



Application of hydration reaction on the removal of recalcitrant contaminants in leachate after biological treatment



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ABSTRACT

Leachate contains amounts of non-biodegradable matters with COD range of 400–1500 mg/L after the biological treatment, and should be removed further to attain the Chinese discharge standards. Hydration reaction has the potential to combine and solidify some recalcitrant substances, and thus could be applied as the advanced leachate treatment process. It was found that COD and NH₃-N decreased from 485 to <250 mg/L and 91 to 10 mg/L, with the removal rate over 50% and 90% in the first 6 d, respectively, and COD and NH₃-N removal capacity were around 23.7 and 9.2 mg/g under the test conditions. The percentage of the substances with low Mn range of <1000 decreased from 32.9% to 3.2% in leachate after hydration reaction. Tricalcium aluminate, tricalcium silicate and dicalcium silicate were the most activity compounds successively for the pollutant removal in leachate, and hydration reaction could be the option for the advanced wastewater treatment process thereafter.

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1. Introduction

Landfill leachate is one of the important byproducts, and around 40,000–60,000 ton leachate generates in landfill per day in China, which is about 20–30% of refuse disposal (Ziyang et al., 2009). Leachate contains dissolved organic matter (DOM), inorganic macro-components, heavy metals, xenobiotic organic compounds (XOCs) and some humic substances (Christensen et al., 2001; Kulkowska and Klimiuk, 2008). Most of readily degradable organic matters, such as volatile fatty acid, could be removed efficiently by the traditional biological treatment processes, for example, lagoon with activated sludge, aged refuse bio-filter, and membrane bio-reactor (Christensen et al., 2001; Fan et al., 2007; Ahmed and Lan, 2012; Renou et al., 2008; Huang et al., 2009; Li et al., 2010; Ziyang et al., 2009). The corresponding COD values in those leachate effluents keep around 400–1500 mg/L even with a longer hydraulic retention time (HRT) and sludge retention time (SRT), according to the practical experiences, since some recalcitrant compounds, such as fulvic-like and humic-like compounds, reductive inorganic components (such as Cl⁻, NO₃⁻), and xenobiotic organic compounds, are still remained, which results in a high COD value, even the corresponding TOC and BOC₅ are low (Li et al.,

2010; Ziyang et al., 2009). N, expressed either as NH₃-N or NO₃-N, is another critical parameter, with a wide range of 50–1000 mg/L (Christensen et al., 2001; Ahmed and Lan, 2012; Ziyang et al., 2009), and should be removed further.

With the stricter leachate discharge standard, some physical-chemical treatment processes, such as chemical oxidation, air stripping, ion exchange, membrane process, adsorption on active carbon and coagulation–flocculation (Meeroff et al., 2012; Foo and Hameed, 2009; Ozturk et al., 2003; Renou et al., 2008), are coupled with those biological treatment processes mentioned above, while a high operation cost, the stricter operation condition, the difficult handling of the by-products, or the low removal efficiency, prohibit the wide application of these techniques in the practical leachate treatment projects.

Hydration reaction has been common used in construction and environmental field (e.g. for stabilization and solidification) to form a solid mass (Bentz, 2008; Kirby and Biernacki, 2012; Paria and Yuet, 2006), and some organic matters, ions (anion and cations) are found to be incorporated into the cement matrix during the hydration process (Paria and Yuet, 2006). It has also been observed that the hydration products, e.g. Calcium silicate hydrates (C–S–H), Ca(OH)₂ (C–H), AFt (Al₂O₃–Fe₂O₃-tri) and AFm (Al₂O₃–Fe₂O₃-mono), could gradually bond with a wide range of contaminants, such as heavy metals, inorganic matter and some organic matters (Jansen et al., 2012; Stampino et al., 2009; Yousuf et al., 1995), which are the typical contaminants involved in the wastewaters.

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Up to now, most of works on hydrating cements focused on the influence of additives, such as metals, anions, polymers, on the cement hydration development processes and the properties of the cementitious products (Jansen et al., 2012; Stampino et al., 2009), and the water/cement ratio and the reaction time were usually around 0.5 and 28 d, respectively, to obtain the hardened cement with the high flexural strength and the tensile strength. To our knowledge, limited works were reported on the application of the hydration reaction in the wastewater treatment (Paria and Yuet, 2006), and the hydration rate, the inclusion of contaminants into the hydrating cement are still the open questions for the hydrating cement–wastewater system.

The aim of the work is to develop a novel advanced leachate treatment process using hydration reaction. The pollutant removal efficiency was evaluated in terms of COD, TOC, $\text{NH}_3\text{-N}$ and TN under the optimum operation conditions, and the interactions between the hydrating cement and contaminants were investigated from the micro-structure perspective, i.e. the pollutant distribution, the speciation of the different phases form and the incorporation appearance.

2. Materials and methods

2.1. Reagents and leachate samples

The hydrating cement applied involved several main mineral compositions, i.e. 50% of C_3S (i.e., $3\text{CaO}\cdot\text{SiO}_2$), 25% of C_2S (i.e., $2\text{CaO}\cdot\text{SiO}_2$), 12% of C_3A (i.e., $3\text{CaOAl}_2\text{O}_3$), 8% of C_4AF (i.e., $4\text{CaO}\cdot\text{Al}_2\text{O}_3\cdot\text{Fe}_2\text{O}_3$), and 3.5% of CSH_2 .

Leachate samples were collected from the collection tank after treated by aged refuse bio-filter in Laogang Refuse Landfill, as reported by Li et al. (2010). COD in leachate sample was 485 mg/L, while BOD_5 were below the determination limit. pH and conductivity were around 8.14 and 1029 $\mu\text{S}/\text{cm}$, respectively. TN was 224 mg/L, and the corresponding $\text{NH}_3\text{-N}$ was 91 mg/L. Cl^- , SO_4^{2-} and F^- concentration were 2303 mg/L, 905 mg/L and 9.9 mg/L, respectively, given that the high percentages of food waste in municipal solid waste (MSW) in China (about 50–70% in fresh refuse). Ca^{2+} in leachate reached to 90.5 mg/L, possibly due to the mix of the construction and demolition waste in landfill. Fe, Al and Zn concentration were 3.2, 6.6 and 2.0 mg/L, respectively.

2.2. Experiments procedure

Fifty gram hydrating cement and 500 mL leachate were introduced into a quartz glass reactor of 1.0 L equipped with a magnetic stirrer at 100 rpm. The supernatant was centralized for analysis after the end of the defined interval time of 24 h, and the residual cement slurry stayed in the reactor for the next round, which was kept running until COD in leachate effluent was above 300 mg/L. Parts of cement slurry samples were taken out from the reactor after 1, 4, 8 and 11 d and prepared for a slice after dried in the glovebox within a dry N_2 atmosphere for the analysis.

2.3. Analytical methods

pH was determined using a portable digital pH meter (PHB-4, Shanghai Leici Company, China). TOC and TN were measured by TOC-V CPH/CPN (Shimadzu, Japan). COD and $\text{NH}_3\text{-N}$ were determined by the standard methods recommended by APHA (1995). Heavy metals were determined by ICP–AES (Inductively Coupled Plasma Atomic Emission Spectrometer, Optima 2100 DV, PerkinElmer, USA) after digested by microwave-assisted acid digestion with combined acid ($\text{HNO}_3\text{:H}_2\text{O}_2\text{:HCl} = 6\text{:}0.5\text{:}3$ mL) using Microwave Extraction/Digestion System ETHOSE (Micesione, s.r.l, Italy).

Anion was analyzed by ICS-1000 (D10NEX ICS 1000 ion chromatography, DIONEX) using ion Pac ASII-HC (4 * 250 mm) as chromatographic column.

Molecular weight distribution in leachate was determined by gel permeation chromatography (SHIMADZU, Lc-10ADVP). TSK gel G4000PWc was chosen as the chromatographic column in GPC with the size of 7.8 * 300 mm, and the free trial easivial PEG/PED standards for SEC (UK, polymer standards) was applied as the standards.

The features of the cement slurry were studied by XRD, SEM and FTIR. XRD analysis was carried out by a siemens D-5000 X-ray diffractometer with Cu $\text{K}\alpha$ radiation (40 kV, 20 mA), ranging between 10° and 70° with 2 h scanning, and the cement slurry from 1, 4, 8 and 11 d were collected and analysis. In order to identify the effect of leachate pollutants on the hydration process, cement slurry from the hydrating cement–leachate system and hydrating cement–pure water system were measured and compared using NOVA Nano-SEM 230 field emission scanning electron microscope (FEI, USA) and FTIR 1000 (Perkin Elmer, USA) of a KBr pellet with a range of $400\text{--}4000\text{ cm}^{-1}$ simultaneously.

3. Results

3.1. Removal efficiency of leachate pollutants by hydration reaction

pH increased dramatically from 8.14 to 12.60 as the hydration reaction proceeded, and maintained at a high level of more than 11.95 after 3rd (Fig. 1a), ascribed that amount of $\text{Ca}(\text{OH})_2$ was generated in the initial period (Jansen et al., 2012), and then $\text{CaO}\cdot\text{SiO}_2\cdot n\text{H}_2\text{O}$ (C–S–H) consumed $\text{Ca}(\text{OH})_2$ greatly (Kirby and Biernacki, 2012), which resulted in the decrease of pH from 11.95 to below 9.00 in the 5th d. Finally it kept at a steady stage of 8.10–8.50. Conductivity in leachate increased from 1029 to about 1950 $\mu\text{S}/\text{cm}$ in the first 2 d, and then decreased to around 1000 $\mu\text{S}/\text{cm}$ after 5 d.

COD and TOC in the resultant leachate were shown in Fig. 1b. COD was around 150 mg/L in the first 2 d, with a removal rate of 70%, and then presented at a range of 150–200 mg/L in the next 6 d. The removal rate of TOC was about 55% in the first 2 d, and then decreased to about 14% dramatically in the next 4 d. Finally it reached to almost zero after the reaction for 10 d. $\text{NH}_3\text{-N}$ in leachate was below 10 mg/L, with a removal rate of above 90% in the entire operation period of 11 d (Fig. 1b). Around 40% TN was removed in leachate in the first 5 d, and then only 22% of TN removed.

Variation of cations and anions in the resultant leachates was shown in Table 1. Ca^{2+} concentration increased from 90 to 635 mg/L in the 1st d, and then decreased to about 5 mg/L, with a removal rate of 94%, which resulted from the balance between the generation of $\text{Ca}(\text{OH})_2$ and the consumption of Ca^{2+} by C–S–H. Al^{3+} was also found to increase from 6.6 to 12.7 mg/L initially, and then C–S–H generated during the hydrating process contributed to the reduction of Al^{3+} to about 2 mg/L. Cl^- , SO_4^{2-} and F^- content decreased significantly in the first 5 d, and then increased as the reaction time extended, compared to the corresponding concentration of 2303, 905 and 9.9 mg/L in raw leachate. The F^- content decreased slightly in the first 5 d through the generation of CaF_2 , since CaF_2 is a slightly soluble compound (Tacker and Stormer, 1993).

3.2. Molecular weight (MW) distribution

Variation of the MW distribution was summarized in leachate effluents, as shown in Fig. 2. The fraction with the Mn (number-average molecular weight) range of <100 K (100,000) predominated in leachate samples, with the percentage over 90%.

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