C, the leaching rate of Cr was increased 11 to 13%.	[92]
Thermal treatment of MSWIFA in Austria	800–1200 °C for 20 h. Chloride addition: 0–200 g Cl per kg ash. Added as NaCl, $CaCl_2$, or $MgCl_2$	At 1200 °C, above 95% of Zn, Cu, Pb, Cd with addition of $CaCl_2$ or $MgCl_2$, 75% Ni ($CaCl_2$), and 30% Cr ($MgCl_2$ or NaCl) could be vaporized.	[89]
Chloride removal by roasting and washing	Washing conditions: solid/liquid = 100 g/L, 1.50 h. Roasting conditions: 600, 800, 950, 1050 °C for 7, 4, 2, 1 h.	1050 °C for 3 h was found to be the optimal condition and removal rate of chloride = 83%. A solid to liquid ratio of 1:10 in washing process can remove 97% of water-soluble chlorides.	[90]

Researches show that temperature, atmosphere, roasting time, and the type and amount of chlorine medium have an effect on the evaporation rates of heavy metals [91,92]. NaCl is inclined to react with heavy metal oxides such as Cd, Cu, Pb, Zn, Ni, and Cr. The generated heavy metal chlorides are easier vaporized. The relevant chlorination mechanism can be expressed as Formulas (3) and (4). The former refers to reactions between the solid heavy metal oxides and solid phase NaCl under a high temperature atmosphere; the Na_2O obtained is easy to further react with SiO_2 and Al_2O_3 in incinerator fly ash, forming aluminosilicates.

$$MO + 2NaCl = MCl_2 + Na_2O \quad (3)$$

$$\alpha MO + 2\alpha NaCl + \beta SiO_2 = \alpha MCl_2 + \alpha Na_2O\ \beta SiO_2 \quad (4)$$

Thus, roasting is especially suitable for the treatment of chloride- and toxic metal-enriched MWIFA in theory. However industrial applications are rare because of the higher facility requirements, energy consumption, and costs that are needed for roasting compared with cement solidification.

3.6. Low Temperature Reatment (LTTT)

LTTT is also called thermal catalysis treatment or Hagenmaier treatment. It was originally applied to treat dioxin-contaminated matter in Germany and was also implemented in several waste incineration plants in Japan [82]. This process is usually executed between 250 and 600 °C [83,93]. After catalyzed or dechlorination/hydrogenation for 0.1–1 h by Cu and other substances in dioxin-contaminated matter [94], most dioxins and other organic pollutants can be destructed. The detoxification efficiency of dioxins is over 90%. Under low temperature conditions, reductive chemicals (such as salts and alkaline metals) are usually collocated to generate reductive gases and finally accelerate the dechlorination reaction [95].

LTTT has the advantages of simple operation and low energy consumption than high temperature treatment such as melting [96]. It is appropriate for developing countries to implement the Stockholm Convention [83]. However, it only decomposes organic pollutants in incinerator fly ash. Most heavy metals still enrich in fly ash sequentially and cannot be immobilized. Thus, LTTT might be inapplicable for treatment of MWIFA with a high content of heavy metals. However, this method can be one of the steps in the treatment of MWIFA and can be used in conjunction with heavy metal removal methods.

3.7. Catalytic Hydro-Dechlorination (CHD)

CHD, a reductive dechlorination method under mild conditions, is mainly used to decompose dioxins in incinerator fly ash. In this process, a suitable amount of catalysts and carriers are used to absorb and dissociate hydrogen under ordinary pressure, and then, reactive hydrogen obtained is used to degrade and remove chlorines in the dioxins. Finally dioxins can be detoxified without toxic intermediate products [97–99]. Supported noble metals, such as Pd, Pt, and Rh, are the most common catalysts, which possess strong capability for adsorption and dissociation of H_2 [100,101]. In addition, carbonaceous materials are widely used carriers because of the ability of resisting acid, alkali, and chlorine poisoning [102,103].

CHD presents high recovery efficiency, strong selectivity, stable reactive performance, low feed ratio, environmental friendly characters, and short reaction time. Additionally, when a water/isopropanol solution is added to the organic solvents the reaction becomes safer. Moreover, regeneration and recycling of Pd/C catalysts can reduce costs. Consequently, CHD has become an effective means of treating chlorinated-organic compounds and hazardous industrial wastes. However, hydrogen chlorides and ammonium chlorides are easy to generate in CHD process, which may inevitably lead to jamming and corroding of the facilities and result in a poor dechlorination performance [104–106]. CHD is an effective intermediate method for the decomposition of dioxins in MWIFA. However, it is still in the laboratory batch experiment stage and some operations may cause severe secondary pollution or dioxin regeneration. However, heavy metals in MWIFA cannot

be removed. From this point of view, this technique can be a sequential disposal step for dioxins cooperating with other methods which can removal heavy metals in MWIFA [107].

3.8. Supercritical Water Oxidation (SCWO)

SCWO has been developed to treat various wastes, such as PCB-contaminated mineral transformer oil, wastewater, sludge, and dioxin-contained fly ash (Table 7) [45,74,108,109]. This technique is an effective, harmless, fast, and violent detoxification method [108,110,111]. Under the condition of water supercritical point (22.1 MPa and 374 °C), supercritical water exhibits a perfect miscibility with organic contaminants such as dioxins. The reaction between free radicals and the pollutant substances comes up to realize the detoxification of MWIFA. To enhance the oxidation effect of supercritical water (SCW), hydrogen peroxide (H_2O_2) or oxygen (O_2) is used. Thereafter, most toxic metals are locked by silica matrix residues and combined with organic substances, which are hardly leached out. Residual heavy metals are precipitated in the form of Fe-Mn oxides which are thermodynamically stable. Finally, harmless products, such as CO_2 and H_2O together with acid and salts are generated [108,112–114] (Figure 2).

Table 7. A recent summary about SCWO treatment technology of incinerator fly ash.

Residues	Objectives	Treatment Conditions	Findings	Reference
MSWIFA from Taiwan and China.	SCW and SCWH treatment of heavy metals in MSWIFA.	The H_2O_2 was added.	This technique can stabilize metals in Fe-Mn oxides and residual fractions. Heavy metal leaching results meets the requirements of USEPA and Chinese EPA permits.	[74]
Oil-contained PCBs and heavy metals in MWIFA	The simultaneous detoxification of PCBs and heavy metals in MWIFA.	7.00–34.40 MPa, 280–410 °C. Reaction was finally quenched by water spray with supercritical water.	The technique meets technical and cost requirements, because MWIFA is found to possess the potential catalysis ability.	[45]
Dioxins in MSWIFA (SCWO)	Dioxin destruction and dioxins adsorption by activated carbon	500 °C, 20 MPa.	Decomposition efficiencies of dioxins and activated carbon were more than 99% and 99.99%.	[108]
Dioxin extraction in fly ash (SCWO)	Extraction of dioxins in fly ash	SC-CO_2 was used, solvent/feed = 5, time = 1 h, 40 °C, 50 MPa.	Extraction efficiency of dioxins attained 99.98%.	[108]
MSWIFA in China	Dioxins degradation by SCWO technique	400–500 °C, 23–29 MPa, fly ash = 0–6 g, 1–2 min, O_2 = 150–300%, H_2O_2 = 0–40 mL.	Mass concentration of dioxins reduces from 28.20 to only 2.79 ng/g, a degradation efficiency > 90%.	[109]

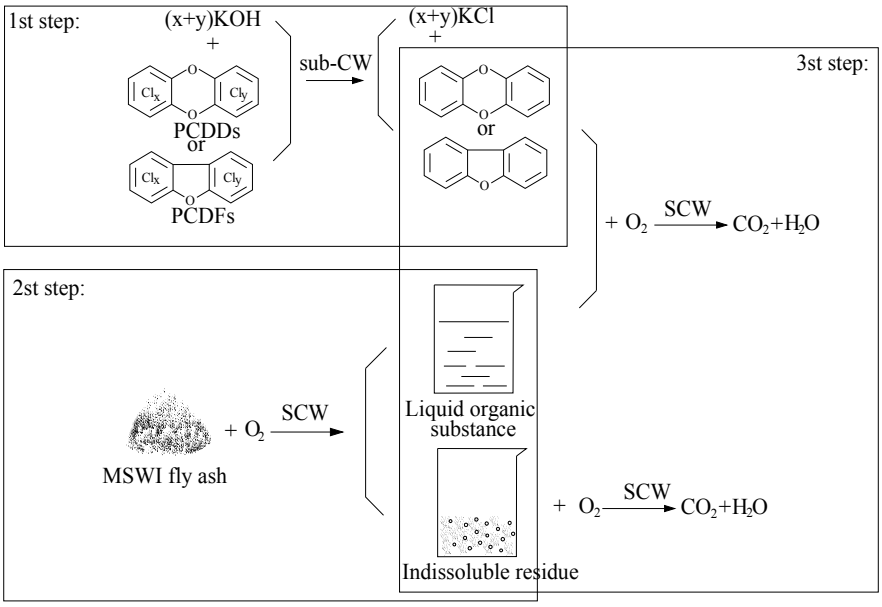

Figure 2. Outline of supercritical water oxidation (SCWO) for treatment of incinerator fly ash.

As one of the highly efficient oxidation technologies, SCWO is probably a promising method for the final treatment of MSWIFA. It is free from the generation of toxic intermediate products and presents a single decomposition route. In addition, there is no heat transfer resistance problem. Especially, high decomposition efficiencies of dioxins can be obtained. Also, the final products obtained are environmental friendly. Although it possesses unique superiorities in decomposing organic pollutants in MSWIFA [109,112], it operates under high temperature and pressure, which poses a high requirement for equipment and has high energy consumption.

3.9. Hydrothermal Treatment (HTT)

HTT can stabilize heavy metals and degrade/decompose dioxins in incinerator fly ash. Stabilization of heavy metals in fly ash is mainly attributed to the synthesis of zeolite-like materials through HTT. Most incinerator fly ash contains large amounts of Si and Al, which are exactly the key ingredients promoting the synthesis of zeolite-like materials with a porous three-dimensional cage network structure and tetrahedron ((SiO_4) and (AlO_4)) [115–117]. This kind of structure could prevent heavy metals from leaching by ion adsorption, ion exchange, precipitation, and physical package [118,119]. Under anaerobic conditions, the factors arranged in order can be listed: reaction temperature > Fe addition > washing pretreatment [120–122]. Under oxide, Fe^{3+}, and H_2O_2 existing conditions, the formation of free radicals is accelerated, which further accelerates above 90% dioxin degradation. Furthermore, the final solid products obtained can be used as acid neutralizing agents and absorbent materials in actual industry [123,124], which can reduce the problem of the secondary pollution (Table 8).

Table 8. Findings of recent studies concerning hydrothermal treatment of incinerator fly ash.

Objectives	Optimal Treatment Conditions	Findings	Reference
HT decomposition of the dioxins in fly ash	300 °C for 20 min; solvent is 1N NaOH solution containing 10 vol% methanol	Dioxins were completely decomposed. Toxicity of dioxins for the treated fly ash decreased to 0.03 ng I-TEQ/g.	[123]
Heavy metal removal from MSWIFA in Japan by washing + HT	Washing 30 min. HT: autoclave pressures = 1.2–2.0 MPa, 150 °C, 5 h, L/s = 10 mL/g.	67% Na, 76% K, and 48% Ca were extracted by washing for 30 min. Final products can produce silicon–sulfur fertilizer after further Cr disposal.	[125]
HT of MSWIFA to produce stable minerals.	NaOH = 0.5 M, L/s = 10 mL/g 180 °C, 48 h.	Most of heavy metals were less released in acid environment. But the concentration of Zn and Cd cannot meet the standard.	[119]
Production of concrete with MSWIFA.	NaOH or Na_2CO_3 10 g fly ash 50 g, 375 °C, 5 h.	Heavy metal leaching concentration decreased by over 58.33%, especially Zn (81.91%/86.89% were leached out by NaOH/Na_2CO_3, respectively).	[126]
Removal of Cu in MWIFA by additives + HT.	325 °C, 2 h, initial Cu^{2+} = 50 mg/L. vessel pressure = 22.2 MPa. L/s = 10 mL/g. ash/Na_2CO_3 = 10.	Temperature has little effect on Cu(II)removal. Removal efficiency increased from 94.80% to 99.90% with the increasing concentration of Cu(II) from 10 to 50 mg L^{-1}.	[127]
Decomposition of dioxins contained in MSWIFA by HT.	290 °C,1 h. A mixture of ferrous and ferric sulphates by 5% (wt/wt) with the Fe (III)/Fe (II) = 2.	90.33% of dioxins were decomposed (in TEQ) with addition of Fe, but the associated decomposition rates were relatively lower.	[122]
Acid treatment + HT and reutilization of MSWIFA	Acid treatment conditions: 30 min. pH = 6.2, L/s = 2 mL/g. HT conditions: 290 °C, 1 h, L/s = 2–2.5 mL/g. 50 g PO_4^{3-} /1 kg fly ash	Heavy metal leaching concentrations meet the Chinese limits. Acid-washing reduced over 79.80% of leaching concentration of heavy metals. Higher concentrations of Cl in acid-washed fly ash were decreased than in water-washed ones.	[128]
HT + additives to stabilize heavy metals in MSWIFA.	CFA and diatomite/MSWIFA = 3:7, 200 °C. Seed or Tobermorite = 3%,	Leaching toxicity of all heavy metals in MSWIFA decreased to the lower than the standard values even for Pb.	[129]
K-zeolite syntheses from biomass incineration ash and coal fly ash via HT.	Biomass incineration ash:2–15 g, KOH: 0.5 and 2 mol/L, CFA:10 g. HT conditions: 160 °C,24 h.	The synthesized K-zeolites can be used to remove radioactive cesium.	[130]
Additives + HT to stabilize heavy metals in MSWIFA.	Coal fly ash/MSWIFA = 3:7, L/s = 10 mL/g. NaOH = 0.5 mol/L. HT conditions:150 °C, 48 h.	Heavy metals were detected on the surface of synthesized tobermorite crystalline. Leaching toxicity of all metals met the standard values.	[131]

"L/S" refers to liquid/solid; "HT" refers to Hydrothermal.

HTT has bright development prospects for harmless treatment of MSWIFA. HTT reduces the requirement of equipment and saves energy compared with high temperature methods [104,122]. However, methanol exhibits some toxicity, and expensive catalysts are liable to be poisoned in the disposal process. Simultaneously, the surface of the equipment is easily corroded under high pressure and an alkaline environment.

Moreover, when HTT is used for MWIFA treatment, there are still other difficulties. As seen in Table 1, there are smaller amounts of the elements Si and Al in MWIFA compared with MSWIFA, which is an obstacle to the formation of both zeolite and tobermorite crystalline unless Si and Al are added. Consequently, more reliable data should be given to facilitate the harmless treatment of MWIFA by HTT.

3.10. Mechanochemical Technique (MCT)

MCT is a nonthermal process and nontail gas disposal method. Operation conditions are usually 100–600 rpm and 2–24 h in a fully closed reactor [51,132] (Table 9). In the MCT process, five types of reagents are usually used: Lewis bases (such as CaO, Al_2O_3, La_2O_3, and Bi_2O_3) which serve as electron donors; neutral substances (such as alumina and SiO_2), which can provide free radicals to decontaminate organics [133]; oxidants (such as δ-MnO_2 and $S_2O_8{}^{2-}$) which can promote the oxidation of organics; reductants including pure metals which can be used as both electron donor and H donor; in addition, in order to reduce the production of hazardous intermediate substances, organic molecules (such as amides, amines, alcohols, ethers, and glycols) are applied together with pure metals to serve as H donor in reaction process [134,135]. With development, Fe-SiO_2, Fe/Zn-SiO_2, and Fe/Ni-SiO_2, and auxiliary materials, such as CaO and quartz sand, are introduced [132,136,137].

The dioxin treatment mechanism by MCT includes the dual paths: destruction and dechlorination. Mechanical force prompts the adsorption of reagents to the surface of incinerator fly ash. Then structures of fly ash are broken, ruptured, and reunited. Specific surface area and pore volume of fly ash are significantly increased and the average pore sizes also decrease after collision and friction. For destruction of dioxins, crystal structures of organic halogenierte stoffes adsorbed on the surface of fly ash are cleavaged and destructed by metals or metallic oxides, and then dissociated as metal halide compounds and carbon oxides [3,138–140]. In addition, dioxins and POP-containing materials can be dechlorinated and decomposed. The activated surfaces of reagents react with organic contaminants in fly ash and electrons ionized by Lewis bases transfer to free radicals or intermediary byproducts. In this process, the carbon–chlorine bond, which is weaker than the carbon–hydrogen bond, is fractured. Finally, formless carbons, inorganic hydrocarbons, inorganic metallic chlorides, and carbon oxides are produced [135,141]. In addition, the mechanism for immobilization/separation of heavy metals from fly ash is thought to be heavy metal resorption by Ca/PO_4-associated immobile salts and then magnetic separation. In the grinding process, the particle size of fly ash is decreased and the active nano-Fe/Ca/CaO/PO_4 additives promote the production of immobile [Ca]/[PO_4] salts and the reunion of fly ash. Meanwhile, a portion of heavy metals are entrapped and resorbed into the newly formed aggregates; the other parts of heavy metals are adsorbed onto the surface of the nano-Fe due to its magnetic properties and then separate [142]. Finally, most heavy metals can be stabilized or separated. The leaching ability of heavy metals meets the threshold of the hazardous waste landfill standard value [51,133].

Applications of this method in treatment of MWIFA have been studied (Table 9). Relatively favorable studies indicated that this method should be paid close attention to as an innoxious treatment of MWIFA in spite of just being implemented under experimental conditions; the mechanism of mechanochemical treatment is relatively ripe. Thus, it might be straightforward to realize industrialization and large-scale applications.

Table 9. A literature overview in the terms of mechanochemical technique in recent years.

Objectives.	Optimal Conditions and Degradation/Destruction Efficiency of Dioxins					Other Findings	References
	Reagent (Ratio)	Rotation Speed	Grinding Time	Grinding Type	Optimal Efficiency		
Destruction of dioxins in MWIFA	CaO/fly ash = 6–60%	400 rpm	2 h	Planetary ball	PCDD = 76.80%; PCDF = 56.80%	Dioxin efficiency rose with increased ratio of CaO.	[3]
Destruction of dioxins in MWIFA	No addition	400 rpm	2 h	Planetary ball (XQM-0.4 L)	Destruction efficiency = 76%	-	[143]
Fly ash smelting with industrial secondary copper	CaO/SiO_2/fly ash = 4:1:5 (wt)	275 rpm	12 h	Planetary ball	Dioxins = 85%	Cu served as the catalyst during dioxin reformation	[135,139]
Decomposition of PCBs in contaminated soil	CaO/SiO_2/soil = 1:1:2 (wt)	400 rpm	20 h	Planetary ball	PCBs = 98%	Dioxins formed in the first 5 h were decomposed with sufficient time.	[144]
Destruction of dioxins and PCBs in MSWIFA	Ca/CaO/fly ash = 1/1/200	400 rpm	20 h	planetary ball, PM-100	completely detoxified	No traces of PCBs and dioxins were detected finally	[145]
Dechlorination/ destruction of dioxins in MSWIFA	Flyash:Ca: Al-powder = 30:4:1	600 rpm	10 h	planetary ball (QXQM-2)	Dioxins = 93.20%	Water washing and Fe/Al/Ca additives present better effect.	[141]
Heavy metal immobilization in MSWIFA with nano-Fe/Ca/CaO/ PO_4 reagent.	CaO:Fe:Ca = 5:2:2. ash: $[PO_4]$:nano-Fe/ Ca/CaO = 20:1:1	150 rpm	2 h	planetary ball (PM-100)	Immobilization rate of heavy metals = 98–100%	Heavy metal leaching concentrations were much lower than Japan standard value.	[132]
Dioxin degradation by washing + Fe/Ni-SiO_2 of MSWIFA	Ni/SiO_2/Fe/ fly ash = 1/2/4/200	400 rpm	24 h	bimetallic ball	Dioxins = 93.20%	Washing conditions: L/s = 4:1, stirring speed = 400 rpm, time = 60 min.	[51]

3.11. Flotation Treatment

Flotation is a physicochemical separation technique, which is generally used for treatment of mineral, wastewater, pollutant soil, and incinerator fly ash [146,147]. Flotation effects depend on many parameters: types and dosages of (flotation agents such as collectors, frothers, and surfactants), air flow rates, flotation time, particle size, pulp pH, and pulp density [148].

For MSWIFA flotation, 41.9% of the total dioxins and 44.1% of PCBs in MSWIFA were achieved by Huang with a mixture of nonionic surfactant of Tween 80 and Span 80 as promoters. Meanwhile, the interaction mechanism between the surfactants and the UC surfaces mainly depends on the hydrogen bonding between the polar oxygen-containing functional groups of the surfactant and the oxygenated functional groups on the UC surface [149–151]. High content of PAC is one of the special characteristics for MWIFA compared with MSWIFA. Dioxins tend to enrich the carbon constituents (including PAC and UC) contained in MWIFA [152]. Recently, flotation of fresh MWIFA was systematically investigated. Liu found approximately 90% of carbon constituents and dioxins in MWIFA could be enriched and then separated as froths by flotation [12,54,153]. A novel process for the successive removal of dioxins and recovery of heavy metals from MWIFA was also developed by Wei [42]. Thereafter, by the process of acid leaching-sulfidation and precipitation–flotation, 42.0% of Pb, 48.7% of Cu, and 49.9% of Zn could be recovered [1]. Furthermore, the destruction efficiencies of dioxins in the froths could exceed 98% at above 1000 °C after reburning treatment of the froths, so it seems that recirculation of froths in the incinerator can serve the dual purpose of destruction of dioxins and energy recovery of carbon constituents [42]. After flotation, dioxin content in the tailings was significantly reduced to approximately 1.55 ng I-TEQ/g, which meets the standard of landfill site of municipal solid waste (3 ng I-TEQ/g). The volume and toxicity of the tailings decreased greatly, which might be landfill disposed [153]. Therefore, flotation is one of the promising alternatives for MWIFA treatment.

The dioxin removal mechanism of MWIFA can be explained by three points: (1) there is a close adsorption relationship between porous PAC and gaseous-phase dioxins in flue gas, and these low chlorinated dioxins are strongly bound on PAC [154]. Thus, these adsorbed dioxins may be removed together with PAC during flotation; (2) some dioxins may enter to the liquid phase from the ash solid particulates during the pulp-conditioning process because of the breakup of ash particles or salt solubilization. Most of these dissolved dioxins may also interact with PAC and be adsorbed by PAC during flotation. This mechanism is similar to sorptive-flotation phenomenon [54]; (3) the free and naturally hydrophobic dioxin molecules may directly attach to kerosene collectors or adhere to air bubbles, and float to the froth product (Figure 3).

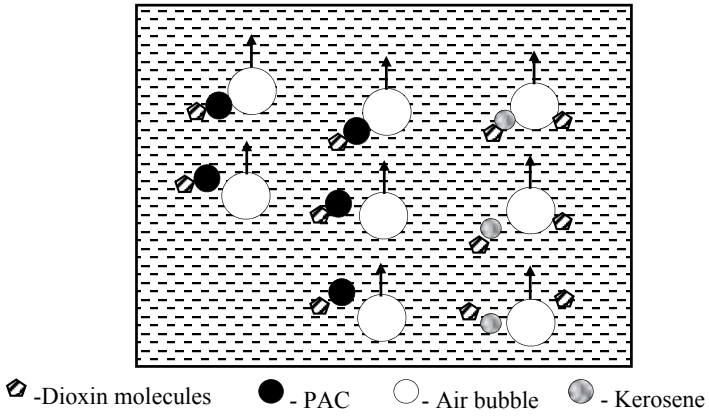

Figure 3. Conceptual diagram of the phenomenon regarding interactions among dioxin molecules, PAC, air bubbles, and kerosene during flotation.

MWIFA flotation has advantages of environmentally benign, flexibility, low-cost, and less usage of reagents. The flotation process does not require organic solvents, heating, expensive reagents, or fancy catalysts [145,146]; only electricity, economic reagents, and water are required [155]. Moreover, no exhaust gases are generated by this method. Especially, a combination of flotation and reburning treatments may represent a promising method to resolve difficulties from MWIFA treatment.

3.12. Microwave Treatment (MWT)

MWT has the characteristics of rapid heating, interior heating, and volumetric and selective heating, which are not present in conventional thermal methods [156]. It has been widely used in organic synthesis, polymer synthesis, food processing, analytical chemistry, and carbon regeneration [157–159]. MWT also has been used for the treatment of POPs (such as dioxins, chlorobenzene, and PCBs in soil or fly ash) and heavy metals in MSWIFA [156,160]. Heavy metals in MSWIFA can be stabilized with the assistance of the dielectric properties of the sample in the microwave field [161]. However, low content of absorbing medium was found in MSWIFA itself. Thus, microwave treatment of MSWIFA usually requires the use of a microwave-absorbing crucibles or the addition of microwave-absorbing additives. Researches have pointed out that PAC in pellets, graphite plates, and containers, which are made by dielectric materials (such as Al_2O_3, c-Al_2O_3, SiO_2, and kaolin), present better microwave absorb performance [156,158,162,163].

PAC demonstrates excellent microwave absorbing characteristics and its temperature can increase quickly and sharply to approximately 1000 °C in 2 min in the microwave field. Different from MSWIFA, a high content of PAC was found in MWIFA. If flotation is used for treatment of MWIFA, most of the PAC would be separated and concentrated into the froths together with dioxins [12]. Wei proved the destruction efficiency of dioxins exceeded 99 wt.% after microwave treatment of the froths. During this process, PAC in the froths would absorb a large amount of microwave energy, which helps form many “hot spots” and promotes the rapid decomposition of dioxins (Figure 4). Finally, the dioxins adsorbed on PAC pores or coexisted with the PAC could be rapidly decomposed

into HCl, CO_2, and H_2O [36,158]. Meanwhile, microwave treatment of the froths under nitrogen atmosphere might realize the regeneration of PAC. If regenerated PAC can be used as a dioxin adsorbent in flue gas, the treated froths might be injected again into APCDs of incinerator, which would significantly reduce the usage amount of parent PAC in the incinerator and decrease the costs. Thus, a combination of flotation and microwave treatment might be a promising method to resolve the MWIFA problem, which would possess many advantages, such as complete decomposition of dioxins, low-cost, and recovery of waste resources [42] (Figure 5).

Comparing with traditional sintering of incinerator fly ash by rotary kiln, a microwave sintering system has no need for gas injection for combustion, and diminishes the emission of hazardous products in flue gas, which indirectly reduces the investment and operation cost of the subsequent flue gas cleaning device. As an innovative technology, the effect of chlorides on dioxin decomposition remains to be further studied. In addition, for full-scale application, there are many obstacles to overcome, such as lack of fundamental data on material dielectric properties, uncontrollable factors, improvement of energy efficiency, and the hurdles in the capacity of the microwave furnace [164]. There is still distance ways to go until the practical application of microwave technology in MWIFA treatment can be realized.

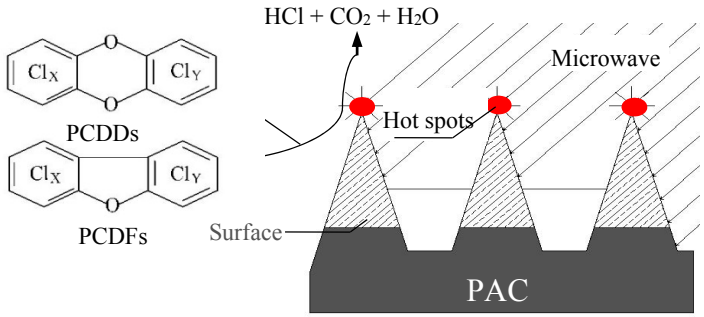

Figure 4. Possible decomposition mechanism of dioxins caused by "hot spot" formation on the surface of surfaces of PAC in the froths during microwave heating.

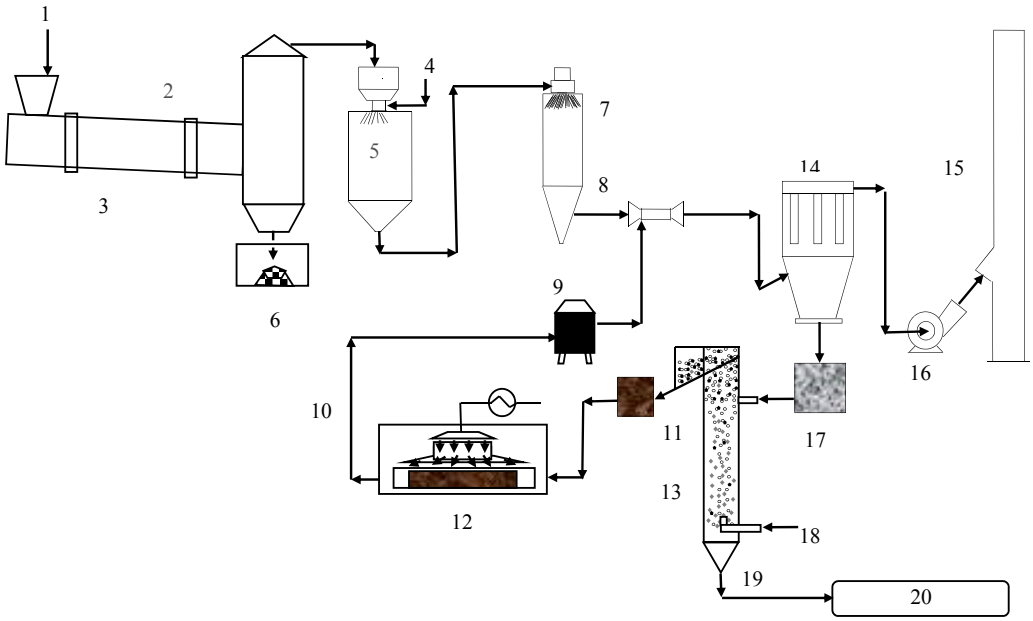

Figure 5. Flow scheme of the combination of flotation and microwave treatment MWIFA. 1—Medical waste; 2—Recombustion chamber; 3—Rotary kiln; 4—Water; 5—Quench tower; 6—Bottom ash; 7—Semidry scrubber; 8—PAC injection; 9—PAC storage tank; 10—Pulverization; 11—The froths; 12—Microwave sintering furnace; 13—Flotation column; 14—Bag filter; 15—Stack; 16—Induced fan; 17—Raw fly ash; 18—Air; 19—Tailings; 20—Landfill disposal.

4. Concluding Remarks and Future Work

MWIFA contains high amount of chlorides, carbon constituents, POPs, and toxic heavy metals, which makes it more difficult to handle compared with MSWIFA. The management of incinerator fly ash will become stricter in the near future, especially for MWIFA. It is necessary to overcome the shortcomings of the existing treatment of MSWIFA and find alternatives which are effective and economical countermeasures to treat MWIFA.

1. The pretreatment methods of acid leaching and water washing are applied to remove chlorine and heavy metals from fly ash, respectively. Both are worth promoting for MWIFA treatment by collaborating with other post-treatment methods. HCl and H_2SO_4 are the most efficient lixiviants at present. However, insoluble chlorides are difficult to remove with the water washing process and wastewater generated must also be treated. These drawbacks should be overcome.
2. The post-treatment methods, such as roasting, residual carbon melting, the mechanochemical technique, flotation, and microwave treatment are recommended for the treatment of MWIFA after overall consideration of the special characteristics of MWIFA, and thorough consideration and balancing of environmental, technological, economical information.
3. Cement solidification can prevent heavy metals leaching out but detoxification of dioxins is unable to realize. Furthermore, the characteristics of high chloride and carbon contents for MWIFA tend to weaken the effect of cement solidification. Thus, a future study is needed to focus on eliminating the influence of chlorides and carbons or thoroughly removing both substances.
4. Melting can efficiently destroy dioxins and stabilize heavy metals; however, its high energy consumption and investment cost restrain its widespread application. This technology is theoretically unsuitable for the treatment of MWIFA at a small scale, except for residual carbon melting furnaces.
5. Roasting treatment is suitable for the treatment of MWIFA and can decompose dioxins and recover heavy metals, which fully combines the characteristics of high chlorines and heavy metals in MWIFA.
6. Intermediate treatment methods such as low-temperature thermal treatment, catalytic hydro-dechlorination, SCWO, and hydrothermal treatment, are effective in the decomposition of dioxins in MWIFA. However, there is limitation in the treatment of heavy metals in MWIFA. In addition, these methods remain in the laboratory testing stage, and some operations may cause severe secondary pollution or dioxin regeneration.
7. The mechanochemical technique is effective at decomposing dioxins and stabilizing/removing heavy metals. It is low-cost, environmental friendly, and has no tail gas production. However, mechanochemical techniques remain in the development stage. Unknown difficulties must be overcome for future application.
8. Flotation technology is recommended for handling MWIFA containing a high content of carbons, dioxins, chlorines, and heavy metals. This technology can efficiently remove carbon constituents, dioxins, and heavy metals from MWIFA. It is helpful to evaluate the harmlessness, reduction, and resource recovery of MWIFA. A combination of flotation and reburning treatment may be an especially promising method to resolve difficulties of MWIFA treatment.
9. The microwave method makes full use of the specific characteristics of MWIFA through a strong microwave-absorbing medium, PAC. After microwave treatment, efficient decomposition of dioxins and solidification of heavy metals can be achieved. Meanwhile, a combination of flotation and microwave treatment can achieve both heavy metal removal and dioxin decomposition. Most successful cases have been demonstrated on the lab scale; however full-scale application is still in development. Comprehensive comparison of treatment techniques of MWIFA is given in Table 10.

Table 10. Comprehensive comparison of treatment techniques of MWIFA.

Technique	Applicability to the Characteristics of MWIFA	Detoxification Effect		Cost of Treatment	Environmental Feedback	Technique Status	Merits	Demerits	Remarks
		Heavy Metals	Dioxins						
WWP	Removal of chlorines	No stabilization	No decomposition	Low	Environmental friendly.	established	Chlorine removal	Waste liquid may need to be treated	pretreatment method
ALP	Leaching out heavy metals	No stabilization	No decomposition	Low	Environmental friendly.	established	Heavy metal leaching	Waste liquid may need to be treated	pretreatment method
CST	High PAC reduces the compressive strength and increases metal leachability	stabilization	No decomposition	Low/moderate	Questionable	Mature	Easy implementing and low expense	consumes mass of cement; products cannot be used; landfill sites are limited	just one of the expedients
MT	High carbon content is harmful to graphite electrode	Partial stabilization	Decomposition	High	Existing a secondary pollution	Developing/ Developed	molten glasses or slag byproducts can be used as road/construction materials	High expense and energy consumption. Metal chlorides were easy volatile.	residual carbon melting furnace is suitable
Roasting	It is suitable for chloride-/ metal-enriched ash	Recovery	Decomposition	Moderate	Slight pollution	Developed	No mentioned	higher facility requirements, energy consumption/costs	Theoretically applicable
LTTT	No mention	Partly stabilization	Decomposition	Moderate	Slight pollution	Developed	Simple operation and low energy consumption	Heavy metals were not considered.	can be a sequential dioxin removal step
CHD	Facilities are easily corroded by HCl.	No stabilization	Decomposition	Moderate	Potentially pollution	laboratory stage	High efficiency, strong selectivity, stable reaction	dioxin regeneration may happen	can be a sequential dioxin removal step
SCWO	Applicability for MWIFA is unknown	Stabilization	Decomposition	High/moderate	Potentially secondary pollution	Developing	Effective, harmless, fast, and violent, and no heat transfer resistance problem	high requirement for equipment and high energy consumption.	Dioxins may regenerated.
HTT	Less Si and Al were bad for the formation of zeolite and tobermorite crystalline	Partial stabilization	Decomposition	Low/moderate	Less pollution	small scale engineering	Energy saving, low facility requirement, and products can be reused.	Catalysts are liable to be poisoned by toxic methanol. Facility is easily corroded	Obstacles need to overcome
MCT	No mentioned	Partial removal or stabilization	Decomposition	Moderate	Environmental friendly.	small scale engineering	Nonthermal process and nontail gas disposal method	Unknown difficulties may need be overcome	has bright prospect of industry application
Flotation	Suitable for high carbon, chlorines and dioxins	Removal	Removal	Low	Environmental friendly	Emerging	Flexibility, less reagent usage,	No actual engineering application	pretreatment method, flotation + reburning is promising
MWT	Suitable for high carbon, and dioxins	Stabilization	Decomposition	Moderate	Less pollution	small scale engineering	Rapid/interior/ volumetric/selective heating	Obstacles need to overcome for industry application	Flotation + MWT is a promising method

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