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Dechlorination of fly ash through co-leaching with organic waste: effect of inoculum

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In order to improve the dechlorination amount of incineration fly ash and increase the mixture amount of fly ash into the kiln for cement kiln co-disposal, this study investigated the dechlorination effect of fly ash using co-leaching method with organic waste. Results revealed that the dechlorination effect of the experimental groups (co-leaching of organic waste and fly ash) was better than that of the control group (separate leaching of fly ash). The chlorine content in fly ash residue of the lactic acid bacteria group (organic waste + fly ash + 10% *Enterococcus faecalis*) was the lowest, at 0.39%. After dynamic leaching, the allowable incorporation rate of fly ash residue into the kiln was 10.30%, which was 9.6 percentage points higher than that achieved by static leaching (three-step water washing). In addition, to achieve the same dechlorination amount, the co-leaching of organic waste and fly ash could save at least 48.7% of water consumption compared to the separate leaching of fly ash. Therefore, the co-disposal of organic waste and incineration fly ash is an economical and efficient method for fly ash dechlorination.

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1. Introduction

In recent years, incineration technology has become one of the mainstream technologies for municipal solid waste treatment and disposal due to its many advantages, such as reduction, stabilization and resource utilization.^{1,2} However, the incineration fly ash produced by incineration has been classified as hazardous waste (code HW18, 772-002-18) according to the China National Hazardous Waste List (2025 Edition), due to its high content of chlorine, heavy metals and dioxins, *etc*³⁻⁵, and its treatment and disposal require emphasis. Many studies have shown that the main compositions of fly ash are similar to the raw materials of cement industry.^{3,6,7} Therefore, using fly ash to replace part of the cement raw materials for co-disposal in the cement kiln has become one of the main technologies for fly ash resource utilization.⁸⁻¹⁰

However, the high chlorine content is the main factor restricting the entry of fly ash into the cement kiln for co-disposal.^{11,12} Therefore, achieving effective chlorine removal

from fly ash is a key issue for its application in cement production. Conventional water-washing could remove most of the chlorine from fly ash,^{9,13-15} but there still remained 1–4% water-insoluble chlorine in the fly ash after water washing, which limited the proportion of fly ash that could be incorporated into cement kilns. Acid washing or salt washing could achieve deep dichlorination.¹⁶⁻²⁰ However, those methods required a large amount of acid or inorganic salts, leading to high chemical costs and potential secondary pollution. In contrast, using organic acids derived from the fermentation of organic waste offers a significant economic advantage. Organic waste is an abundant and low-cost resource. Its fermentation produces a mixed organic acid broth *in situ*,^{21,22} eliminating the need for purchasing and transporting large quantities of industrial chemicals, thereby substantially reducing the overall operational cost of the fly ash treatment process. Moreover, preliminary experimental results from our team shown that when using lactic acid fermentation solution to wash fly ash, the static leaching method of two-step acid addition (*i.e.*, in a three-step washing process, the first step involved water washing, while the second and third steps utilized lactic acid fermentation solution) had the best dechlorination effect. This method could remove 98.7% of total chlorine and 95.6% of water-insoluble chlorine from fly ash, and increased the yield of fly ash into the kiln by 6 percentage points compared with three-step pure water washing.²¹ Furthermore, the dechlorination capability of other organic acids (*e.g.*, acetic acid, butyric acid, propionic acid, *etc.*) was similar to that of lactic acid.²³ Therefore, different kinds of organic acid fermentation broths

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produced by organic waste are expected to achieve deep chlorine removal from fly ash.

Organic waste is an important part of municipal solid waste. With the rapid development of the economy, the production of organic waste is increasing. It is well known that organic waste contains a large amount of easily degradable organic matter, N, P, K, Ca, and various trace elements, which gives it high recycling value.²⁴ Studies have found that organic waste contains abundant indigenous microbial flora; that is, in a natural stacking state, the diverse microorganisms present in organic waste can, through hydrolytic acidification reactions, lead to the production of large amounts of organic acid mixtures (*e.g.*, lactic acid, acetic acid, formic acid).²⁵ Our team has previously demonstrated that the lactic acid fermentation solution derived from organic waste can achieve deep dechlorination of fly ash, but the fermentation cost increases due to the inoculation of lactic acid bacteria.^{21,23} Therefore, it is necessary to investigate whether the natural hydrolytic acidification solution of organic waste has similar dechlorination effect and whether synergistic effects exist among various organic acids in the hydrolytic acidification solution.

In summary, building upon previous static leaching experiments, this study introduced a novel dynamic co-leaching strategy that integrated the treatment of two solid waste streams: organic waste and incineration fly ash. The key innovation lay in utilizing the *in situ* produced organic acids from the organic waste's hydrolytic acidification to enhance the dechlorination of fly ash within a single system. This study investigated the effect of inoculum (*Enterococcus faecalis*) on this process, aiming to evaluate the dechlorination performance and the potential for enhancing fly ash incorporation in cement kilns under dynamic leaching conditions. The results are expected to provide a scientific foundation for developing a cost-effective and synergistic co-disposal technology for organic waste and incineration fly ash.

2. Materials and methods

2.1 Experimental materials

The fly ash samples used in this study originated from a municipal solid waste incineration plant in Beijing. The incinerator of the plant was a mechanical grate furnace. In the process of flue gas purification, $\text{Ca}(\text{OH})_2$ was used to neutralize the acid gas generated during incineration. The collected fly ash was dried at 105 °C to constant weight and then stored separately for later use. Co-leaching experiments were carried out in a series of 5 cm diameter plexiglass columns (Fig. 1). Organic waste (dry weight) and fly ash were added to the plexiglass column in two layers with a mass ratio of 1 : 1, separated by a filter screen. Different inoculation conditions were shown in Table 1. The bottom of the column was lined with gravel and fine sand to prevent the loss of fly ash. During the experiment, the dripping port was opened regularly to introduce 15 mL of ultrapure water into the column, and a conical bottle at the bottom of the column was used to collect the leaching solution. The entire experiment lasted 64 days. During the 64 day experiment, 15 mL of ultrapure water was introduced into each column at the following time points: days 1, 2, 3, 4, 5, 6, 7, 9, 11,

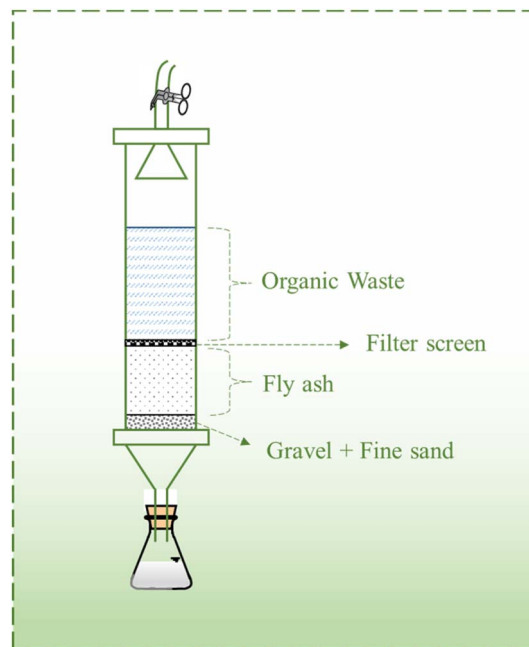


Fig. 1 Experimental setup schematic diagram.

15, 19, 21, 23, 25, 27, 30, 32, 38, 40, 43, 47, and 64. The frequent additions in the first week (daily) were designed to rapidly leach water-soluble chlorine, while the gradually extended intervals thereafter allowed for microbial acid production and simulated the intermittent infiltration typical of landfill environments. The pH value, organic acid content and chloride ion content of the leaching solution collected were measured. At the end of the experiment, the leaching column was opened, and the fly ash was collected and dried, after which the residual mass and chlorine content of the fly ash residue were determined. All experiments were conducted in triplicate using three identical leaching columns for each treatment.

2.3 Analysis method

The content of lactic acid in leachate was determined by high performance liquid chromatography (LC-20A, Shimadzu, Kyoto, Japan). The contents of acetic acid, propionic acid, butyric acid, caproic acid and valeric acid in leachate were determined by gas chromatography (CP3800, Shimadzu, Kyoto, Japan). Chlorine content in leachate was determined by ion chromatograph (ICS 600, Thermo Fisher Scientific (Shanghai) Instrument Co., Ltd). The mineral facies characteristics of fly ash before and after leaching were analyzed by X-ray diffraction (LabX XRD-6000, Shimadzu International Trade Shanghai Co., Ltd).

3. Results and discussions

3.1 Effect of inoculum on the dechlorination amount of fly ash

As can be seen from Fig. 2, the chlorine removal from fly ash in each group showed a similar trend. During the initial stage of leaching (the first 10 days), the chlorine in fly ash was quickly



Table 1 Experimental scheme of co-leaching of organic waste and fly ash

Experimental group	Leaching method	Single leaching amount
Inoculation of lactic acid bacteria group (LAB)	150 g organic waste (wet weight) + 30 g fly ash + 10% <i>Enterococcus faecalis</i>	15 mL
Non-inoculation group (NI)	150 g organic waste (wet weight) + 30 g fly ash	15 mL
Control group	30 g fly ash	15 mL

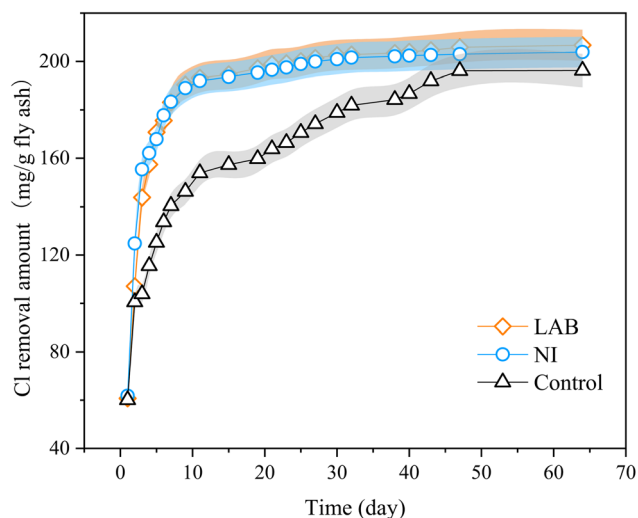


Fig. 2 Variation of dechlorination amount with leaching time.

dissolved. Subsequently, the removal rate of chlorine from fly ash gradually slowed down and stabilized (days 10–64). The main reason for this pattern is that during the early stage of leaching, there was a large amount of water-soluble chlorine in the fly ash, and this portion of chlorine could be quickly dissolved in the leaching solution and removed from the leaching system by the flow of leaching solution, thus showing a rapid removal rate at the early stage. In addition, for the LAB and NI groups, the organic wastes in the system produced organic acids (*e.g.*, lactic acid, acetic acid, propionic acid, *etc.*) under the action of microorganisms during the whole leaching process, and the presence of these organic acids could also promote the rapid dissolution of water-soluble chlorine in fly ash. With the removal of water-soluble chlorine from fly ash, the water-insoluble chlorine in fly ash became the main form of chlorine in the leaching system, which resulted in the observed gradual slowdown of chlorine removal rate at the late leaching stage. At the end of leaching, the dechlorination amounts of LAB, NI and control groups were 206.7 mg g⁻¹, 203.9 mg g⁻¹ and 196.2 mg g⁻¹, respectively. Compared with the control group, an additional 10.5 mg g⁻¹ and 7.7 mg g⁻¹ of chlorine was removed from the LAB and NI groups, respectively. This additional chlorine removal primarily originated from the water-insoluble chlorine in the fly ash.

3.2 Effect of inoculum on the properties of hydrolytic acidification broth

Our previous static leaching experiments have shown that the lactic acid fermentation broth from organic wastes has a good

effect on the chlorine removal from fly ash.²¹ On this basis, this study further investigated the dechlorination effect of lactic acid fermentation broth under dynamic leaching. Organic waste can also undergo hydrolytic acidification by environmental microorganisms when it is naturally stacked, leading to the production of large amounts of organic acids, among which lactic acid is typically the most abundant.²⁶ Therefore, this study investigated the co-leaching of organic waste with fly ash both with and without the addition of lactic acid bacteria (*i.e.*, natural hydrolysis and acidification), and compared the effect of lactic acid fermentation broth to that of hydrolysis acidification broth from organic waste on chlorine removal from fly ash during dynamic leaching. The experimental results are shown in Fig. 3. From Fig. 3, the daily yield of organic acids in the leaching systems of LAB and NI groups both increased first and then decreased with increasing leaching time, which was mainly due to the decrease in bioavailable organic matter within the organic waste at the later stage of leaching. During the whole leaching cycle, the maximum concentration of lactic acid in the LAB group was 24.5 g L⁻¹, which was higher than that in the NI group (14.2 g L⁻¹). This was mainly attributed to the addition of *Enterococcus faecalis*, which directed the organic waste towards lactic acid production. In addition, Fig. 3 also shows that the cumulative production of total organic acid and of lactic acid in the LAB group were 4054.1 mg and 3582.4 mg, respectively, which were 1197.3 mg and 2332.3 mg higher than those in the NI group. Thus, the addition of *Enterococcus faecalis* enhanced the substrate utilization efficiency of organic waste and increased lactic acid production, which is favorable for chlorine removal from fly ash. However, although organic acids are the primary drivers of chlorine removal, other components in the organic waste (*e.g.*, ammonium, phosphate, sulfide, and metal ions) may also influence the dechlorination efficiency. For instance, ammonium can act as a complexing agent, while phosphate may precipitate with calcium, potentially affecting the solubility of chlorine-bearing phases. Nevertheless, in the present study, the enhanced chlorine removal is primarily attributed to the chelating and acidifying effects of the organic acids produced during hydrolysis and acidification; the contributions of other components are considered secondary and warrant further investigation.

Furthermore, a Pearson correlation analysis revealed a positive correlation between the chlorine removal amount from fly ash and the production of organic acids in the leachate (Fig. 4). However, the correlation between the dechlorination amount and the production of propionic acid was stronger, while the correlation with other organic acids such as acetic acid and with the total production of organic acids was weaker. Therefore,



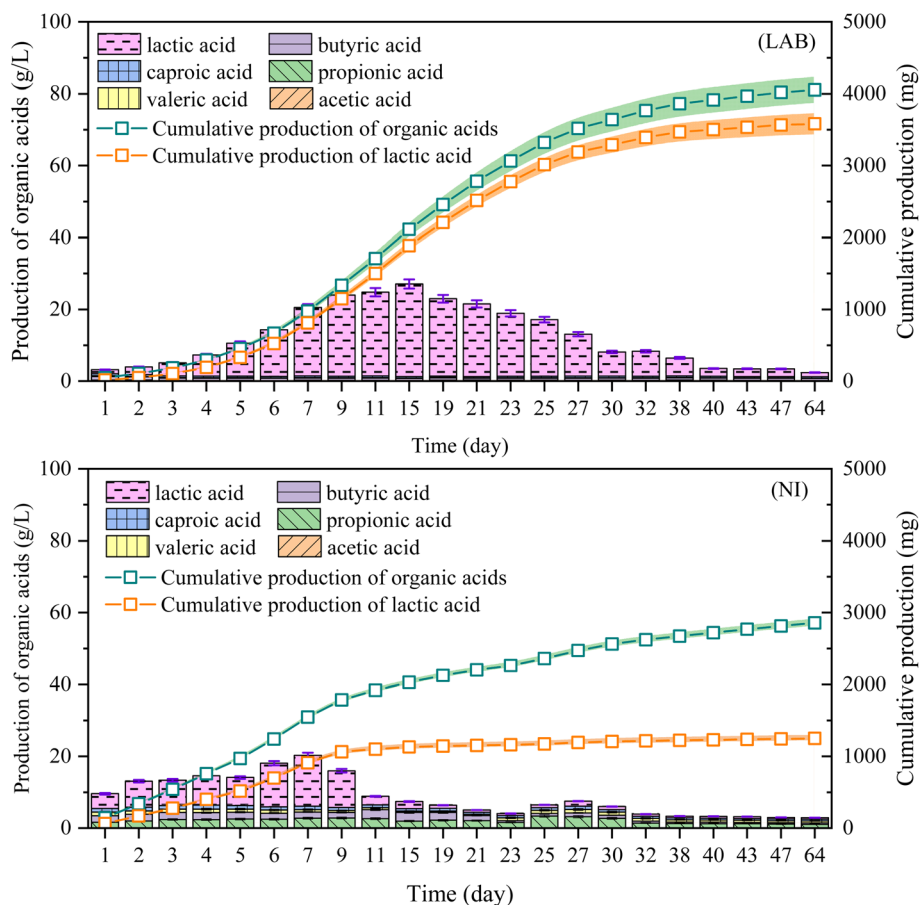


Fig. 3 Organic acid production profile over time.

when organic waste is co-processed with incineration fly ash, targeted measures to increase the production of propionic acid from the organic waste could enhance the dechlorination efficiency of fly ash, thereby facilitating the co-disposal of organic waste and incineration fly ash.

3.3 Effect of inoculum on the pH of the leachate in the co-leaching system

Leachate pH would affect the removal of water-insoluble chlorine from fly ash. In the simulated co-leaching experiments of incineration fly ash and organic waste, the variation of leachate pH of different leaching columns with the leaching time was shown in Fig. 5. At the beginning of co-leaching, the LAB, NI and control groups all showed an increase in leachate pH with time. For the control group, the main reason for this phenomenon was that the fly ash contained a large amount of alkaline substances, which were dissolved in the leachate and carried out of the co-leaching system, resulting in an increase in the leachate pH. For the LAB group, the organic acid content in the leachate during the initial stage of leaching was not sufficient to neutralize the alkaline substances in the fly ash, which led to an increase in the pH value of the leachate. Similar to the LAB group, the organic acid production in the NI group at the beginning of the co-leaching period was also insufficient to neutralize the alkaline substances in the fly ash. Therefore, it also showed similar pH trends to the other two groups.

With the extension of leaching time, the leachate pH value in the control group decreased slightly and remained at about 12, which was similar to the initial pH value of the fly ash. Subsequently, the leachate pH of the control group increased again. In addition, the leachate pH of the LAB and NI groups showed

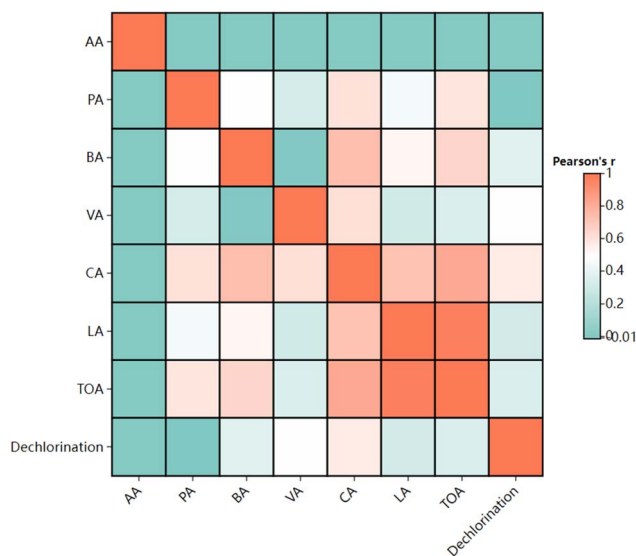


Fig. 4 Pearson correlation analysis.



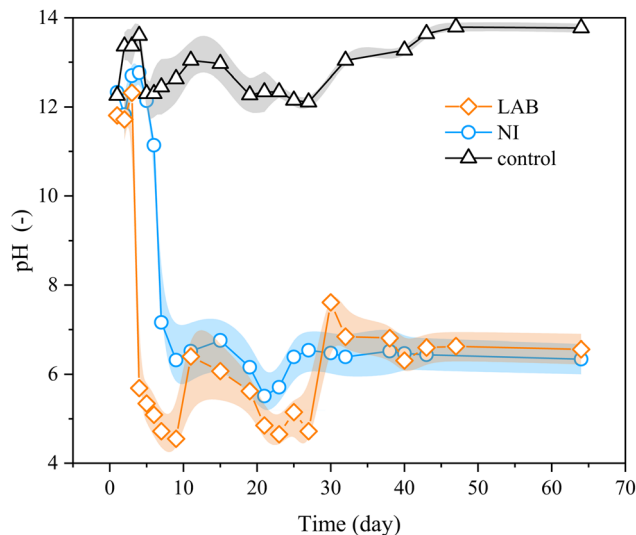


Fig. 5 Variation of pH with time.

a sharp decrease from day 3 and day 7 of leaching, respectively. Combined with Fig. 3, it is clear that the organic waste in the system produced a large amount of organic acids under the action of microorganisms, and these organic acids neutralized the alkaline substances in the fly ash during the leaching process, which led to the decrease of leachate pH. At the middle and late stage of leaching (day 20 to day 64), the leachate pH of the LAB and NI groups increased slowly with time, and finally stabilized at about 6. The main reason for this phenomenon is that as the degradation of organic waste proceeds, the bioavailable organic matter content in the organic waste gradually decreases, resulting in a decline in the acid production capacity of organic wastes. However, the alkaline substances in fly ash continue to be dissolved and released during the leaching process, which eventually causes the leachate pH to gradually increase and then stabilize.

3.4 Effect of inoculum on chlorine content in fly ash residues

The residual mass of fly ash and its chlorine content of each group were measured after the end of co-leaching, and the results were shown in Fig. 6. The mass of residual fly ash residue in the LAB, NI and control groups accounted for 65.6%, 67.1% and 84.3% of the initial mass of fly ash, respectively (Fig. 6). The mass loss of fly ash was mainly due to the following two reasons: (1) fly ash contained a large amount of soluble substances, such as soluble chloride salts (*e.g.*, NaCl, KCl, Ca(OH)Cl, *etc.*),^{27–29} which were dissolved in the solution and removed from the co-leaching system during the leaching process; (2) the organic acids produced by the organic waste under the action of microorganisms could disrupt the mineral structure of the fly ash and dissolve some water-insoluble substances present in fly ash. This also led to the fact that the residual fly ash mass of the two experimental groups (*i.e.*, LAB group and NI group) was 18.7 and 17.2 percentage points lower than that of the control group, respectively.

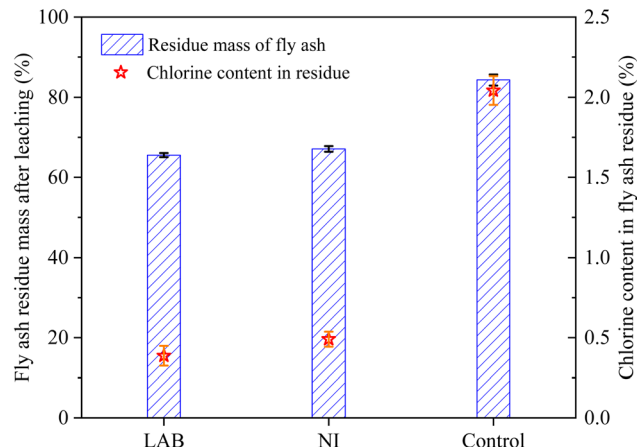


Fig. 6 Residual mass and chlorine content of fly ash after leaching under different treatments.

3.5 Effect of inoculum on the mineral phase of fly ash

In order to further investigate the changes of mineral phase composition of the fly ash before and after co-leaching, the mineral phase composition of the residues from original fly ash, and from the LAB group, NI group and control group after leaching was determined by X-ray diffraction (Fig. 7). The water-soluble chlorine in the original fly ash mainly existed in the form of KCl, NaCl, Ca(OH)Cl, and CaCl₂·4H₂O. For the control group, these water-soluble chlorine salts could not be observed in the XRD spectrum of the fly ash residue after leaching, and the relative abundance of water-insoluble chlorine (*i.e.*, AlOCl) increased. This was mainly due to the dissolution of soluble substances in the fly ash during the leaching process, which increased the relative abundance of insoluble AlOCl. In addition, for the LAB group and NI group, water-soluble chlorine (KCl, NaCl, Ca(OH)Cl, CaCl₂·4H₂O) in the fly ash residue of both groups also completely disappeared after leaching.

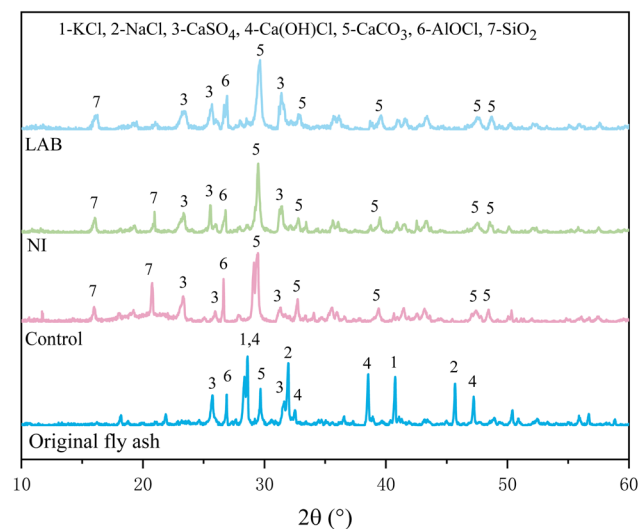


Fig. 7 XRD patterns of original fly ash and residues after different leaching treatments.

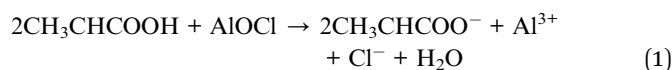


Table 2 Chlorine content of the fly ash residues under different leaching methods

Leaching methods	Cl (%)	Maximum addition ^a (%)
LAB	0.39	10.30
NI	0.49	8.16
Control	2.04	1.96
Three-step lactic acid fermentation broth washing ^b	1.5	2.67
Three-step water washing ^b	6.0	0.67

^a Assuming that other raw materials for cement kiln are chloride-free. ^b The data are based on our previous research published in Waste Management.

However, different from the control group, the XRD spectra showed that the relative abundance of water-insoluble chloride salts (*i.e.*, AlOCl) in the fly ash residue of the LAB and NI groups decreased to different degrees. This may be due to the reaction of AlOCl with organic acids (*e.g.*, lactic acid) in the leachate during leaching (eqn (1)).



Notably, XRD patterns also showed that the relative abundance of CaCO₃ in the fly ash residues of the LAB and NI groups was greater than that of the control group. It is well known that a large amount of CO₂ is produced during the anaerobic fermentation of organic waste (*e.g.*, eqn (2)),^{30–32} and this CO₂ can be neutralized by the alkaline substances of the fly ash, which leads to an increase in the CaCO₃ content of the fly ash residue. This phenomenon also indicates that co-leaching of organic waste and incineration fly ash can not only realize the dechlorination and calcium preservation of fly ash, but also reduce the greenhouse gas emissions during the fermentation of organic waste.



3.6 Effect of inoculum on the proportion of fly ash that can be added to the kiln

The amount of water-insoluble chlorine removed from the fly ash can be calculated by measuring the chlorine content in the fly ash residue after the leaching process, and the calculated results are shown in Table 2. As shown in Table 2, the water-insoluble chlorine removal amount from the fly ash in the LAB and NI groups (the dechlorination amount for the control group represents the removal of water-soluble chlorine only) was 85.11% and 80.89%, respectively. Meanwhile, combined with the requirement of chlorine amount in the raw materials of cement kiln co-disposal (HJ662-2013) (<0.04%), the proportion of fly ash that could be blended into the kiln for LAB, NI and control groups was 10.30%, 8.16% and 1.96% of the total kiln feed, respectively. Compared with the optimal result from two-step static leaching,²¹ the co-leaching of organic waste and fly ash can further increase the kiln incorporation rate of fly ash by 3.6 percentage points. In summary, co-leaching can increase the removal efficiency of water-insoluble chlorine from fly ash, thus

significantly increasing the proportion of fly ash that can be used in cement kiln co-disposal processes. This is conducive to establishing cement kiln co-disposal technology as the mainstream technology for incineration fly ash treatment and disposal.

3.7 Calculation of water savings from co-leaching of organic waste and fly ash

According to Fig. 2 and Table 1, when achieving the same amount of dechlorination as that of the control group (separate leaching of fly ash), the water consumption of the LAB group and the NI group was 160.2 mL and 169.4 mL, respectively. Therefore, compared with separate fly ash leaching, the dechlorination of incineration fly ash by co-leaching of organic waste and fly ash could save at least 48.7% of the pure water required.

4. Conclusion

In this study, the effect of inoculum on the dechlorination efficiency of fly ash was investigated by co-leaching of organic waste and incinerated fly ash. Results shown that at the end of co-disposal leaching, the chlorine content in fly ash residue of LAB, NI and control groups (pure fly ash) was 0.39%, 0.49% and 2.04%, respectively. Deep dechlorination could be achieved even without inoculation of exogenous bacteria when co-disposing organic waste and fly ash. Moreover, the allowable proportion of fly ash in the kiln feed for LAB group, NI group and control group was 10.30%, 8.16% and 1.96% of the total material, respectively, which was up to 3.6 percentage points higher than the optimal proportion achieved in static leaching. To achieve the same amount of dechlorination, the co-leaching of organic waste and fly ash can save at least 48.7% of water compared with treating incineration fly ash alone. Therefore, co-disposal of organic waste and incineration fly ash is an economical and efficient method for dechlorination.

Conflicts of interest

There are no conflicts of interest to declare.

Data availability

The data that support the findings of this study are available from the corresponding author on reasonable request.



Acknowledgements

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