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Investigation of Mechanochemically Treated Municipal Solid Waste Incineration Fly Ash as Replacement for Cement

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Abstract: Municipal solid waste incineration (MSWI) fly ash has been classified as hazardous waste in China because of the leachable toxic heavy metals and high concentrations of chlorides and polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDD/Fs). Currently, the main treatment method is still landfilling after chemical treatment or cement solidification, and an effective approach to realize fly ash utilization is still lacking. In the present work, the fly ash was firstly water-washed to remove the soluble chlorine salts, which can improve the performance of the produced cement mortar in later work. Mechanochemical pre-treatment was adopted to destroy the PCDD/Fs and improve the heavy metals' stabilization. The results show that 75% of PCDD/Fs can be degraded and that most of the heavy metals are stabilized. After the mechanochemical pre-treatment, the average particle size of the fly ash decreases to 2–5 μm, which is beneficial for promoting the activation energy and accelerating the hydration process in cement mortar production. The compressive and flexural strengths of the fly ash cement mortar improve to 6.2 MPa and 32.4 MPa, respectively, when 35% of the OPC is replaced by treated fly ash. The similarity in the 3-day and 28-day strength with or without the addition of the treated ash shows the light influence of the fly ash addition. Thus, the mechanochemical process can stabilize the heavy metals and activate the fly ash, allowing it to partly substitute ordinary Portland cement in building materials, such as cement raw materials and concrete.

Keywords: mechanochemical treatment; fly ash; PCDD/Fs; heavy metals

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1. Introduction

Incineration has been the major approach to reduce the amount of municipal solid waste in China. A total of 136 million tons of solid waste were incinerated, and more than 500 incineration plants were run in 2021 in China [1]. However, large amounts of fly ash (FA) are produced from municipal solid waste incineration (MSWI), with a production amount of more than 8 million tons per year, which is still increasing due to the increasing amount of solid waste [2]. MSWI FA is intercepted by the baghouse filter in air pollution control devices. MSWI FA is classified as hazardous solid waste because of the leachable toxic heavy metals, such as Cu, Cd, Zn, and Pb, and chloride salts that are leachable; thus, FA can be a serious environmental risk [3–5]. Moreover, polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are formed and adsorbed in fly ash in a concentration range of 76–2652 ng TEQ/kg [6,7], which has caused widespread concern due to the harmful effects on human health and the environment [8]. Currently, the main treatment method is still landfilling after chemical treatment or cement solidification [9]. However, the land in East China is currently seriously scant, and landfilling is not a sustainable solution for MSWI FA treatment [10].

Due to the high concentrations of CaO, SiO_2, Al_2O_3, Fe_2O_3, MgO, etc., in fly ash, several researchers have studied the utilization and recovery of MSWI fly ash to prevent adverse environmental risks, including its utilization as a raw material for tile manufacturing and as a substitute for ordinary sand in concrete [11–14]. In previous studies, coal-fired fly ash and MWSI bottom ash have been studied for their potential use in concrete, road pavement, ceramics, and so on [15–20]. When MSWI fly ash is added to cement-based products, to meet the technical and environmental requirements, such as enough strength, durability performance, and leaching limits of heavy metals from the products, pre-treatment is necessary to reduce the leaching of hazardous components [21,22]. Thermal pyrolysis was studied to treat the MSWI fly ash and the PCDD/Fs concentrations can be lower than 20 ng TEQ/kg after the pyrolysis process at 350 °C [8]. However, thermal pyrolysis at a low temperature cannot reduce heavy metal leaching concentrations [23]. Fly ash co-processing in cement kilns has been adopted to destroy hazardous waste in China; however, the restricted addition amount because of the chlorine concentration has limited the utilization of fly ash [24].

Phosphorus-based inhibitors can efficiently prevent dioxin formation in the post-combustion zone but have a poor effect on the heavy metals' solidification in fly ash [25]. To achieve complete detoxification of fly ash and make it recyclable, further detoxification is needed. The mechanochemical (MC) process is simple and can simultaneously solidify heavy metals and degrade PCDD/Fs in fly ash [26]. Birke et al., performed mechanochemical treatment by adding magnesium, alcohols, and ethers to achieve complete dehalogenation of POPs in several minutes [27]. Lu et al., used a mechanochemical method with CaO/SiO_2 addition to destroy 2,4,6-trichlorophenol, and a 99% degradation efficiency was achieved [28]. Yan et al., studied the performance of mechanochemical treatment to destruct PCDD/Fs with CaO addition, and 90% of the PCDD/Fs were destroyed [29]. By analyzing the dichlorination and fingerprints of PCDD/Fs, destruction and dichlorination were proposed as major mechanisms for PCDD/Fs degradation [29]. Selena et al., used hydroxyapatite as the agent to treat soil containing heavy metals such as Cd, Zn, and Pb by a mechanochemical method, in which immobilization rates of 99.89%, 97.44%, and 99.7% were obtained, respectively [30]. Chen et al., found that $Ca_3(PO_4)_2$ addition facilitated the mechanochemical stabilization of toxic heavy metals in fly ash, and the Nemerow pollution index reduced from 9.35 to 0.71 [26]. Yuan et al., used mechanochemical treatment to improve heavy metal stabilization in coal-fired fly ash, and the water-soluble and acid-soluble components of Cu, Pb, and Cr decreased by more than 25% [31]. Geng et al., found that the mechanochemical process was beneficial for stabilizing Pb in fly ash because it crushed FA particles and destroyed the crystal structure, further developing its surface area and generating more surface active sites, which was beneficial to increasing the diffusivity and adsorption ability of heavy metals in FA [32].

Moreover, particle refinement contributes to improving the mechanical performance of the produced cement mortar by promoting the pozzolanic and hydration activity [33]. Decreasing the particle size will promote the activation energy and accelerate the hydration process [33,34]. Cordeiro et al., (2008) studied the addition of sugar cane bagasse incineration ash to cement mortar and found that the fine particles boosted the pozzolanic reactions [35].

In this study, a mechanochemical method was adopted to pre-treat municipal solid waste incineration fly ash and promote its characteristics. The evolution of PCDD/Fs and the leaching characteristics of heavy metals in the fly ash were comprehensively studied. In addition, raw and treated fly ash were used as partial substitutes for ordinary Portland cement, and the strength performance and heavy metal leaching of the fly ash cement mortar were investigated.

2. Materials and Methods

2.1. Fly Ash Samples

The fly ash sample was collected from the baghouse filter in a solid waste incineration plant, which has a total treatment capacity of 2250 t/d with three 750 t/d incinerators. The incinerator studied in this work was equipped with a circulating fluidized bed furnace, and the furnace temperature was kept above 850 °C to prevent dioxin formation. The solid waste was stored and fermented for more than one week before burning to remove the water and increase the calorific value. Typically, the air pollution control devices (APCDs) for this incinerator include selective noncatalytic reduction (SNCR), a semidry scrubber, and activated carbon injection coupled with a baghouse filter. To ensure that the dioxins emissions met the national standard, the APCDs were ungraded with the addition of an inhibitor injection system at the flue pipe, where the flue gas temperature was approximately 500 °C. A novel S–N–P-based inhibitor was chosen with a flow of 20 kg/h. Attributed to the advanced combustion technology and APCDs, the initial dioxin concentration in the fly ash was only 255 ng TEQ/kg, much lower than that reported in reference [7]. Before use, the fly ash was water-washed twice to remove the chlorine with a solid:liquid ratio of 1:6. After water washing, the soluble chlorine concentration was 0.86%, which is lower than the national standard of 1%.

2.2. Mechanochemical Treatment of Fly Ash

To study the effect of mechanochemical treatment on the heavy metals and dioxins, planetary ball milling equipment (QXQM-2-DW, Changsha Tianchuang, China) was adopted in this study. A total of 15 g fly ash was put in the zirconia ball milling tank (250 mL) with zirconia spheres (total mass of 375 g, diameters of 10 mm and 5 mm, and mass ratio of 1:1). The rotation speed of the planetary disk was 500 rpm, and the durations were 6 h and 10 h.

CaO was adopted as the addition in ratios of 15%, 30%, and 45%, which was beneficial for destroying the PCDD/Fs and solidifying heavy metals. The sample abbreviations under different operating conditions were water-washed fly ash (WFA) and mechanochemically treated fly ash (MFA-x-y, where x is the amount of CaO addition and y is the milling time).

2.3. Fly Ash Cement Mortar Preparation

The effects of WFA or MFA addition on the mechanical properties and the heavy metal leaching concentrations of the fly ash cement mortar were examined. The ordinary Portland cement (OPC) was used as the raw material, which has a density of 3.15 g/cm^3 with a surface area of 3460 cm^2/g. The mortar was mixed with a mass ratio of 2:6:1 of binder, fine aggregate, and water. In the experiments, WFA, MFA-0%-10 h, MFA-15%-10 h, MFA-30%-10 h, and MFA-45%-10 h were used to replace OPC. To evaluate the effect of the fly ash addition on the mortar performance, fly ash replaced 35%, 50%, and 60% OPC, as shown in Table 1.

Table 1. Ratio of fly ash used to replace OPC (wt %).

	WFA	MFA-0%	MFA-15%	MFA-30%	MFA-45%	OPC
SN0						100
SN1	35					65
SN2		35				65
SN3			35			65
SN4			50			50
SN5			65			35
SN6				35		65
SN7				50		50
SN8				65		35
SN9					35	65
SN10					50	50
SN11					65	35

In accordance with the Chinese standard (GB/T 17671), after mixing, the mortar was poured into steel molds. Then, the molds were cured at 20 °C with 95% relative humidity for 24 h. After the mortar had cured, it was taken out from the mold and placed in water with a constant temperature of 20 °C. Compressive strength tests of mortar samples were performed after 3 and 28 days according to GB/T 17671. The pretreatment and experiment process is shown in Figure 1. The heavy metal leaching concentrations were also tested by ICP.

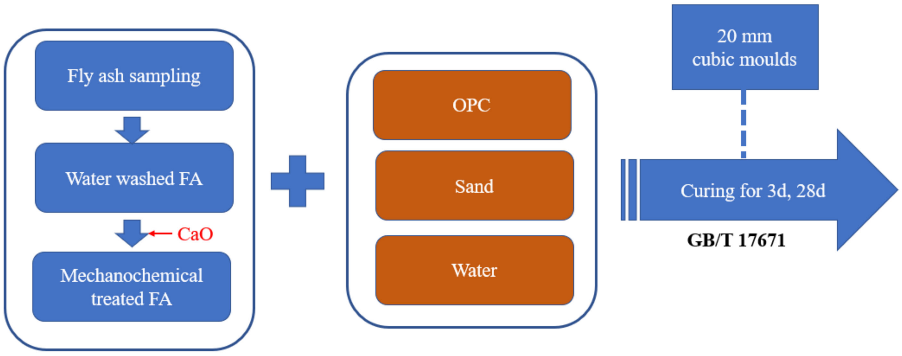

Figure 1. The schematic of the experiment procedure.

2.4. Characterization and Analysis

The chemical composition of the fly ash samples was determined by X-ray fluorescence spectroscopy (XRF, SMS-Omega XRF, Thermo Fisher, Waltham, MA, USA). The chlorine content was tested by ion chromatography (Integrition, Thermo Fisher).

The fly ash was pre-treated according to US EPA 23, and the PCDD/Fs concentrations were measured by high-resolution gas chromatography/high-resolution mass spectrometry (HRGC/HRMS, JMS-800D, JEOL, Tokyo, Japan). First, the fly ash samples were soaked for 4 h in HCl solution to remove the metallic impurity. Then, the concentrated solutions were transferred to acid silica columns and activated carbon mini-columns to remove metals and other organic pollutants. In the pre-treatment process, samples were spiked with ^{13}C-labeled standard compounds of PCDD/Fs. Finally, the treated samples were analyzed with HRGC/HRMS with DB-5 MS column (60 m $\times$ 0.25 mm $\times$ 0.25 mm). HRMS was operated in electron ionization and selected ion monitoring modes with a resolution of >10,000. The detailed pre-treatment and analysis processes were introduced in our previous work [36].

The heavy metals were extracted from the samples following Chinese national standard HJ/T 300-2007 ("Solid Waste-Extraction Procedure for Leaching Toxicity—Acetic Acid Buffer Solution Method"). The samples were detected with an iCAP 6300 instrument (Thermo Fisher).

3. Results and Discussion

3.1. Characterization of the Raw and Treated Fly Ash

The compositions of the raw fly ash were determined by XRF, and the results show that Ca, Si, and Al dominate fly ash. However, the chlorine content in the present work is only 6 wt %, which is much lower than contents from incinerators without an inhibition system [37]. The low content of chlorine is consistent with the low PCDD/Fs concentration, which is only 255 ng TEQ/kg. The S–N–P inhibitor helps to reduce the PCDD/Fs formation in two ways. On the one hand, the S from the inhibitor is oxidized to SO_2, which can inhibit Cl_2 conversion from HCl and decrease dioxin formation. On the other hand, the conversion to $CuSO_4$ from $CuCl_2$ with SO_2 is an important factor to reduce organic chlorine formation [38].

Figure 2 and Table 2 shows the particle size distribution of the OPC, WFA, and MFA. The average particle size of the WFA is 56 µm, which is larger than that of OPC (8 µm). With the treatment of the mechanochemical method, the average particle size decreases to 2–5 µm, showing three peaks of the treated fly ash. When the average particle size is lower than 10 µm, the force between particles increases significantly, and the particle size does not decrease but remains in a reasonable range [39].

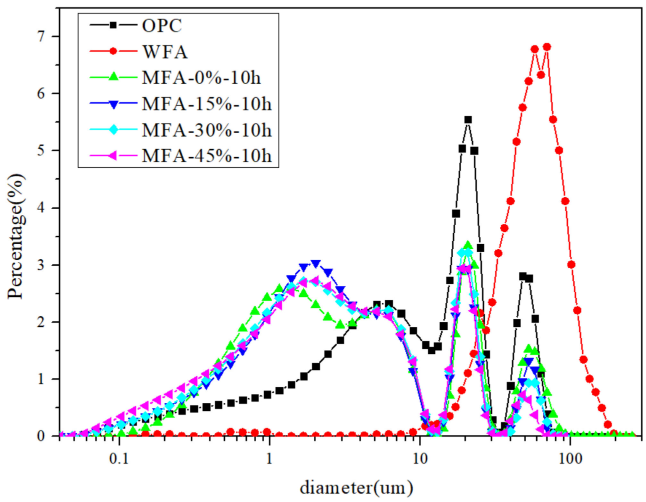

Figure 2. Particle size distribution of the samples under different operating conditions.

Table 2. D50 and D90 of the samples under different operating conditions (μm).

Samples	D50	D90
OPC	16.1	45.2
WFA	49.6	96.5
MFA-0%-10 h	4.2	46.8
MFA-15%-10 h	3.3	23.7
MFA-30%-10 h	3.2	23.1
MFA-45%-10 h	2.7	21.4

3.2. PCDD/Fs and Heavy Metal Concentrations before and after MC Treatment

After being washed with water, the water-soluble chlorides, such as $CaCl_2$, KCl, and NaCl, easily washed away. The total PCDD/Fs concentrations increased from 5035 in the raw fly ash to 5671 ng/kg after water washing. The TEQ concentration of water-washed fly ash (WFA) was 230 ng TEQ/kg. After water washing, the water-soluble chlorides concentration was 0.86%, which can reduce the PCDD/Fs formation in the mechanochemical process.

Figure 3 shows the changes in the PCDD/Fs TEQ of fly ash under different working conditions. Without the addition of CaO to fly ash, the degradation efficiency of PCDD/Fs is quite low. After 10 h of mechanochemical treatment, only 10.9% of the PCDD/Fs was destroyed. When 15% CaO was added to the fly ash, the destruction efficiency reached 22.3% after 6 h of treatment and increased to 35.4% with 10 h of milling. In particular, the PCDD/Fs concentration was only 55.82 ng TEQ/kg with a 45% addition, indicating that a certain amount of CaO can better promote the PCDD/Fs degradation in fly ash by mechanochemical methods. The degradation rate increases with increasing addition ratio and milling time. Compared to the results in a previous work [40], the destruction efficiencies in the present work are much lower. Wei et al., also found a similar phenomenon in their study [41]. In this work, the initial PCDD/Fs concentration is much lower due to the inhibitor addition in the flue gas. The destruction efficiency is much lower when the PCDD/Fs concentration in fly ash is low due to less contact between balls and dioxin pollutants.

By analyzing the PCDD/Fs fingerprints in the raw and treated fly ash, changes in the contents of 17 kinds of 2,3,7,8-polychlorinated toxic PCDD/Fs were observed. According to Figures 4 and 5, after 6 h of ball milling without CaO addition, the OCDD content decreased by 29.6%, while after 10 h of ball milling, the OCDD content continued to decrease to 1.295 ng TEQ/kg, resulting in a degradation rate of 41.2%. In addition, MC treatment has an obvious effect on the degradation of 1,2,3,4,6,7,8-HpCDD, 1,2,3,4,6,7,8-HpCDF, and OCDF. This shows that without additives in the MFA-0%, the PCDD/Fs can still be degraded, and when CaO is added, the CaO in the raw fly ash can help to dechlorinate the PCDD/Fs [40].

Compared to MFA-0%, the degradation efficiency of the samples with the CaO addition obviously improved, and the degradation efficiency increased with increasing addition ratio and ball milling time. When the CaO addition amount was 45%, the degradation rate reached 88.1% after 10 h of ball milling, indicating that CaO has a better promoting effect on PCDD/Fs degradation. Moreover, it is worth noting that when the addition ratio increased from 30% to 45%, the degradation efficiency improvement rate was quite low [28]. In addition, it can be observed from Figures 4 and 5 that the mechanochemical method has a suitable degradation effect on high-chlorinated PCDD/Fs but a mediocre degradation effect on low-chlorinated PCDD/Fs.

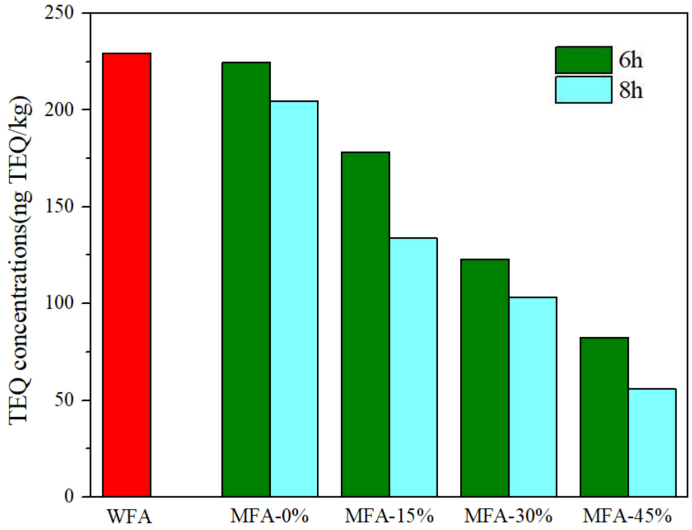

Figure 3. TEQ concentrations of the MC-treated fly ash.

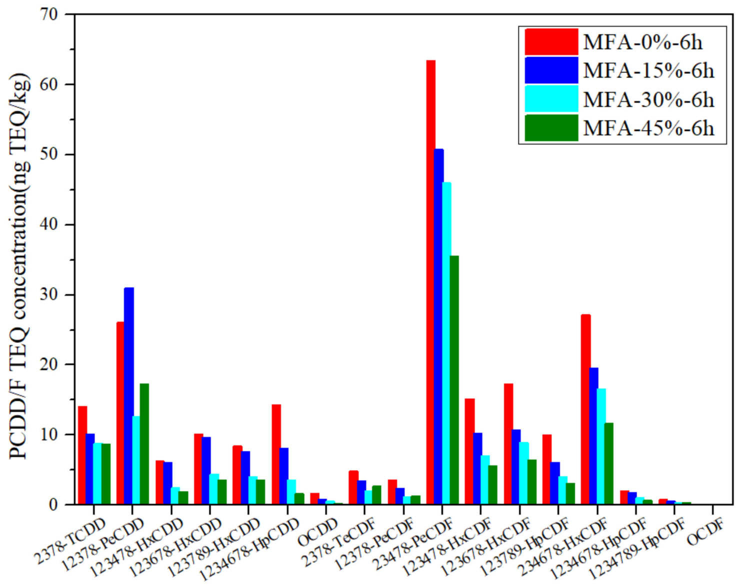

Figure 4. Concentrations of 17 2,3,7,8-substitued PCDD/Fs after MC treatment for 6 h.

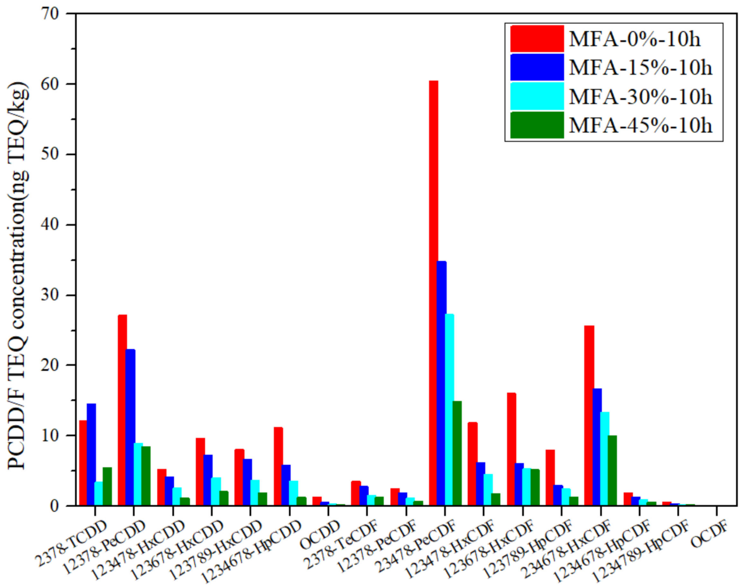

Figure 5. Concentrations of 17 2,3,7,8-substitued PCDD/Fs after MC treatment for 10 h.

Figures 4 and 5 show the changes in the toxicity equivalent of 17 toxic PCDD/Fs under four different working conditions. It can be seen from the figure that mechanochemistry treatment has a significant degradation effect on 17 toxic PCDD/Fs toxic equivalents, and the largest contribution of the toxicity equivalent is still PCDFs, especially 2,3,4,7,8-PeCDF, which is approximately 30%. It is worth mentioning that mechanochemistry has a light effect on degrading low-chlorinated PCDD/Fs, especially 2,3,7,8-TCDD, 1,2,3,7,8-PeCDD, and 2,3,4,7,8-PeCDF. The reason may be that on the one hand, high-chlorinated PCDD/Fs are easily converted into low-chlorinated PCDD/Fs, which exhibit higher toxicity [29]. On the other hand, the chlorine at position 2,3,7,8 is not easy to remove, and its toxicity factor is relatively high, resulting in a high TEQ concentration [42].

Table 3 shows the heavy metal leaching concentrations under different working conditions. It shows that after the mechanochemical treatment, the heavy metal leaching concentration in the fly ash decreases, indicating that mechanochemical solidification can block the heavy metals. Li et al., (2010) showed that the change in the crystal structure of fly ash by mechanical force is the main reason for its ability to solidify heavy metals [43]. When CaO is not added, most of the heavy metals are stabilized after mechanochemical treatment, but the Ni content increases, which may be caused by the wear of the grinding balls or tank walls. In addition, the leaching concentration of Cd still exceeds landfill standards. When 15% CaO is added, the leaching concentration of each heavy metal is far lower than the landfilling standard. This shows that CaO has a favorable synergistic effect on the heavy metals in the solidified fly ash. Studies have shown that CaO reduces the leaching concentration of heavy metals in fly ash by increasing the pH value of the leaching solution and leading to part of the heavy metals in the solution being adsorbed [33,43].

Table 3. Leaching concentrations of heavy metals in the different fly ash (mg/L).

Heavy Metal	Cd	Cr	Ni	Cu	Pb	Zn	PH
Standard	0.15	1.5	0.5	40	0.25	100	2–12.5
WFA	0.41	0.66	0.40	9.45	0.68	35.73	7.2
MFA-0%-6 h	0.32	0.06	0.50	1.25	0.30	22.67	7.8
MFA-15%-6 h	0.006	ND	ND	0.2	0.005	0.02	8.2
MFA-30%-6 h	0.002	ND	ND	0.06	0.002	0.004	8.9
MFA-45%-6 h	ND	ND	ND	0.02	0.002	0.002	9.6

ND: not detected.

3.3. Compressive Strength of the Fly Ash Cement Mortar

Figures 6 and 7 show the compressive and flexural strengths of cement mortar prepared by OPC and partly substituted by fly ash according to GB/T 17671, respectively. The ratios of OPC substituted by raw or treated fly ash are shown in Table 1. For all cement mortar blocks, the flexural and compressive strengths at 28 days are greater than those at 3 days, and their mechanical strength properties decrease with increasing fly ash content. Compared with the cement mortar without fly ash, the flexural and compressive strength of the cement mortar greatly decrease when the amount of added WFA is 35% (decrease from 9.3 and 64.8 MPa to 5.6 and 22.3 MPa, respectively). The water-washed fly ash addition has a notable negative effect on the strength of the cement mortar. The flexural and compressive strengths of the fly ash mortar improve to 6.2 MPa and 32.4 MPa, respectively, when 35% of the OPC is replaced by MFA-0%. When the same proportion of MFA-15%-10 h is added, the strength of the cement mortar improves significantly. Its 28-day flexural and compressive strengths are 7.9 MPa and 45.7 MPa, respectively. When 35% MFA-30%-10 h is added, its 28-day flexural and compressive strengths are 8.2 MPa and 51.6 MPa, respectively. The flexural strength reaches 88.2% of the pure cement mortar, and the compressive strength reaches 79.6% of the pure cement mortar. Even when the addition ratio is increased to 65%, the flexural and compressive strengths at 28 days can reach 7.4 MPa and 43.5 MPa, indicating that the performance of MFA-30%-10 h is greatly improved after mechanochemical treatment with the CaO addition. When 35% MFA-45%-10 h is added, the strength is lower than the MFA-15%-10 h and MFA-30%-10 h cement mortars, and its 28-day flexural and compressive strengths are only 6.1 MPa and 32.7 MPa, respectively. The results indicate that when the amount of free calcium is too high, it will affect the stability of cement and reduce the strength [44]. In conclusion, the performance of fly ash treated by the mechanochemical method is significantly improved. The fly ash after water washing and mechanochemical treatment can be used as cement.

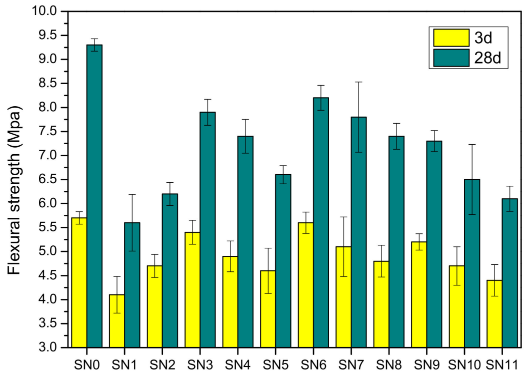

Figure 6. Flexural strength of the cement mortar.

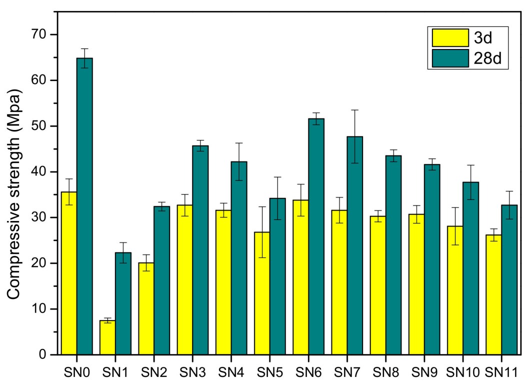

Figure 7. Compressive strength of the cement mortar.

3.4. Leaching Concentration of Heavy Metals in the Fly Ash Cement Mortar

Table 4 shows the leaching concentrations of heavy metals in the cement mortar under different working conditions. The results show that the leaching concentrations of heavy metals under all working conditions are quite low. Comparing SN1 to SN0, the 35% fly ash addition can increase the leaching concentration of cement mortar. When MC-treated fly ash is added, the leaching concentrations of heavy metals in cement mortar are much lower than those substituted by raw fly ash. The results of SN3–SN5 indicate that the amount of treated fly ash that is added has an important influence on the leaching of heavy metals in the cement mortar, even though all the values meet the standard. With increasing CaO amounts, the performance of the treated fly ash shows a better stabilization effect on heavy metals. The leaching concentrations of heavy metals in SN6–SN11 are much lower than those prepared by pure cement, indicating that MC-treated fly ash can be used in building materials, such as cement raw materials and concrete.

Table 4. Leaching concentration of heavy metals in cement mortars (mg/L).

Heavy Metal	Cd	Cr	Cu	Ni	Pb	Zn
Standard	0.15	1.5	0.5	40	0.25	100
1-7 SN0	ND	0.338	0.003	0.057	ND	0.013
SN1	0.25	2.643	0.422	0.106	ND	0.506
SN2	0.005	0.228	0.040	ND	ND	0.312
SN3	0.004	ND	0.020	ND	ND	0.173
SN4	0.003	ND	0.027	ND	ND	0.170
SN5	0.048	ND	0.048	ND	ND	0.723
SN6	ND	ND	ND	ND	ND	0.002
SN7	ND	ND	0.002	ND	ND	0.001
SN8	ND	ND	0.007	ND	ND	0.013
SN19	ND	ND	0.001	ND	ND	0.001
SN10	ND	ND	0.003	ND	ND	0.001
SN11	ND	ND	0.002	ND	ND	0.002

ND: not detected.

4. Conclusions

In this work, we used a mechanochemical method with CaO as an additive to destroy PCDD/Fs and stabilize the heavy metals in water-washed fly ash. The results show that the PCDD/Fs concentration decreased to 55.8 ng TEQ/kg after mechanochemical treatment, with a degradation efficiency of 75.6%. After MC treatment with 15% CaO addition, the leaching concentrations of heavy metals met the landfilling standard in China. In particular, the leaching concentration of Pb decreased from 0.68 to 0.005 mg/L. Furthermore, raw fly ash and MC-treated fly ash were used as substitutes for cement in cement mortar production. The performance of fly ash obviously improved after MC treatment. When 35% cement was replaced by MC-treated fly ash, the 28-day flexural and compressive strengths were 8.2 MPa and 51.6 MPa, respectively, reaching 88.2% and 79.6% of those of pure cement mortar, showing a satisfactory mechanical performance. The heavy metal leaching concentrations of the fly ash cement mortar were quite low, close to those of the pure cement mortar. The results indicate that MC-treated municipal solid waste fly ash showed a performance similar to that of cement and can be used in building materials, such as cement raw materials and concrete.

Author Contributions: S.P.: investigation, methodology. J.D.: data curation, visualization, writing—original draft. Y.P.: conceptualization, funding acquisition, writing—review and editing. S.L. and X.L.: conceptualization, supervision. All authors have read and agreed to the published version of the manuscript.

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