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Research on the Optimization and Application of the Washing Dechlorination Process for Municipal Solid Waste Incineration Fly Ash

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ABSTRACT: In this paper, the fly ash cyclic gradient washing dechlorination process is systematically studied through experiments, and the effects of process parameters such as liquid−solid ratio, the number of leaching, and process pulping on the dechlorination effect of fly ash are investigated and analyzed with the currently operating three-stage counter-current washing dechlorination process. The experimental results indicate that with the liquid−solid ratio of 3:1, the number of leaching of 4, and the primary process pulping, the chlorine content of washing fly ash is reduced to 0.5−0.6%. The Baume degree in the washing filtrate is increased to 11−12 °Bé, the total amount is reduced by about 15%, and the average turbidity value is ≤5NTU. Meanwhile, the moisture content of the washing fly ash is reduced to 28−30%. By comparing with the actual construction project, it is found that under a disposal capacity of 100 t/d, the cyclic gradient washing dechlorination process can reduce the installed power by 30.3%, the floor space by 32.9%, the treatment volume of washing filtrate by 11.1%, and the drying load by 27.9% compared to the traditional three-stage counter-current washing and dechlorination process.

1. INTRODUCTION

With the development of the social economy and the improvement of residents' living standards, residents' domestic waste has increased yearly.[1−4] According to the data of the China Statistical Yearbook,[5,6] in the five years from 2016 to 2020, the amount of municipal solid waste incineration (MSWI) in China increased by about 98%. A development pattern has gradually formed in which incineration mainly treats new waste. It is estimated that by the end of 2025, the treatment capacity of MSWI in China will reach about 800,000 tons/day.[7] A large amount of fly ash is produced during domestic waste incineration.[8−11] Because it contains toxic and hazardous substances such as heavy metals and dioxins, it is listed on the National Hazardous Waste List.[12] The resource disposal of fly ash is a widespread concern in society.[13−15]

In addition to soluble salts, the composition of MSWI fly ash is mainly CaO, SiO_2, and Al_2O_3, which are similar to the raw materials of the building material industry.[16,17] The resource disposal of fly ash from domestic waste incineration shows a diversified development trend, mainly including cement kiln co-processing,[18] high temperature/plasma melting,[19,20] and catalytic pyrolysis.[21] Among them, cement kiln co-processing of fly ash is currently the main way for resource utilization of fly ash in China. The first cement kiln co-processing line for MWSI fly ash in China was put into operation in Fangshan, Beijing, in 2012.[22] By the end of 2021, more than 10 fly ash cement kiln co-processing production lines were put into operation in China. The capacity of cement kiln co-processing fly ash has reached 100 wt/a, but compared to the amount of fly ash generated, there is still a huge gap.

The MAWI fly ash contains high chlorine salts, which have strong corrosiveness to equipment and influence the quality of building materials in resource utilization.[16,23] Washing fly ash can remove soluble chlorine economically and effectively, which has become the main form of dechlorination of fly ash at home and abroad.[24,25] Previous studies have shown that chlorine in fly ash is divided into soluble and insoluble chlorine. Insoluble chlorine is difficult to be removed by simple washing, and fly ash washing dechlorination is mainly for soluble chlorine removal.[26] The main process parameters of fly ash washing dechlorination, such as the number of leaching, leaching time, liquid−solid ratio, and water temperature, have different influences on the dechlorination effect of fly ash.[27−29]

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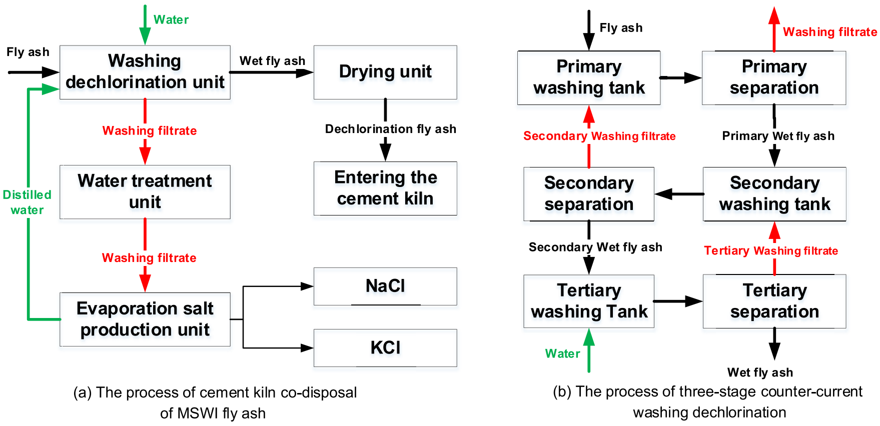

Figure 1. Schematic process flow of fly ash co-processing.

The research of Wang shows that when the washing liquid−solid ratio is 10 mL/g, the washing time is 28 min, the water temperature is 30 °C, and the stirring speed is 450 rpm, the removal of chlorine element in fly ash reaches 93.3% through two cycles of washing.[30] The high solid−liquid ratio is beneficial for fly ash washing dechlorination, but it also increases the load on the subsequent wastewater treatment. To achieve the same dechlorination effect of fly ash washing, the liquid−solid ratio can be reduced by increasing the number of leaching. Wang constructed a three-stage counter-current process, with the washing liquid−solid ratio of 6 mL/g and washing time of 20 min, and the elemental chloride removal from the fly ash reached 95.2%.[31] The above research mostly stays at the experimental stage without considering the influence of the liquid−solid ratio on the process of engineering applications.

At present, the commonly used three-stage counter-current washing dechlorination process for fly ash has problems such as high investment, long process flow, more process equipment, large floor space, high operational-energy consumption, high moisture content of washing fly ash, and incomplete separation of washing filtrate. Therefore, it also causes problems such as complex treatment processes for subsequent washing filtrate, high operational energy consumption for drying of washing fly ash, and evaporation of washing filtrate for salt production. To solve the above engineering problems, this paper proposes a fly ash cyclic gradient washing dechlorination process and optimizes the process parameters of liquid-solid ratio, the number of leaching, and primary process pulping through the experimental research. The optimal cyclic gradient washing dechlorination operation parameters are obtained, which are applied in engineering practice to verify the feasibility of the process.

2. PROCESS PROGRAM DETERMINATION

2.1. Analysis of the Mechanism of Dechlorination in the Process of Washing Fly Ash. The effect of washing dechlorination of fly ash is related to the form of chloride ions in the fly ash, which is primarily determined by the distribution characteristics of the chloride ions in fly ash. Insoluble chlorine in fly ash mainly exists in AlOCl and Ca_4OCl_6 that cannot be removed by simple washing.[32] Soluble chlorine mainly exists in NaCl, KCl, and CaClOH,[16] accounting for 82−96% chlorine content in fly ash.[33] Research shows that NaCl and KCl are enriched in fly ash with flue gas condensation after evaporation in the process of MSWI. CaClOH in fly ash is the intermediate product generated by the reaction of excess $CaO/Ca(OH)_2$ with HCl during flue gas deacidification,[34] and the reaction process is as follows:

$$CaO(s) + HCl(g) \rightarrow CaClOH(s) \tag{1}$$

$$Ca(OH)_2(s) + HCl(g) \rightarrow CaClOH(s) + H_2O \tag{2}$$

$$Ca(OH)_2(s) + CaCl_2(s) \rightarrow CaClOH(s) \tag{3}$$

The washing dechlorination of fly ash is the process of removing chlorine in soluble form. The removal of soluble components from the solid state can be divided into two physical processes: dissolution and desorption. The study shows that the washing process is mainly dissolution when the liquid−solid ratio is less than 20. When the liquid−solid ratio is more than 20, the washing process is mainly desorption.[35] In this paper, the experimental study of the washing dechlorination process has a liquid−solid ratio ≤3, and the elution of chloride ions is dominated by dissolution. In washing dechlorination of fly ash, soluble chlorine salts (such as NaCl and KCl) in fly ash are quickly dissolved in water. In addition, CaClOH reacts with water to form soluble $CaCl_2$, which is then dissolved in water. The reaction is as follows:

$$CaClOH(s) + H_2O(l) \rightarrow Ca(OH)_2(aq) + CaCl_2(aq) \tag{4}$$

The process of dissolving chlorine salts in water is mainly related to the solubility of chlorine salts. The dissolution of Ca is also internally affected by the solubility and ionization balance of $CaSO_4$ and $CaCO_3$ in fly ash.[36] In this process, the dissolution of Cl^- is the largest, followed by Ca, Na, and K, so as to achieve the purpose of dechlorination.[37] In the industrial production process of fly ash washing dechlorination, the dechlorination effect is influenced not only by the liquid−solid ratio but also by the number of leaching and the process pulping. This paper focuses on the influence of the parameters of the washing dechlorination process through experiments.

2.2. Fly Ash Washing Dechlorination Process. At present, the engineering application process of cement kiln co-disposal of MSWI fly ash in China is shown in Figure 1a. The fly ash washing dechlorination unit mainly adopts three-stage counter-current washing dechlorination; the schematic process flow is demonstrated in Figure 1b. The liquid−solid ratio (clear water/fly ash) of the washing dechlorination process is 3:1. Combined with the operation data of BBMG Liulihe, Jurong TCC, and CONCH Wuhu, the elution rate of chlorine in fly ash is required to be ≥95%.

The cyclic gradient washing dechlorination process for MSWI fly ash proposed in this paper is illustrated, and the operation process is presented in Figure 2. As shown in Figure

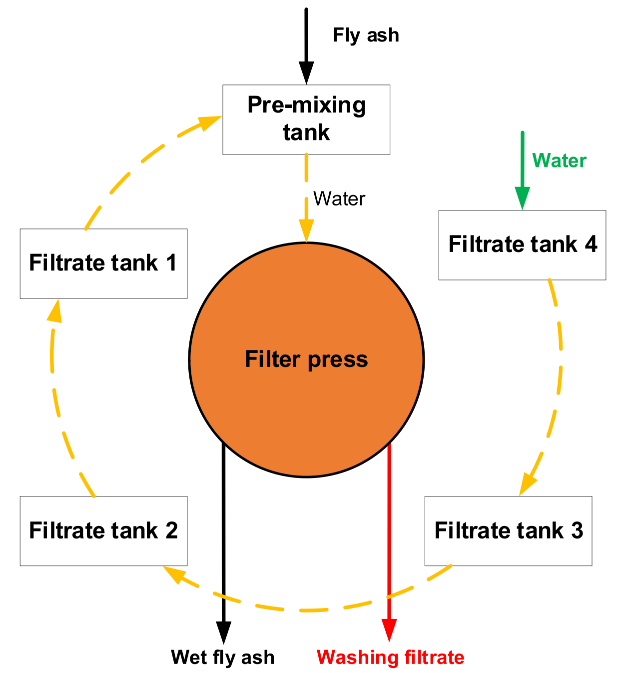

Figure 2. MSWI fly ash cyclic gradient water washing dechlorination process.

2, the first step is that the fly ash enters the pre-mixing tank to mix with the 5-stage washing filtrate for pulping and enters the filter press for separation, and the separated washing filtrate enters the subsequent disposal process. The second step is the washing filtrate in filtrate tank 4 to leach and separate the fly ash in the filter press, and the separated 5-stage washing filtrate enters the pre-mixing tank for mixing with the next batch of fly ash to make pulp. The third step is the washing filtrate in filtrate tank 3 to leach and separate the fly ash in the filter press, and the separated 4-stage washing filtrate enters filtrate tank 4. The fourth and fifth steps are the washing filtrate in filtrate tank 2 and filtrate tank 1, as in the third step of the process. The final step is to use clean water to leach and separate the fly ash in the filter press, the separated 1-stage washing filtrate into the filtrate tank 1, and the washing fly ash into the subsequent disposal process.

In practical industrial applications, according to the fly ash washing dechlorination requirement, the water washing gradient can be set reasonably by adjusting the number of filtrate tanks. And the fly ash will be drenched accordingly, which makes the washing filtrate form a concentration gradient. In conclusion, the cyclic gradient washing dechlorination process for MSWI fly ash only requires one-time process pulping and several leachings to achieve dechlorination requirements. Compared with the traditional three-stage counter-current washing dechlorination process, the process has the advantages of low investment, short process flow, less process equipment, small footprint, low operating energy consumption, low moisture content of washing fly ash, and complete separation of washing filtrate.

3. EXPERIMENTAL SECTION

3.1. Experimental Samples. The MSWI fly ash generated from Beijing's domestic waste incineration power plants in Gao'antun, Shunyi, Huairou, Tongzhou, and Dagong Village under stable operating conditions is sent to the BBMG (Beijing Building Materials Group) Liulihe Cement Plant for cement kiln co-processing. The samples required for the tests in this paper are taken by the systematic sampling method from the discharge port of the incoming fly ash transport tanker. All samples are not pretreated except for mixing and homogenization. The components of the samples are detected and analyzed. The average values of the admission fly ash composition tests for each month of 2021 are compared, and the results are presented in Figure 3. As can be seen from

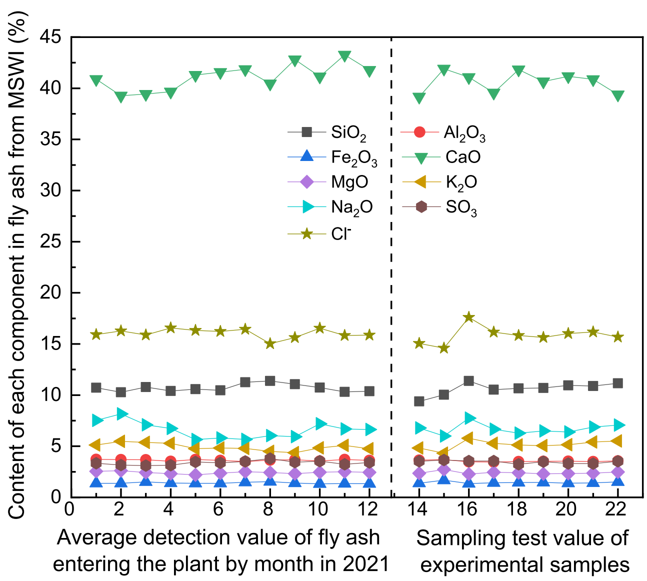

Figure 3. Comparative analysis of sample components for test.

Figure 3, the composition of the samples taken for the experiment and the composition of the incoming fly ash have basically remained the same, indicating that the samples used in this experiment have good representativeness.

3.2. Experimental Devices. The parameters of the main test equipment of the fly ash cyclic gradient washing dechlorination process are listed in Table 1. The filter press for leaching and solid−liquid separation of fly ash is indicated in Figure 4. The type, manufacturer, and origin of the devices are listed in Table 9 of the Appendix.

3.3. Design of the Experimental Program. Each test is conducted by weighing 80 kg of mixed and homogenized fly ash samples, which are put into a pre-mixing tank and stirred for 15 min to pulping. And after leaching, solid−liquid separation, and air-pressing in the filter press, the washing fly ash and washing filtrate are obtained. After the test, the samples are taken to test the moisture content and dry basis chlorine content (the same as below) of the washing fly ash as well as the Baume degree and turbidity parameters of the washing filtrate.

During the experiment, the liquid−solid ratios are 3:1 and 2:1, and the number of leachings are 3, 4, 5 and 6, respectively. The fly ash after the 4th and 5th leaching is stirred in the pre-mixing tank for primary process pulping and pumped into the

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Table 1. Main Equipment Configuration of Fly Ash Cyclic Gradient Washing Dechlorination Test

number	equipment	specifications	quantity	remarks
1	filter press	filter area: 2 m^2;volume: 70 L;power: 15 kW	1	affiliated hydraulic oil station and squeeze water station
2	pre-mixing tank	diameter: 1000 mm;height: 1200 mm	1	stirring power: 2.2 kW
3	washing filtrate tank	length: 1000 mm;width: 1000 mm;height:1000 mm	4	
4	feed pump	flow rate: 5 m^3/h;lift: 120 m;power: 7.5 kW	1	variable frequency regulation, screw type
5	washing filtrate pump	flow rate: 5 m^3/h;lift: 120 m;power: 7.5 kW	1	variable frequency regulation, centrifugal

Figure 4. Fly ash cycle gradient water washing dechlorination system filter press.

filter press again for solid−liquid separation and air-press separation to obtain washing fly ash and washing filtrate.

In the first test, water is placed in the pre-mixing tank and each filtrate tank, and fly ash samples are sent to the pre-mixing tank for pulping preparation and pumped into the filter press for filtration. Then, the washing filtrate enters the subsequent disposal process. From the filtrate tank 4, the fly ash is progressively leached. After each leaching, the Baume degree of the washing filtrate is detected, and the final washing fly ash enters the subsequent disposal process. When the above test is repeated three times, the Baume degree of each water washing filtrate tank tends to be stable, and the sample is carried out since the 4th test.

3.4. Analysis Methods. In this paper, the moisture content and chlorine content of fly ash and the turbidity and Baume degree of fly ash washing filtrate are tested. The main instruments are listed in Table 2.

Table 2. Cyclic Gradient Washing Dechlorination Detection Method

number	detection items (fly ash)	detection method	main instruments
1	moisture content	gravimetric method	rapid moisture meter (DHS-2DA)
2	chlorine content	ammonium thiocyanate volumetric method	circulating multipurpose vacuum pump (SHZ-D (III))
3	potassium and sodium content	flame photometry	flame photometer (FP6410)
4	sulfur trioxide content	barium sulfate gravimetric method	burette, beaker, bench scale, etc.
5	calcium, magnesium, aluminum, and iron content	EDTA titration method	burette, beaker, etc.
6	silicon content	potassium silicate volumetric method	burette, beaker, etc.
7	heavy metal content	inductively coupled plasma mass spectrometry	ICP−MS (7800)
8	turbidity of washing filtrate	spectrophotometry	multi-parameter controller [5B−3B (VII)]
9	Baume degree of washing filtrate	Baume meter	measurement range 0−35 °Bé

4. RESULTS AND DISCUSSION

4.1. Effect of Liquid−Solid Ratio on Dechlorination. The liquid−solid ratio directly affects the dissolution efficiency of soluble salts in fly ash. A lower liquid−solid ratio is not conducive to the dissolution of soluble salts in fly ash. It affects the dechlorination effect, while a higher liquid−solid ratio increases the subsequent fly ash washing filtrate disposal load. To investigate the influence of liquid−solid ratio on the effect of fly ash cyclic gradient washing dechlorination, the author combined the actual operation of MSWI fly ash cement kiln co-disposal lines such as BBMG Liulihe, Jurong TCC, and CONCH Wuhu. The test parameters are listed in Table 3. The two liquid−solid ratios are repeated 4 times. After the test, the washing fly ash is taken, and the chlorine content is detected after drying. The results are presented in Figure 5. It can be

Table 3. Test Parameters for the Effect of Liquid−Solid Ratio on Dechlorination

factors	test parameters	
liquid−solid ratio	2:1	3:1
test temperature (°C)	40	40
the number of leaching	4	4

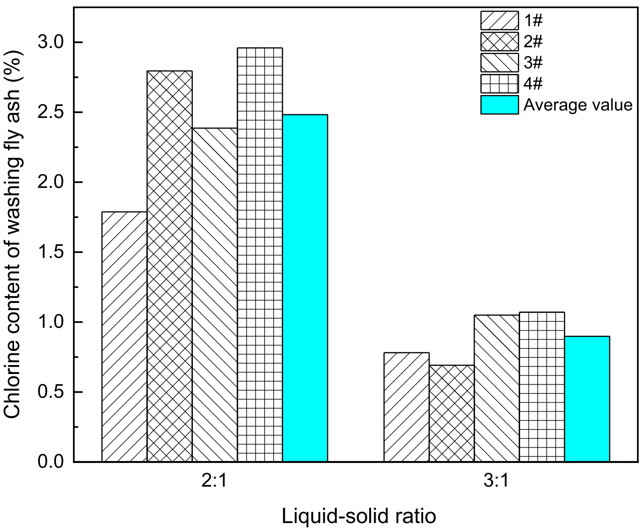

Figure 5. Influence of liquid−solid ratio on the effect of washing dechlorination.

seen from Figure 5 that when the liquid−solid ratio is 2:1, the chlorine content of the washing fly ash is 1.79−2.96% after 4th leaching. Under the same conditions with a liquid−solid ratio of 3:1, the chlorine content of washing fly ash is 0.78−1.07%. Compared with the liquid−solid ratio of 2:1, the average value of dechlorination effect is increased by 63.8%.

In Figure 3, the chlorine content in the MSWI fly ash is 15−20%. When the liquid−solid ratio of fly ash cyclic gradient washing dechlorination is 2:1, the average chlorine content of the washing fly ash is <3%. Thus, it can meet the requirements of China's "Standard for pollution control on the hazardous waste landfill pollution" (GB 18598-2019) on entering flexible landfills on the total amount of water-soluble salts <10%, which is suitable for landfill disposal after chelate curing.[38] When the liquid−solid ratio of fly ash cyclic gradient washing dechlorination is 3:1, the average chlorine content of the washing fly ash is <1%. Hence, it can meet the recommended requirement of chlorine content ≤1% of washing fly ash in China's "Technical specification for pollution control of fly ash from municipal solid waste incineration" (HJ1134-2020).[39] And the cement kiln co-processing is carried out through reasonable compatibility with cement production raw materials.

The experiments of the fly ash cyclic gradient washing dechlorination process prove that increasing the liquid−solid ratio can significantly improve the dechlorination effect. However, it also increases the operating load of the subsequent treatment of the washing filtrate. According to the author's statistics in the operation practice of fly ash disposal, for every 1 increase in the liquid−solid ratio, the power consumption per ton of fly ash increases by about 58 kW. The detailed statistics and calculations are presented in Table 10 of the Appendix. Therefore, the appropriate liquid−solid ratio should be selected in combination with the chosen disposal form of fly ash in industrial applications. In satisfying the requirements of fly ash dechlorination, the operating load of water washing filtrate treatment is reduced so that the fly ash disposal operates under optimal working conditions.

4.2. Effect of the Number of Leaching. Based on investigating the effect of liquid−solid ratio on fly ash cyclic gradient washing dechlorination, the impact of the number of leaching on the dechlorination effect of fly ash and washing filtrate Baume degree and turbidity are explored to find the optimal leaching number. The test parameters are listed in Table 4 and three replicate tests are conducted for each leaching number.

Table 4. Test Parameters for the Effect of the Number of Leaching

factors	test parameters			
liquid−solid ratio	3:1	3:1	3:1	3:1
test temperature (°C)	40	40	40	40
the number of leaching	3	4	5	6

4.2.1. Effect of the Number of Leaching on the Dechlorination. After the test, the chlorine content of washing fly ash was detected after drying, and the results are presented in Figure 6. As can be seen from Figure 6, with the increase of the number of leaching, the chlorine content of washing fly ash gradually decreases, but the reduction range of the chlorine content gradually narrows. After five leachings, the chlorine content of the washing fly ash stabilizes at 0.5−0.8%.

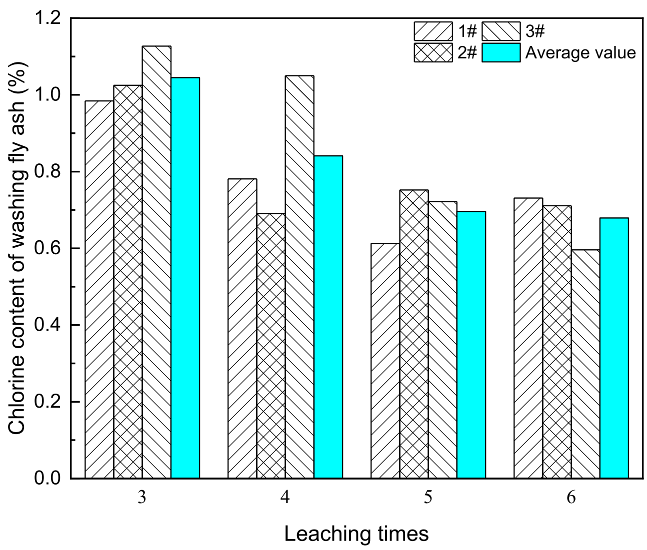

Figure 6. Effect of the number of leaching on the dechlorination.

At present, the actual operation of MSWI fly ash cement kiln co-disposal lines in China has different requirements on the chlorine content of washing fly ash into the kiln. BBMG Liulihe requires that the chlorine content of washing fly ash into the kiln be ≤0.6%. Most cement kiln co-disposal lines, such as Jurong TCC and CONCH Wuhu, require the chlorine content of washing fly ash into the kiln to be ≤1%. In the actual engineering application, according to the process conditions of fly ash disposal, while satisfying the requirements of fly ash dechlorination, the appropriate leaching number is selected to improve the system processing efficiency and make

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the fly ash disposal system operate under optimal working conditions.

4.2.2. Effect of the Number of Leaching on Baume Degree of Filtrate. The Baume degree of the washing filtrate was detected after the test, and the results are displayed in Figure 7.

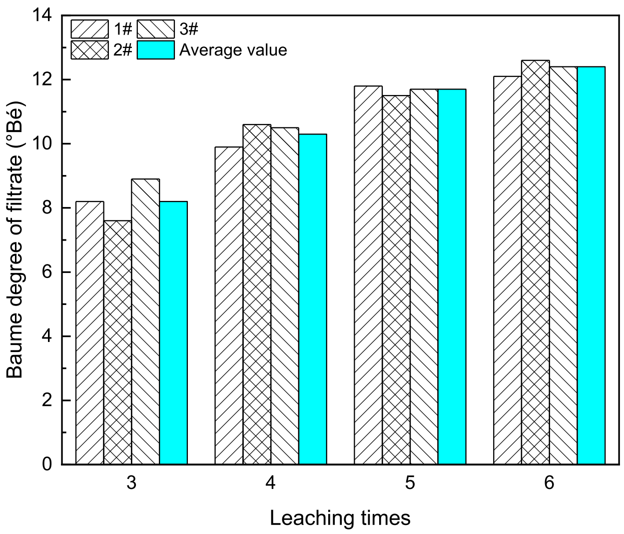

Figure 7. Effect of the number of leaching on the Baume degree of filtrate.

As shown in Figure 7, with the increase in the number of leaching, the Baume degree of the washing filtrate gradually increased, but the increasing amplitude gradually narrowed. After the 5th leaching, the Baume degree of washing filtrate was stabilized at 11−12 °Bé, which is confirmed by the stabilization of the chlorine content of the washing fly ash at the 5th leaching. The experiment proves that the ability to reduce the chlorine content of the washing fly ash and improve the Baume degree of the washing filtrate by simply increasing the number of leaching is limited.

Compared with the actual operation MSWI fly ash cement kiln co-disposal lines in China, the Baume degree of washing filtrate in BBMG Liulihe and Jurong TCC is about 10 °Bé. The Baume degree of washing filtrate in CONCH Wuhu and Yiyang is about 7−9 °Bé. The experiment demonstrates that the fly ash cyclic gradient washing dechlorination process can reduce the amount of washing filtrate treated by about 15% through reducing the water introduced by the production process chemicals. It also significantly reduces the operating load of the subsequent treatment of the washing filtrate, thus reducing the investment and operating costs of the fly ash disposal system.

4.2.3. Effect of the Number of Leaching on Filtrate Turbidity. In the operation practice of fly ash disposal, the turbidity of the washing filtrate in the evaporation salt unit is required to be ≤5NTU. After the test, the turbidity of the washing filtrate is tested, and the results are presented in Figure 8. In Figure 8, the average turbidity of the washing filtrate is <5NTU. Considering the error of on-site detection, the number of leaching has no effect on the turbidity of the washing filtrate, which can satisfy the turbidity requirement of the subsequent treatment of the washing filtrate.

The turbidity of the washing filtrate has a more significant impact on the subsequent washing filtrate treatment. A comparison of the washing filtrate of the fly ash disposal line of BBMG Liulihe is demonstrated in Figure 9. In Figure 9, a and c are the actual operating washing filtrate, and b and d are the test washing filtrate of the cyclic gradient washing dechlorination process. a and b are the states of still standing for 1 h, and c and d are the states without still standing. Figure 9 shows that the turbidity of the washing filtrate produced by the cyclic gradient washing dechlorination process is significantly better than that of the washing filtrate of the three-stage counter-current washing dechlorination process in actual operation. Currently, the actual operating cement kiln co-processing of MSWI fly ash in China must pass the process of still standing, sand filtration, and fine filtration to meet the requirement of turbidity <5NTU. The fly ash cyclic gradient washing dechlorination process can significantly reduce the operating load of the subsequent washing filtrate treatment process, shorten the process flow, and reduce equipment investment, operating costs, and floor space.

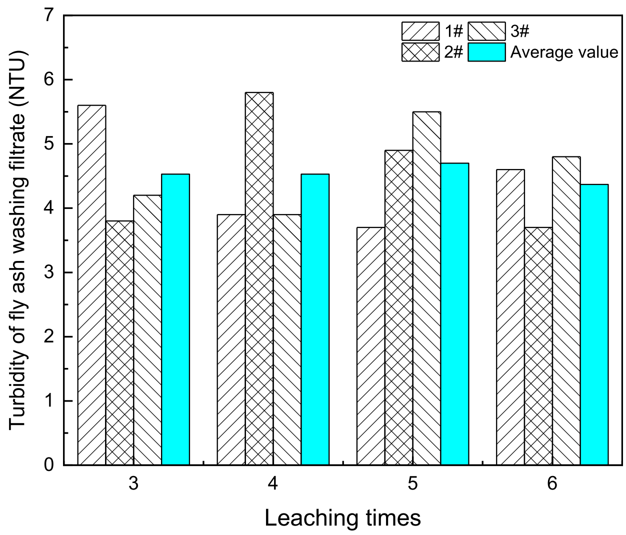

Figure 8. Turbidity of fly ash washing filtrate.

Figure 9. Comparison of the washing filtrate of two washing dechlorination processes. (a,c) Actual operating washing filtrate, and (b,d) test washing filtrate of the cyclic gradient washing dechlorination process. (a,b) States of still standing for 1 h; (c,d) states without still standing.

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4.3. Effect of Process Pulping. The above test shows that simply increasing the number of leaching cannot ensure that the chlorine content of washing fly ash is ≤0.6%. The main reason may be that the fly ash filter cake layer is partially penetrated in the leaching process, resulting in a short circuit, as shown in Figure 10. Hence, the surrounding fly ash leaching is insufficient. To solve the above problem, after the 4th leaching, the fly ash in the filter press is sent to the pre-mixing tank for mixed pulping with clean water and then pumped into the filter press for solid−liquid separation. The chlorine content is tested after drying the washing fly ash at the end of the test. And the result is compared with washing five times, as shown in Figure 11. After changing the 5th leaching to the primary stirring and pulping, the chlorine content of the washing fly ash is 0.5−0.6%, with a slight decrease and more stability. It indicates that the primary process pulping in the leaching process can effectively alleviate the incomplete leaching phenomenon caused by the partial breakdown of the fly ash filter cake layer.

Figure 10. Local breakdown and short circuit of the fly ash filter cake layer during the leaching process.

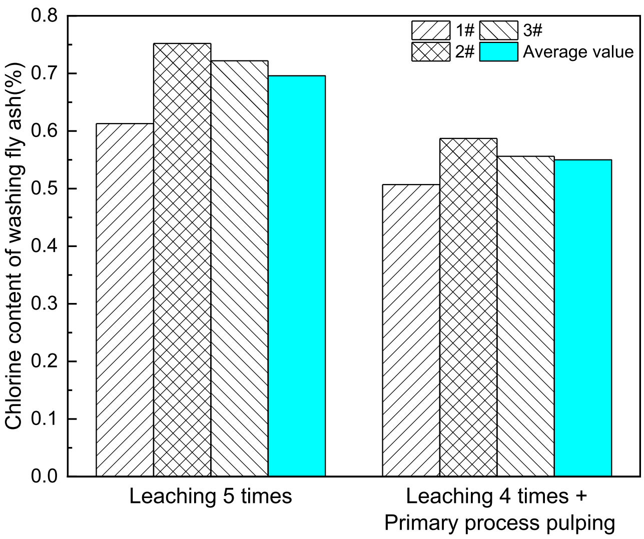

Figure 11. Effect of primary process pulping on the dechlorination.

In the actual operation of the MSWI fly ash cement kiln co-processing line, the three-stage counter-current washing dechlorination process requires the completion of pulping three times. Therefore, in the actual production process of applying the gradient washing dechlorination process, parameters such as the liquid−solid ratio and the number of leaching should be combined to comprehensively consider whether to carry out primary process pulping. Under the condition of meeting the requirement of fly ash dechlorination, the fly ash disposal system can operate under optimal conditions. In addition, since the filter press used in this experiment is the original model, the industrial application is an optimized model with the inlet port of the drenching tilted upward by 15° to reduce the possibility of a partial breakdown of the fly ash filter cake layer.

4.4. Effect of the Moisture Content of Washing Fly Ash. The chlorine content of washing fly ash is positively correlated with its moisture content, mainly because it contains soluble chlorine salts in the water. In addition, more energy is consumed in the subsequent drying process of washing fly ash. Therefore, reducing the moisture content of washing fly ash can reduce its chlorine content and the operational energy consumption of subsequent disposal of washing fly ash. Based on the above experiments to explore the effect of the liquid−solid ratio, a control test is added with the liquid−solid ratio of 3:1, and other conditions are the same.

Based on the above experiments to explore the effect of the liquid−solid ratio, the air-pressing process is added further to reduce the moisture content of washing fly ash. The main experimental parameters are listed in Table 5. The physical washing of fly ash after adding an air press is shown in Figure 12a, and the three-stage counter-current washing fly ash of the BBMG Liulihe fly ash disposal line is illustrated in Figure 12b. By comparing Figure 12a,b, it can be seen that the washing fly ash after adding the air press is in the shape of a cake, which has a lower moisture content and is easy to transport.

Table 5. Control Test Parameters before and after Adding Air Pressing

factor	test parameters	
liquid−solid ratio	3:1	3:1
test temperature (°C)	40	40
the number of leaching	4	4
separation method	mechanical filter pressing	mechanical filter pressing + air pressing
compressed air pressure (MPa)	0.2−0.38	0.2−0.38
pressing time (min)	10	10

The relationship between the variation of moisture content and chlorine content of the washing fly ash after passing through the air press and the separation method is presented in Figure 13. As can be seen from Figure 13, the moisture content of washing fly ash can be further reduced by air pressing. This is because the water contained in the washing fly ash is mainly free water, and more free water can be squeezed out of the washing fly ash by compressed air, thus reducing the moisture content of the washing fly ash and its chlorine content. In addition, due to the limited test conditions, the pressure of compressed air is low, and the compressed air of 0.8−1.0 MPa can be used for pressing in industrial applications, which can further reduce the moisture content of washing fly ash.

The drying process of washing fly ash requires a lot of energy, and its heat source is hot air from the cement kilns. According to the author's statistics on the operation of the MSWI fly ash cement kiln co-processing line in CONCH Wuhu, the drying costs of washing fly ash account for about 19% of the direct operating costs of fly ash. Meanwhile, the process also requires a closer distance between the washing fly ash dechlorination workshop and the cement kiln. The moisture content of the washing fly ash of the cyclic gradient washing dechlorination process is lower and easy to transport. This creates conditions for the subsequent study of directly transporting the washing fly ash into the cement kiln for co-

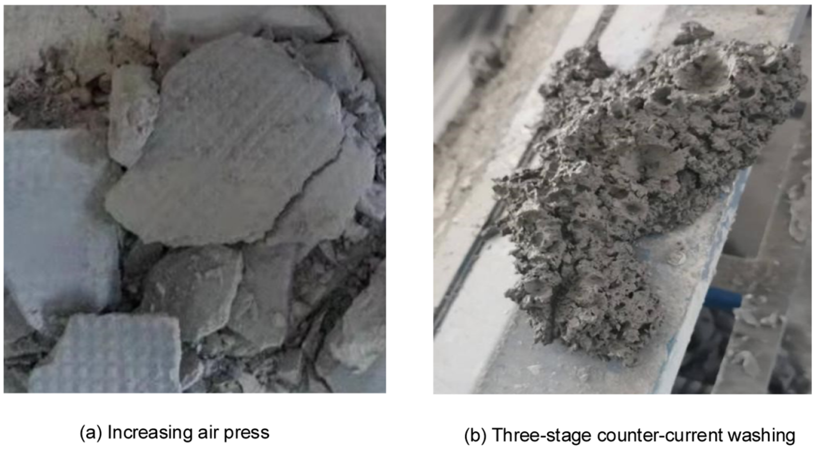

Figure 12. Physical washing of fly ash with different washing processes.

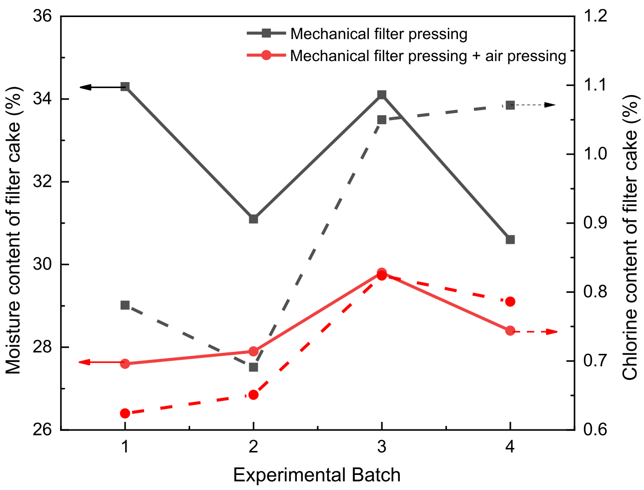

Figure 13. Moisture content of washing fly ash before and after increasing air pressing.

processing. It can reduce the operation energy consumption of fly ash disposal and the investment and floor space of drying equipment. On the other hand, it also can break the restrictions on the site selection of fly ash dechlorination workshops by cement kilns and create conditions for establishing a centralized washing dechlorination pretreatment center for fly ash.

4.5. Effect of Washing Filtrate Temperature. The fly ash cyclic gradient washing dechlorination experiment is greatly affected by the ambient temperature. When the ambient temperature decreases, the temperature of the fly ash washing filtrate is lower, and the leaching rate is significantly reduced. The statistical results are listed in Table 6. Considering that the filter cloth is not replaced in the whole process of the test, the water permeability of the filter cloth is tested. Comparing its water permeability with that of the unused filter cloth shows that the water permeability of the filter cloth is not significantly attenuated, and the results are presented in Table 7. It can be seen from Table 7 that the water permeability of the filter cloth after multiple recycling has not been significantly attenuated. The reason for the significant decrease in the drenching speed is speculated to be the lower ambient temperature, which leads to a rapid heat loss in the system, resulting in a reduction of the temperature of the washing filtrate and a slow separation of liquid and solid in the fly ash leaching process. As a result, the time of the leaching process is significantly increased, and the operating efficiency of the fly ash disposal system is reduced.

Table 7. Test Results of Water Permeability of Filter Cloth

number	filter cloth	filtration capacity (L)	time-consuming (min)	filter cloth material
1	in use	200	4	polypropylene
2	unused	200	3.8	polypropylene

During the test, an electric heater is used to heat the washing filtrate to 50 °C to balance the effect of the reduced ambient temperature on the drenching rate. Due to the small size of the experimental devices, the temperature of the washing filtrate in actual operation is still significantly reduced, which is not ideal for improving the leaching rate. The decrease in ambient temperature reduces the temperature of the fly ash washing filtrate, resulting in a reduction of the leaching rate. Combined with the operation of the fly ash co-disposal project, due to the large material handling capacity and the small percentage of heat loss in the system, the liquid phase temperature is maintained at about 40 °C all year round. Therefore, in industrial applications, the fly ash cyclic gradient washing dechlorination process can be used to balance the effect of environmental temperature variations on the drenching rate by adding preheating and heat preservation measures.

5. ANALYSIS OF ENGINEERING APPLICATIONS

Based on the actual operation of the MSWI fly ash cement kiln co-processing line in BBMG Liulihe, Jurong TCC, and CONCH Wuhu, the authors apply the fly ash cyclic gradient washing process to an actual MSWI fly ash cement kiln co-processing project in Yunnan. The main control requirements

Table 6. Statistics of Leaching Time of Fly Ash Cyclic Gradient Washing Dechlorination

number	test month	ambient temperature (°C)	inlet water temperature (°C)	washing filtrate temperature (°C)	leaching time (min)
1	September	32	40	35	15−20
2	November	8	40	15	40−50

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are that the liquid−solid ratio is not more than 3:1 and the chlorine content of washing fly ash is ≤1%. The design schemes of traditional three-stage counter-current and cyclic gradient washing dechlorination processes are compared and analyzed, in which the latter has 3 leachings, primary process pulping, and 0.8 MPa air-press pressure. The main design results are listed in Table 8. As can be seen from Table 8,

Table 8. Comparison of Design Schemes for Fly Ash Cement Kiln Collaborative Project in Yunnan

number	sub-item	unit	three-stage counter-current washing dichlorination	cyclic gradient washing dechlorination
1	disposal capacity	t/d	100	100
2	installed power	kW	3560	2480
3	floor space	m^2	1680	1127
4	treatment capacity of washing filtrate	m3/d	360	320
5	total water volume of drying	t/d	45.6	32.9

under the same disposal capacity, the cyclic gradient washing dechlorination process can reduce the installed power by 30.3%, save 32.9% of the floor space, reduce the washing filtrate treatment capacity by 11.1%, and reduce the drying load by 27.9%.

Compared with the traditional three-stage counter-current washing dechlorination process, the cyclic gradient washing dechlorination process proposed in this paper has three main advantages. First, the washing dechlorination unit of the process is short and has less process equipment. Hence, the installed power and floor space are less. Second, the turbidity of the washing filtrate of the process can meet the process requirements for subsequent disposal. And there is no need for static sedimentation, sand filtration, fine filtration, and so forth. Additionally, since flocculants are not required, the total amount of washing filtrate treatment is reduced, which helps to reduce further the process flow, installed power, and floor space of subsequent washing filtrate treatment units. Third, the moisture content of the washing fly ash is lower in the cyclic gradient washing dechlorination process, thus reducing the drying load of the washing fly ash.

6. CONCLUSIONS

By exploring the optimization of the washing dechlorination process for MSWI fly ash, this paper establishes the test devices for the fly ash cyclic gradient washing dechlorination process and investigates the effects of process parameters such as liquid−solid ratio, the number of leaching, pulping process, and air pressing on the chlorine content and filtrate in washing fly ash. The conclusions are as follows.

The liquid−solid ratio, the number of leaching, and the pulping process greatly influence the dechlorination of fly ash. Under the conditions of liquid−solid ratio of 3:1, the number of leaching of 4, and the primary process pulping, the chlorine content of washing fly ash is stabilized between 0.5 and 0.6%, which has a good dechlorination effect.

By increasing the number of leaching, the fly ash cyclic gradient washing dechlorination process can increase the washing filtrate Baume degree to between 11 and 12 °Bé and meet the average turbidity value of ≤5NTU. Compared with the traditional fly ash washing dechlorination process, it shortens the process flow and reduces the amount of washing filtrate by about 15%. In addition, the combination of mechanical filter press and air press can reduce the moisture content of washing fly ash to 28−30%, which creates conditions for subsequent research on the direct transportation of washing fly ash to cement kilns for co-disposal.

APPENDIX

The type, manufacturer, and origin of the devices are listed in Table 9.

In the actual industrial production process, the chlorine content in fly ash is fluctuating. To ensure the stability of chlorine content in washing fly ash, the solid-liquid ratio should be properly adjusted according to the chlorine content in fly ash. It will impact the power consumption per ton of fly ash disposal. The fly ash disposal line of CONCH Wuhu is taken as an example, processing about 140 tons of fly ash per day. The actual operation data statistics are shown in Table 10.

Table 10. Statistical Table of Disposal Power Consumption per Ton of Fly Ash

sub-item	operating condition 1	operating condition 2	operating condition 3
liquid−solid ratio	2.8	3	3.4
the power consumption per ton of fly ash (kW)	314	327	349
applicability remarks	fly ash with chlorine content <18%	fly ash with chlorine content before both	fly ash with chlorine content >22%

As per Table 10, the liquid−solid ratio increases by 0.6, and the power consumption per ton of fly ash disposal increases by about 35 kW. Thus, it can be deduced that an increase of 1 in the liquid−solid ratio increases the power consumption of disposing of per ton of fly ash by about 58 kW.

Table 9. Type, Manufacturer, and Origin of the Devices

number	equipment	brand	manufacturer	origin
1	filter press	Tongxing	Yantai Tongxing Industrial Group Co., Ltd	Yantai, Shandong Province, China
2	pre-mixing tank	Tongxing	Yantai Tongxing Industrial Group Co., Ltd	Yantai, Shandong Province, China
3	washing filtrate tank	Zhengfeng	Anhui Zhengfeng New Material Technology Co., Ltd	Lu'an, Anhui Province, China
4	feed pump	Gaofeng	Sichuan Zigong Pump Industry Co., Ltd	Zigong, Sichuan Province, China
5	washing filtrate pump	Gaofeng	Sichuan Zigong Pump Industry Co., Ltd	Zigong, Sichuan Province, China

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Notes

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REFERENCES

(1) Xiao, S. J.; Dong, H. J.; Geng, Y.; Francisco, M. J.; Pan, H. Y.; Wu, F. An overview of the municipal solid waste management modes and innovations in Shanghai, China. *Environ. Sci. Pollut. Res.* **2020**, *27*, 29943−29953.

(2) Scarlat, N.; Fahl, F.; Dallemand, J. F. Status and Opportunities for Energy Recovery from Municipal Solid Waste in Europe. *Waste Biomass Valorization* **2019**, *10*, 2425−2444.

(3) Chen, Y. C. Effects of urbanization on municipal solid waste composition. *Waste Manag.* **2018**, *79*, 828−836.

(4) Nanda, S.; Berruti, F. Municipal solid waste management and landfilling technologies: a review. *Environ. Chem. Lett.* **2021**, *19*, 1433−1456.

(5) Compiled by National Bureau of Statistics of China. *China Statistical Yearbook, Section 8: Resources and Environment*; China Statistical Press: Beijing, 2017; p 20.

(6) Compiled by National Bureau of Statistics of China. *China Statistical Yearbook, Section 8: Resources and Environment*; China Statistical Press: Beijing, 2021; p 18.

(7) Ministry of housing and urban rural development, National Development and Reform Commission. *The 14th Five-Year Plan for the Development of Urban Household Waste Sorting and Treatment Facilities*, 2021.

(8) Fan, C. C.; Wang, B. M.; Ai, H. M.; Liu, Z. A comparative study on characteristics and leaching toxicity of fluidized bed and grate furnace MSWI fly ash. *J. Environ. Manage.* **2022**, *305*, 114345.

(9) Azam, M.; Setoodeh Jahromy, S. S.; Raza, W.; Wesenauer, F.; Schwendtner, K.; Winter, F. Comparison of the Characteristics of Fly Ash Generated from Bio and Municipal Waste: Fluidized Bed Incinerators. *Materials* **2019**, *12*, 2664.

(10) Wang, P.; Hu, Y. N.; Cheng, H. F. Municipal solid waste (MSW) incineration fly ash as an important source of heavy metal pollution in China. *Environ. Pollut.* **2019**, *252*, 461−475.

(11) Shunda Lin, S. D.; Jiang, X. G.; Zhao, Y. M.; Yan, J. H. Disposal technology and new progress for dioxins and heavy metals in fly ash from municipal solid waste incineration: A critical review. *Environ. Pollut.* **2022**, *311*, 119878.

(12) Ministry of Ecological Environment of China. *Solid Waste Center*; National Directory of Hazardous Wastes, 2020.

(13) Ferraro, A.; Farina, I.; Race, M.; Colangelo, F.; Cioffi, R.; Fabbricino, M. Pre-treatments of MSWI fly-ashes: A comprehensive review to determine optimal conditions for their reuse and/or environmentally sustainable disposal. *Rev. Environ. Sci. Bio/Technol.* **2019**, *18*, 453−471.

(14) Zhang, Y. Y.; Wang, L.; Chen, L.; Ma, B.; Zhang, Y. K.; Ni, W.; Tsang, D. C. Treatment of municipal solid waste incineration fly ash: State-of-the-art technologies and future perspectives. *J. Hazard. Mater.* **2021**, *411*, 125132.

(15) Xue, Y.; Liu, X. M. Detoxification, solidification and recycling of municipal solid waste incineration fly ash: A review. *Chem. Eng. J.* **2021**, *420*, 130349.

(16) Shao, N. N.; Wei, X. K.; Monasterio, M.; Dong, Z. J.; Zhang, Z. T. Performance and mechanism of mold-pressing alkali-activated material from MSWI fly ash for its heavy metals solidification. *Waste Manag.* **2021**, *126*, 747−753.

(17) Yu, J.; Sun, L. S.; Xiang, J.; Jin, L. M.; Hu, S.; Su, J. R.; Qiu, J. Physical and chemical characterization of ashes from a municipal solid waste incinerator in China. *Waste Manage. Res.* **2013**, *31*, 663−673.

(18) Baidya, R.; Ghosh, S. K.; Parlikar, U. V. Co-processing of Industrial Waste in Cement Kiln - A Robust System for Material and Energy Recovery. *Procedia Environ. Sci.* **2016**, *31*, 309−317.

(19) Wang, X. T.; Jin, B. S.; Xu, B.; Lan, W. J.; Qu, C. R. Melting characteristics during the vitrification of MSW incinerator fly ash by swirling melting treatment. *J. Mater. Cycles Waste Manage.* **2017**, *19*, 483−495.

(20) Ma, W. C.; Shi, W. B.; Shi, Y. J.; Chen, D. M.; Liu, B.; Chu, C.; Li, D.; Li, Y. L.; Chen, G. Y. Plasma vitrification and heavy metals solidification of MSW and sewage sludge incineration fly ash. *J. Hazard. Mater.* **2021**, *408*, 124809.

(21) Gao, N. B.; Li, J. Q.; Quan, C.; Tan, H. Z. Product property and environmental risk assessment of heavy metals during pyrolysis of oily sludge with fly ash additive. *Fuel* **2020**, *266*, 117090.

(22) Zhang, G. L.; Li, Z. F.; Han, L.; Zhu, Y. C.; Gu, J.; Wei, B. Y. Engineering application of cement kiln co-disposal of waste incineration fly ash technology. *Cement* **2017**, *S1*, 8−12.

(23) Boğa, A. R.; Topcu, i. B. Influence of fly ash on corrosion resistance and chloride ion permeability of concrete. *Constr. Build. Mater.* **2012**, *31*, 258−264.

(24) Joseph, A. M.; Snellings, R.; Van den Heede, P.; Matthys, S.; De Belie, N. D. The Use of Municipal Solid Waste Incineration Ash in Various Building Materials: A Belgian Point of View. *Materials* **2018**, *11*, 141.

(25) Chen, W. S.; Chang, F. C.; Shen, Y. H.; Tsai, M. S.; Ko, C. H. Removal of chloride from MSWI fly ash. *J. Hazard. Mater.* **2012**, *237−238*, 116−120.

(26) Yan, M.; Jiang, J. H.; Zheng, R. D.; Yu, C. M.; Zhou, Z. H.; Hantoko, D. Experimental study on the washing characteristics of fly ash from municipal solid waste incineration. *Waste Manage. Res.* **2022**, *40*, 1212−1219.

(27) Wei, Y. M.; Liu, S. J.; Yao, R. X.; Chen, S.; Gao, J. M.; Shimaoka, T. Removal of harmful components from MSWI fly ash as a pretreatment approach to enhance waste recycling. *Waste Manag.* **2022**, *150*, 110−121.

(28) Huang, H. G.; Liu, W.; Zhang, L.; Fang, J. M.; Xu, F.; Bu, S.; Xu, W. G.; Xu, C.; Yao, H. Q.; Ma, Z. L. A microscopic and quantitative analysis on the separation of chloride ion by fly ash washing: Effect of liquid-to-solid ratio, washing time and temperature. *Environ. Sci. Pollut. Res.* **2022**, *29*, 36208−36215.

(29) Zhao, K. X.; Hu, Y. Y.; Tian, Y. Y.; Chen, D. Z.; Feng, Y. H. Chlorine removal from MSWI fly ash by thermal treatment: Effects of iron/aluminum additives. *J. Environ. Sci.* **2020**, *88*, 112−121.

(30) Wang, Y.; Liu, Y.; Deng, H. N. Optimization for dechlorination washing of fly ash from municipal solid waste incineration by response surface method. *Environ Pollut Prev* **2020**, *42*, 1259−1262.

(31) Wang, Y. T.; Tang, M. H.; Zong, D.; Chen, Z. L.; Lin, X. Q.; Lu, S. Y. Two-stage countercurrent water-washing characteristics of municipal solid waste incineration fly ash from grate furnace. *J. Zhejiang Univ. Eng. Sci.* **2019**, *53*, 981−987.
(32) Yue, Y.; Zhang, J.; Sun, F. C.; Wu, S. M.; Pan, Y.; Zhou, J. Z.; Qian, G. R. Heavy metal leaching and distribution in glass products from the co-melting treatment of electroplating sludge and MSWI fly ash. *J. Environ. Manage.* **2019**, *232*, 226−235.
(33) Wang, Y. X.; Shao, L. Y.; Xu, T. N.; Cai, C. H.; Yuan, J. S.; Zhang, Y. W.; Qian, G. R. Study on existing speciation and deep dechlorination of chlorine in waste incineration fly ash. *Inorg. Chem.: Indian J.* **2021**, *53*, 78−83.
(34) Partanen, J.; Backman, P.; Backman, R.; Hupa, M. Absorption of HCl by limestone in hot flue gases. Part II: importance of calcium hydroxychloride. *Fuel* **2005**, *84*, 1674−1684.
(35) Bai, J. J.; Zhang, Z. Q.; Yan, D. H.; Li, L. Study on the removal of chlorine and heavy metals in incineration fly ash during water washing process. *Environ. Eng.* **2012**, *30*, 104−108.
(36) Zhu, F. F.; Takaoka, M.; Shiota, K.; Oshita, K.; Kitajima, Y. Chloride chemical form in various types of fly ash. *Environ. Sci. Technol.* **2008**, *42*, 3932−3937.
(37) Chang, W.; Liu, H. H.; Jiang, X. G. Study on dechlorination and heavy metal leaching characteristics of MSWI fly ash during water washing. *Inorg. Chem.: Indian J.* **2022**, *54*, 113−118.
(38) Ministry of Ecological Environment of China; State Adminintration for Market Regulation. *Standard for Pollution Control on the Hazardous Waste Landfill Pollution*, 2021.
(39) Ministry of Ecological Environment of China. *Technical Specification for Pollution Control of Fly Ash from Municipal Solid Waste Incineration*, 2021.

https://doi.org/10.1021/acsomega.2c07032
ACS Omega 2023, 8, 4081−4091