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Characterization of solidification for disposal of hazardous waste landfill leachate

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Abstract

Hazardous waste landfill leachate (HWLL) with high concentrations of salt and pollutants has created a bottleneck at hazardous waste landfills. This study applied a cement-based curing method to the disposal of HWLL. The highest contaminant fixing rate was achieved by adjusting the composition and proportion of the curing base, the content of additives, and the liquid-solid (L/S) ratio of the leachate to the curing base. The fixing rates for chemical oxygen demand and salt content in HWLL reached the highest values of 95.1% and 86.1%, respectively, when the Portland cement to metakaolin ratio was 3:2; the L/S was 1; and diatomite and activated carbon were added at 0.5% and 0.25%, respectively. The addition of glass fiber to the curing base improved the crack resistance of the solidified product. A simulated landfill experiment further indicated that after 116 days of leaching, the leachate effluent pollutant concentrations of the landfill column were lower than the effluent standard. Solidification is a feasible method for HWLL disposal.

Keywords Hazardous waste landfill leachate · Solidification · Fixing · Glass fiber · Landfill

Introduction

Safe landfilling is an important means for hazardous waste disposal. It is widely used because of simple operation, but this method can create leachate problems. Hazardous waste landfill leachate (HWLL) is quite complex. It contains a large amount of toxic and hazardous substances, and the concentration of chemical oxygen demand (COD) in leachate can be up to tens of thousands of milligrams per liter. It also contains a large number of refractory macromolecular organic substances (Suliasih et al. 2010). At present, the leachate treatment process is generally a combined process of biological and physicochemical methods (Colombo et al. 2019). However, HWLL is more harmful than the leachate from municipal solid waste landfills. It has higher salinity and

refractory COD and harmful components, such as heavy metals, which can kill microorganisms and affect the biological treatment processes (Chen et al. 2019; Macêdo et al. 2019).

Cement is widely used in civil engineering, water conservancy, national defense, and other projects as an important construction material (Liu et al. 2018). It also has been applied in waste disposal (Pan et al. 2018). Cement is a powdery hydraulic, inorganic cementitious material. Concrete made of cement-bonded macadam is not only strong after hardening but also resistant to salt-water corrosion to some extent by using noncorrosive reinforcement (Younis et al. 2018). Metakaolin (MK) is an anhydrous aluminum silicate ($\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) formed by dehydration of kaolin ($\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$). MK is a mineral admixture with high volcanic ash activity. Due to its irregular molecular arrangement, it exhibits a thermodynamic stability state and is gelatinous under appropriate excitation. It is mainly used as a concrete additive. MK can form a polymer under an alkali activator and polymerizes under alkaline conditions (Arellano-Aguilar et al. 2014). We used CaO to adjust the pH and provided basic conditions for the polymerization of MK. At the same time, the calcium is more favorable for the formation of polymer structures (Si et al. 2018). Diatomite is a siliceous rock with the SiO_2 -based chemical composition, containing a small amount of Al_2O_3 ,

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Fe₂O₃, CaO, MgO, and other organic matters. Further, it has a specific surface area of around 40–65 m²/g with high adsorption potential (Mu et al. 2018; Nikjoo et al. 2018). Generally, diatomite is often used to treat wastewater because of its adsorption ability (Chu et al. 2008; Sun et al. 2018); in addition, adding an appropriate amount of diatomite to cement can increase cement strength (Ergün 2011); activated carbon (AC) is a highly adsorbable material, which is widely used in waste treatment processes for adsorbing pollutants (Mohammad-Pajooch et al. 2018). Adding it to the curing base can increase the fixing rate of pollutants in HWLL. In order to improve the crack resistance of the curing blocks and enhance the curing effect, we added glass fiber with the crack resistance as the curing materials. Glass fiber is an inorganic nonmetallic material with excellent insulation, heat resistance, corrosion resistance, and mechanical strength (Li et al. 2014).

The concept of geo-polymer was proposed by Davidovits in 1978 (Davidovits 1994). It is an inorganic polymer composed of AlO₄ and SiO₄ tetrahedral structural units. This material has excellent mechanical properties and resistance to acids, alkalis, fire, and high temperature (Bonet-Martínez et al. 2018; Zuhua et al. 2009). It also has a certain salt tolerance and can be used in some special construction situations, such as buildings that have been eroded by seawater for many years; the addition of MK to concrete increases its resistance to chloride ion penetration (Hou et al. 2016). In this research, we tested the HWLL solidification using the materials described above using the geo-polymer concept. The objective was to find a feasible HWLL disposal method to overcome the bottleneck problem at hazardous waste landfills. We used the results of laboratory testing to carry out a landfill column scale-up experiment for the analysis of the leaching behavior of pollutants when external water intrudes into cured leachate blocks buried in a landfill.

Materials and methods

Batch tests of HWLL solidification effect

HWLL was collected from the hazardous waste landfill in Shaoxing City, Zhejiang Province, China. The curing base used included CaO (CAS1305-78-8, Wuxi), Portland cement (PC, grade P.O 42.5R), and MK obtained after 600 °C–900 °C of dehydration, and the additives used included diatomite (CAS 91053-39-3), AC (CAS 7440-44-0), and glass fiber.

Three experimental scenarios were conducted at room temperature in the laboratory, and the total curing base mass was maintained at 20 g; all experiments were performed three times as parallel.

The first scenario was designed to find the optimal solidification base composition and the liquid-solid (L/S) ratio of the HWLL in the curing system. First, the pH value of HWLL

was adjusted to 9.5 ± 0.5 using CaO. Then, the solidification base composition included PC and MK, and PC and MK were mixed and stirred in batch mass ratios of 1:0, 3:1, 3:2, 1:1, 2:3, and 1:3 to form a curing base. The pH-adjusted HWLL was added at L/S ratios of 0.7, 0.8, 1, 1.2, and 1.3. Then, the matrix was stirred evenly into a thick paste and placed in dish mold to air-dry at room temperature. Five days later, leaching test on the blocks cured by air-drying was carried out (HJ 557-2010).

The second scenario was designed to find the best quantity of diatomite and AC added in the curing system. The solidification base composition and L/S ratio of HWLL were based on the best results from the first experiment. This scenario included single-factor and multifactor experiments. The single-factor experiment design was as follows: first, 0.1 g, 0.2 g, 0.3 g, 0.5 g, and 0.8 g of diatomite were added to admixture ratios at PC/MK ratio of 3:2 and at an L/S ratio of 1:1. Then, 0.05 g, 0.1 g, 0.15 g, 0.2 g, and 0.3 g of AC were added to admixture ratios at PC/MK ratio of 3:2 and at an L/S ratio of 1:1. The Design-Expert software was used to design the multifactor experiments, and its user-defined design function was used to optimize the curing recipe parameters. The detailed experimental design was listed in Table 1. A total of 36 experiments were performed. Mixing, molding, and curing were performed the same as in the first scenario.

In the third scenario, curing system materials and the L/S ratio of HWLL were fixed. In this scenario, 0 g, 0.3 g, 0.5 g, and 0.8 g of glass fiber were added to the curing system to enhance the crack resistance of the solidified block. The experimental procedure is shown in Fig. 1. Afterward, cracking resistance tests of the curing blocks were carried out (JC/T 951-2005).

Evaluation of solidified HWLL leaching behavior after landfilling

Landfill simulation reactor

A set of cylindrical landfill simulation reactors with inner diameter of 40 cm and height of 200 cm was constructed. The top of the landfill column had a water distribution device, the upper layer was not covered, the middle was filled with cured blocks, a steel plate with holes was installed at the bottom, and the bottom had a leachate collecting space and water outlet.

Reactor filling process and operation

HWLL was pretreated according to the best curing formula that was developed in the above laboratory test, namely, 1.7 L of HWLL (adjusted to pH 9.5 ± 0.5 by CaO addition), 1-kg PC, 0.7-kg MK, 4.2-g AC, 8.3-g diatomite, and 41.7-g glass fiber. The curing process is shown in Fig. 1. After air-drying for 5 days, 150 kg/0.163 m³ of cured blocks was placed into

Table 1 The detailed experimental design of multifactorial experiment

PC (g)	MK (g)	HWLL (mL)	Diatomite (mg)	AC (mg) 1	AC (mg) 2	AC (mg) 3	AC (mg) 4	AC (mg) 5	AC (mg) 6
12	8	20	0	0	0.05	0.1	0.15	0.2	0.3
12	8	20	0.1	0	0.05	0.1	0.15	0.2	0.3
12	8	20	0.2	0	0.05	0.1	0.15	0.2	0.3
12	8	20	0.3	0	0.05	0.1	0.15	0.2	0.3
12	8	20	0.5	0	0.05	0.1	0.15	0.2	0.3
12	8	20	0.8	0	0.05	0.1	0.15	0.2	0.3

(The addition amount of PC, MK, HWLL, and diatomite is determined, six kinds of quality AC are sequentially added, and a total of 36 experiments are included)

the landfill column reactor, and the operation process was shown in Fig. 2.

The landfill reactor experiment simulated the annual rainfall in Shaoxing City, Zhejiang Province, China. The annual rainfall of 1700 mm of Shaoxing City was cited from China Meteorological Administration website. Simulated precipitation was applied to the reactor weekly with 4 L of water. A leachate sample was collected from the bottom of the reactor weekly and analyzed, three samples were taken as parallel each time, and every sample was measured three times during the measurement.

Analyses

Batch test

The mass of cured blocks was weighed. Then, the blocks were ground to a particle size of less than 5 mm. 5 g of the ground sample was mixed with 250 mL of distilled water. Then, the mixture was shaken in a horizontal shaker at a speed of 120 rpm at room temperature. Eight hours later, the mixture was allowed

to settle for 16 h. Finally, a 0.45-μm membrane (13 mm, water system, JIN TENG) was used to filter the extract. The original HWLL and the extract from the cured block were simultaneously measured by conductivity meter (Mettler-Toledo, FE30, China) and COD meter (HACH, DR2800, Germany).

The HWLL salt fixing rate was calculated as Eq. 1:

$$F = [(V1 \times SAL1 - SAL2 \times V2 \div M2 \times M1) / (V1 \times SAL1)] \times 100\% \tag{1}$$

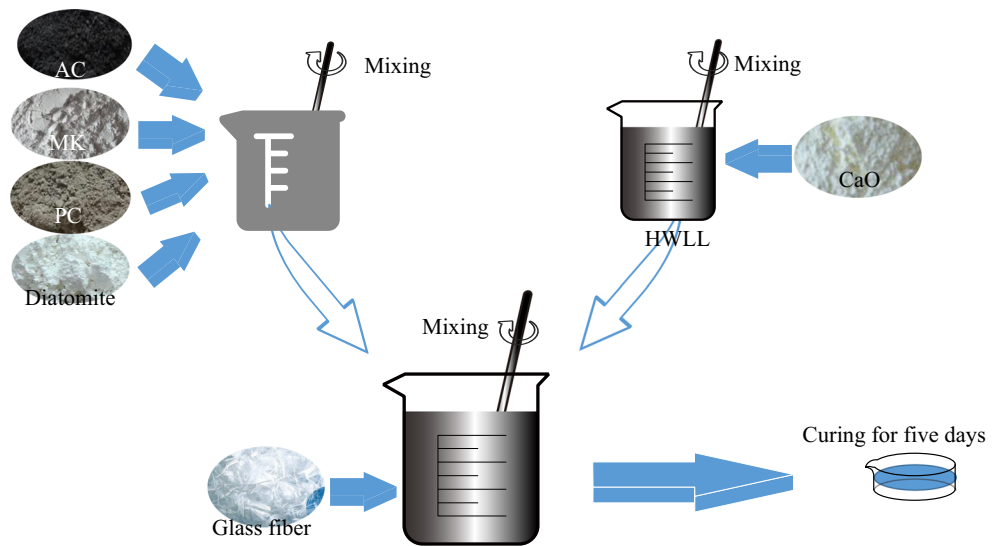
where *F* is the salt fixing rate, *V1* is the volume of HWLL added, *V2* is the volume of the extract (250 mL), *SAL1* is the salt concentration of HWLL, *SAL2* is the salt concentration of the extract, *M1* is the total mass of the curing block, and *M2* is the mass of the cured block used for the leaching tests (5 g).

The HWLL COD fixing rate was calculated as Eq. 2:

$$G = [(V1 \times COD1 - COD2 \times V2 \div M2 \times M1) / (V1 \times COD1)] \times 100\% \tag{2}$$

where *G* is the COD fixing rate, *V1* is the volume of HWLL added, *V2* is the volume of the extract (250 mL), *COD1* is the

Fig. 1 Experimental process flow



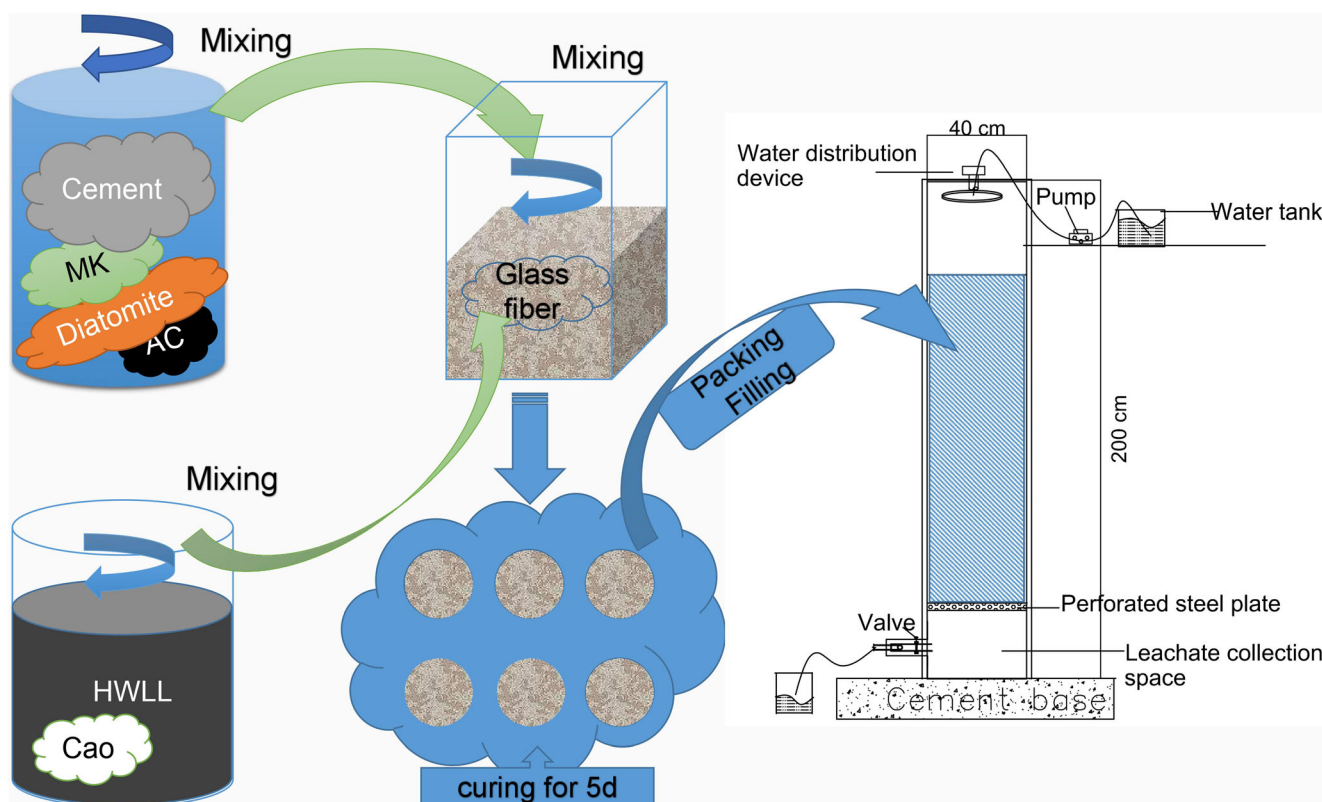


Fig. 2 Simulated landfill reactor filling process

COD of HWLL, COD_2 is the COD of the extract, M_1 is the total mass of the cured block, and M_2 is the mass of the cured block used for the extraction (5 g).

conductivity meter (Mettler-Toledo, FE30, China), and NH_3-N was measured by an ultraviolet–visible spectrophotometer (Metash, V-5800, China).

Leachate test

The volume of the collected leachate was measured with a graduated cylinder. The concentrations of Cl^- were measured by an ion chromatograph (Metrohm, 863 compact IC Autosampler, Switzerland), COD was measured by a COD meter (HACH, DR2800, Germany), salt was measured by a

Statistical analysis method

The SPSS software was used to analyze the significant differences between measured data results at the 0.05 probability level. Different letters represent the difference between the two, and the same letter represents that the difference is not significant.

Fig. 3 The effect of PC/MK on the COD and salt fixing rates

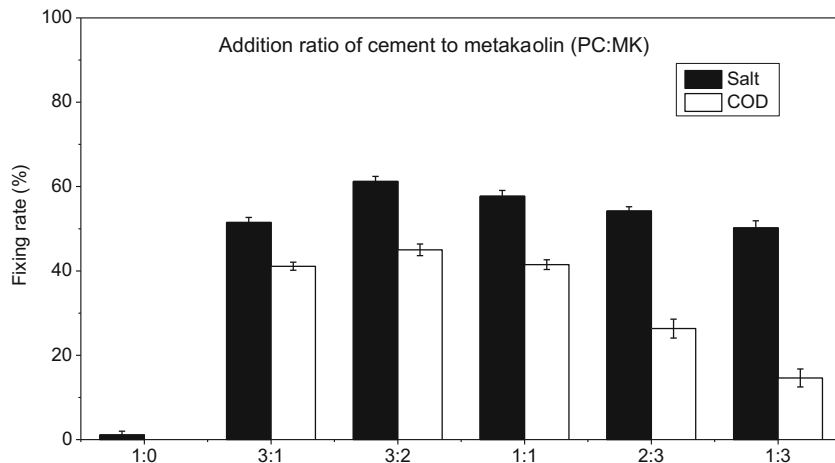
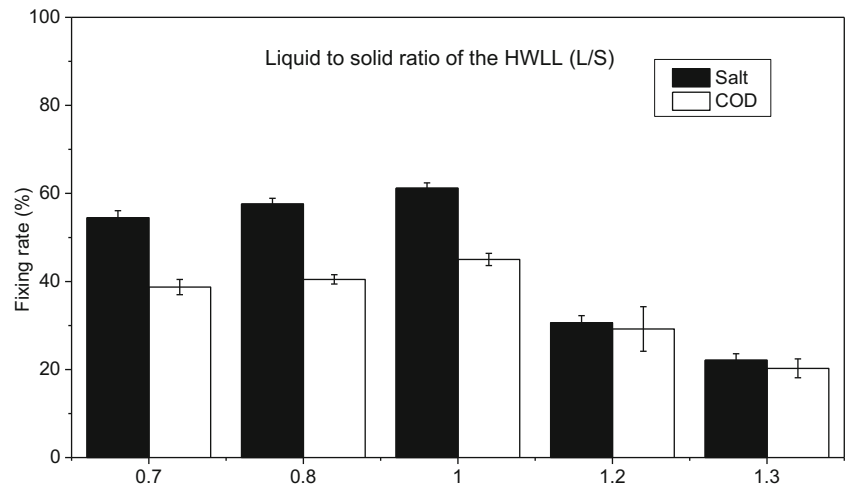


Fig. 4 The effect of L/S ratios on the COD and salt fixing rates



Results and discussion

Optimization of HWLL solidification

Characteristics of the HWLL

The characteristics of the HWLL used in the laboratory experiment were as follows: COD_{Cr} , 15,763 mg/L; salt, 76.6 g/L; Cl^- , 35,709 mg/L; $\text{NH}_3\text{-N}$, 9756.5 mg/L; Cr, 5.52 mg/L; Ni, 12.67 mg/L; Zn, 1068 mg/L; and Cu, 10.21 mg/L. The content of Hg and As was extremely low in the HWLL and pH was 6.8.

Effects of PC/MK and L/S ratios on pollutant fixing rate

PC and MK content had a measurable effect on HWLL solidification. As shown in Fig. 3, when the L/S was 1, the COD fixing rate increased from 0% to $45 \pm 1.37\%$ as the PC/MK ratio increased from 1:0 to 3:2. The corresponding salt fixing rate increased from $1.10 \pm 0.9\%$ to $61.2 \pm 1.2\%$. However, the COD and salt fixing rate decreased from $45 \pm 1.37\%$ and $61.2 \pm 1.2\%$, respectively, to $14.6 \pm 2.1\%$ and $50.2 \pm 1.7\%$ as the

PC/MK ratio further increased from 3:2 to 1:3. The highest fixing rates for COD and salt were observed at a PC/MK ratio of 3:2. Thus, an MK content that is too low or too high is not conducive to the optimal pollutant fixing rate, which is consistent with other published studies (Wang et al. 2019). MK undergoes activated and polycondensation reactions in this system to form a geo-polymer with three-dimensional network structure composed of AlO_4 and SiO_4 , which has excellent resistance to acids, alkalis, fire, high temperature, and salt (Belmokhtar et al. 2017). At the same time, the PC undergoes a hydration reaction and forms a hard cured block that fixes contaminants within the block (Wei 2017).

Then, the effect of L/S ratio on pollutant fixing rate was studied. As shown in Fig. 4, when the PC/MK ratio was 3:2, with L/S ratios from 0.7 to 1, the HWLL COD and salt fixing rate increased from $38.7 \pm 1.7\%$ and $54.5 \pm 1.6\%$ to $45 \pm 1.4\%$ and $61.2 \pm 1.2\%$, respectively. However, as the L/S ratio increased further from 1 to 1.3, the HWLL COD and salt fixing rate decreased from $58.9 \pm 1.4\%$ and $61.2 \pm 1.2\%$ to $20.3 \pm 2.1\%$ and $22.1 \pm 1.5\%$, respectively. When too much HWLL was added, the curing system could not accommodate it.

Fig. 5 The effect of adding diatomite to 20 g of curing base after PC/MK solidification on the COD and salt fixing rates (a, b, c, and d stand for the salt fixing rate were significant difference at the 0.05 probability level and A, B, C, and D stand for COD fixing rate)

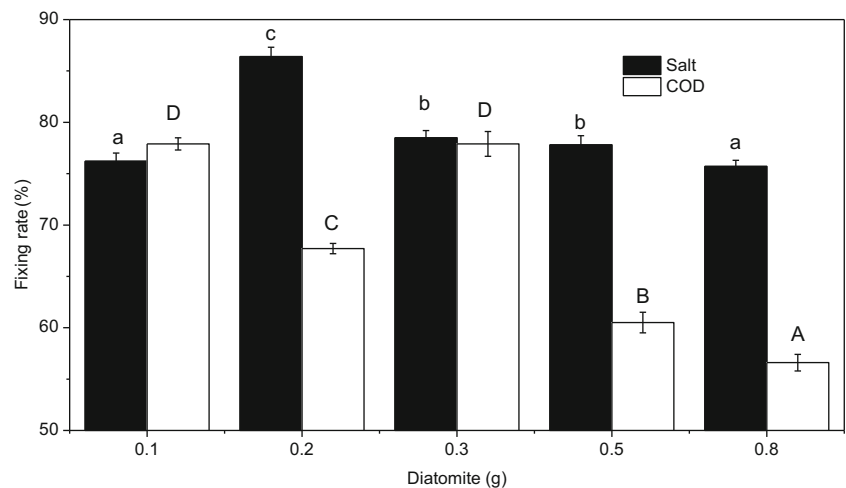
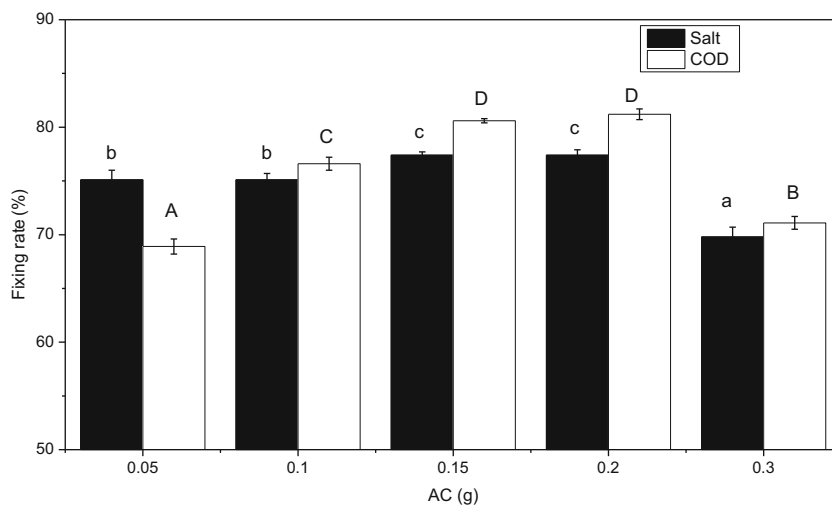


Fig. 6 The effect of AC on curing base after PC/MK solidification (the lowercase and uppercase letters stand for the salt and COD fixing rate were significant difference at the 0.05 probability level, respectively)



Therefore, the L/S ratio also affected the pollutant fixing rate. The highest fixing rates for COD and salt were observed when the L/S ratio was 1.

Additives to improve the curing system

According to the cured block leaching results in Figs. 3 and 4, when the PC/MK and L/S ratios were 3:2 and 1, respectively,

the HWLL COD and salt fixing effect were the highest. In order to get the best fixing results, additives were added to mixtures with the optimal PC/MK and L/S ratios. As shown in Fig. 5, when the diatomite content increased from 0.1 g to 0.2 g in 20 g of curing base after PC/MK solidification, the salt fixing rate increased from $76.2 \pm 0.8\%$ to $86.4 \pm 0.9\%$. However, after that, adding more diatomite reduced the salt fixing rate. This is related to the adsorption and cohesiveness

Fig. 7 The combined effect of diatomite and AC to 20 g on curing base solidification (a and b represent the salt fixing rate and the COD fixing rate, respectively)

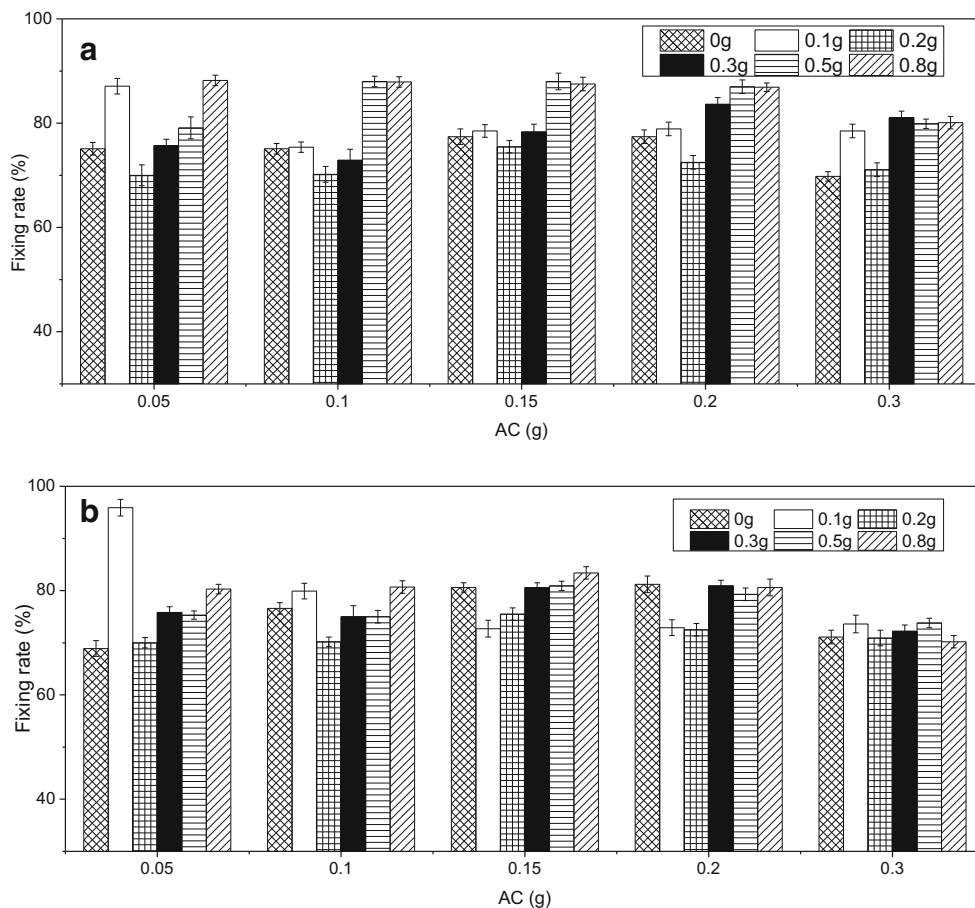
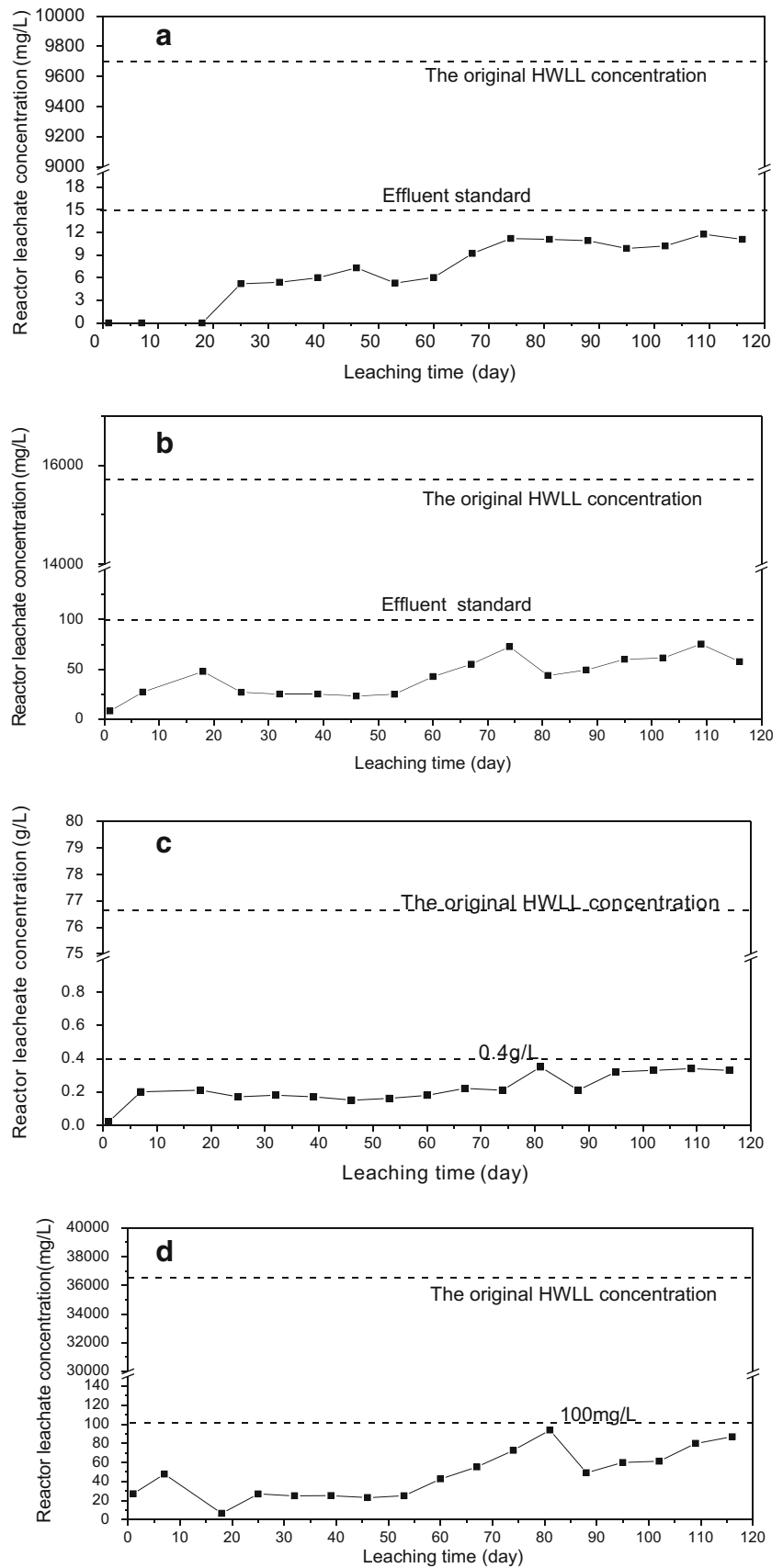


Fig. 8 The variation of $\text{NH}_3\text{-N}$ (a), COD (b), salt (c), and Cl^- (d) concentrations in leachate effluent



of diatomite (Houwaida Nefzi et al. 2019). As for the COD fixing rate, there was a small difference from the salt fixing rate. The highest COD fixing rate was $77.9 \pm 1.2\%$, when the diatomite content was 0.3 g in 20 g of curing base after PC/MK solidification.

We found that the fixing rate was also improved by adding AC to 20 g of curing base after PC/MK solidification. As shown in Fig. 6, when the AC content increased from 0.05 g to 0.2 g in 20 g of curing base after PC/MK solidification, the HWLL COD and salt fixing rate increased from $68.9 \pm 0.7\%$ and $75.1 \pm 0.9\%$ to $81.2 \pm 0.5\%$ and $77.4 \pm 0.5\%$, respectively. However, after that, as more AC was added, the observed COD and salt fixing rate decreased. The highest COD and salt fixing rates were $81.2 \pm 0.5\%$ and $77.4 \pm 0.5\%$, respectively, when the AC content was 0.2 g in 20 g of curing base after PC/MK solidification, which was related to the adsorption of AC (Yang et al. 2019).

The combined effect of adding diatomite and AC to 20 g of curing base after PC/MK solidification was studied. As shown in Fig. 7a, the highest HWLL salt fixing was $88 \pm 1.1\%$ when the contents of diatomite and AC were 0.5 g and 0.1 g, respectively, but the highest HWLL COD fixing rate was $95.1 \pm 1.6\%$ when the contents of diatomite and AC were 0.1 g and 0.05 g, respectively (Fig. 7b); the salt fixing rate at this point was $87.1 \pm 1.5\%$. No change in the salt fixing rate was observed with AC contents of 0.05 g to 0.3 g, when the COD fixing rate first increased and then decreased. This is due to the antagonistic effect between AC and diatomite, so as the combined content of diatomite and AC increased, the observed pollutant fixing rate decreased. When the added content of diatomite was from 0.2 g to 0.8 g, the salt and COD fixing rate gradually increased. This may be due to the increasing content of diatomite, and it began to play a more dominant role that did the AC.

Increase in crack resistance

Glass fiber was added to the curing base after treatment with PC/MK, diatomite, and AC. When the mass ratio of curing base to glass fiber was 40:1, the cracking index was 760 mm, namely, 0.25 times that of the cured block without glass fiber. It was found that the addition of glass fiber could increase the crack resistance of the curing body fourfold when the mass ratio of the curing base to the glass fiber was 40:1. This finding was consistent with published research (Lacki et al. 2019). Adding glass fiber was beneficial to the long-term effect of curing.

Leaching behavior of solidified HWLL in landfill site

The landfill column reactor was subjected to simulated rainwater leaching for 116 days, and during this period, only a small amount of salt and organic matter leached out. As shown in Fig. 8, the COD and $\text{NH}_3\text{-N}$ concentrations in leachate

effluent from the landfill column could meet the first order of national integrated wastewater discharge standard (GB 8978–1996), the first-order standard COD limit is 100 mg/L, and the $\text{NH}_3\text{-N}$ is 15 mg/L. And the COD, $\text{NH}_3\text{-N}$, Cl^- , and salt concentrations in the landfill column leachate effluent were far lower than the corresponding initial HWLL concentrations. The chloride ion and salt concentrations in the leachate also did not exceed 100 mg/L and 0.4 g/L, respectively. This means that the HWLL pollutants can be safely fixed in the cured blocks after solidification. Thus, this process is a feasible disposal method for HWLL and other wastewater with high salt and organic concentrations.

Conclusion

A cement-based curing method was used to dispose of HWLL. When the PC/MK and L/S ratios were 3:2 and 1, respectively, adding 0.1 g diatomite and 0.05 g AC to 20 g of curing base after PC/MK solidification achieved the highest HWLL COD and salt fixing rates of $95.1 \pm 1.1\%$ and $87.1 \pm 1.5\%$, respectively. The addition of glass fiber can further improve the crack resistance of cured blocks and benefit long-term curing. The landfill leaching simulation experiment showed that all of the leachate pollutant concentrations from solidified HWLL meet the effluent standard after 116 days of leaching. Thus, cement-based solidification is a feasible disposal method for HWLL and other wastewater with high salt and organic concentrations.

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