Influence of alkalinity on the stabilization of municipal solid waste in anaerobic simulated bioreactor

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ABSTRACT

Four simulated landfill anaerobic bioreactors were performed to investigate the influence of alkalinity on the anaerobic treatment of municipal solid waste (MSW). Leachate was recirculated in all the four reactors. One reactor was operated without alkalinization. The other three were operated under alkaline conditions. Na_2CO_3, $NaHCO_3$ and $NaOH$ were added to leachate in the second, third and fourth reactor, respectively. Experimental results showed that CO_3^{2-} and HCO_3^- addition had a more pronounced effect on MSW stabilization while the effect of addition of OH^- was weak. The concentration of COD, BOD_5, total nitrogen (TN), ammonium nitrogen (NH_4^+-N) and nitrate nitrogen (NO_3^--N), etc. in leachate significantly reduced in four reactors. The removal efficiencies were 90.56%, 92.21%, 92.74% and 90.29% for COD, 66.45%, 72.38%, 68.62% and 68.44% for NO_3^--N, and 96.5%, 98.75%, 97.75% and 98% for NO_2^--N in the control, Na_2CO_3, $NaHCO_3$ and OH^- added reactors, respectively. The final BOD_5/COD was 0.262, 0.104, 0.124, and 0.143, and pH was 7.13, 7.28, 7.42, and 7.24 for control, Na_2CO_3 added, $NaHCO_3$ added, and OH^- added reactor, respectively. Therefore, alkalinity addition had positive effect on the stabilization of MSW.

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1. Introduction

Anaerobic bioreactor disposal is one of the most cost-effective municipal solid waste (MSW) treatment processes. So researchers have paid more attention to this technology in recent years. However, MSW treated in anaerobic bioreactor have to meet some requirements, such as uniformly distributed moisture and water for effective pollutants removal; degradation of MSW may be retarded in their absence. Anaerobic bioreactor based on the leachate recirculation can solve these problems. Leachate recirculation enhances and accelerates conversion and stabilization of MSW by increasing the uniformity of moisture, substrate and nutrient distribution and by creating an environment that promote the rapid development of the desired microbial population [1]. Thus, landfill gas (LFG) generation rates are enhanced while the environmental impacts are reduced by containing the leachate and controlling the LFG emissions. The volume of leachate is reduced by maximizing evaporative losses during recirculation which can reduce leachate treatment capital and operation cost, and thus the overall contaminating life span of the landfill is reduced [2,3]. Circulating leachate between a landfill and a methanogenic reactor (mature landfill) can take advantage of adapted microflora and high alkalinity of effluent in the methanogenic reactor to buffer pH and inoculate the land-

fill, thus providing optimal environmental and nutrient conditions for acidogenic bacteria and methanogens, and improve the performance of landfill system [4–6].

However, in the anaerobic bioreactor, continuous hydrolysis of solid waste and the microbial conversion of biodegradable organic content may lead to the production of intermediate volatile fatty acids (VFAs) at high concentrations, which could inhibit methanogenesis owing to the accumulation of VFAs and low pH [7,8]. Some reports have shown that methane production remains inhibited for over 1-year, even in a landfill moistened by leachate recycle [9]. According to the degradation model proposed by Veeken and Hamelers [10], the accumulation of metabolic intermediary products such as volatile acid, not only hinder methanogenesis but also hydrolysis and acidogenesis. Therefore, a balance between acid production and acid consumption is essential for a stable anaerobic process for optimized methanogesis and waste stabilization. Some of the operational techniques such as two-phase process (acidogenic process and methanogenic process) [11–14,22], aeration [12,15–18,28], pH control/buffer addition [19,20] have been presented in recent years.

Compared with conventional process, two-phase process has following advantages: maintains optional environmental conditions for each group of microorganisms and accelerates the conversion of substrate thus increasing stability of MSW by balancing the acidogenesis and methanogenesis. Using of aeration based on the fact that methanogenic bacteria are generally believed to be more sensitive to oxygen and other inhibitors than other

Table 1
Physical and chemical properties of the waste

Components	Value	Components (mg kg^{-1})	Value
Moisture content (%)	28.4	Lead	39.2
Volatile solid (%)	33.2	Cadmium	0.52
Fixed solid (%)	65.5	Cuprum	40.2
Total nitrogen (mg g^{-1})	5.1	Zinc	31.5
Total phosphorus (mg g^{-1})	8.3	Chromium	26.4

Table 2
Physical and chemical properties of the leachate

Components	Concentration (mg L^{-1})	Components	Concentration (µg L^{-1})
pH	6.14	Cd	11.3
Eh(mV)	−283.8	Cr	72.5
COD	12,265.5	Fe	2650.1
BOD$_5$	4783.6	Ni	258.4
NH$_4^+$-N	1161.6	Pb	103.5
NO$_3^-$-N	8.2	Cu	10,343.9
NO$_2^-$-N	0.4	Mn	8031.7
Total nitrogen	709.1	Zn	9345.8
Total alkalinity	7507.5	Al	2030.3
Chloride	1759.3	Conductivity (ms m^{-1})	155.8
Humic acid	1872.0	VFA (mol L^{-1})	114.0

bacteria involved in the methane fermentation, which could achieve two-phase separation and accelerate MSW stabilization. The study carried out by San and Onay [21] showed that a four times per week recirculation strategy with a pH control provided the highest degree of stabilization. Dinamarca studied the influence of pH on the anaerobic digestion of the organic fraction of the urban solid waste in a two-phase anaerobic reactor, and the results showed that the higher degradation of total suspended solid (TSS) and volatile suspended solid (VSS) were obtained in the reactors operated at pH 7 and 8 [22].

The ammonium bicarbonate alkalinity could maintain a pH close to neutral inside cells [23,24]. This was called "metabolism generated alkalinity" inside cells. The degradation of cation releasing nitrogenous organics (proteins) could double the alkalinity concentration generated during biodegradation of proteins in organic solid waste [23,24]. On the other hand, VFAs alkalinity contributes to the buffering of H$_2$CO$_3$, but is transient since the VFAs varies and therefore cannot be consistently relied upon. Therefore, adequate alkalinity, or buffer capacity, is necessary to maintain a stable pH in the digester for optimal biological activity. Alkalinity addition was used in numerous studies to neutralize the pH in the anaerobic treatment of MSW [21,25]. Lens found that the lack of bicarbonate delayed the conversion of organic solid wastes to methane resulting in low methane productions and percentages in control reactor [26]. Alkalinity addition reduced waste quantity, organic content of the solid waste and biodegradation time [24].

Limited studies have been performed to investigate the influences of alkalinity on the anaerobic disposal of the MSW in bioreactors. There are few reports about the impacts of alkalinity on simulated landfill anaerobic reactors treating MSW [24]. The objective of this paper is to study the effect of different types of alkalinity (HCO$_3^-$, CO$_3^{2-}$, and OH$^-$) on stabilization of MSW in simulated anaerobic reactors.

2. Materials and methods

2.1. Sources of wastes and leachate

Samples of MSW and leachate were obtained from Paijia Landfill of Changchun, China. Physical and chemical properties of the samples are shown in detail in Tables 1 and 2. All MSW samples were shredded to less than 1 cm diameter prior to physicochemical analysis and column study.

2.2. Experimental setup

Four simulated landfill reactors were constructed in the laboratory using four Plexiglas columns of length 60 cm and a diameter of 17 cm. Leachate was recirculated in all the four reactors. Reactor A (control reactor) was operated without alkalinization; reactor B, C and D were operated under alkaline conditions, and Na$_2$CO$_3$, NaHCO$_3$ and NaOH were added to leachate, respectively. The reactors were kept at a constant room temperature (25 °C) to enhance the growth of microorganisms. Simulated landfill reactors were

filled with 7.5 kg shredded and synthetic MSW; the density of MSW is 0.934 g/cm^3 approximately.

2.3. Experiment operation

The frequency of leachate recirculation was three times per week for each reactor, and the maximum volume of recirculated leachate did not exceed 1.8 L in order to avoid flooding of the reactor. Throughout the experiment, water was added to the reactors at a constant rate of 600 ml per week in order to simulate the 20 cm annual infiltration.

2.4. Analytical methods

Leachate samples were collected at the bottom of the reactors and analyzed for chemical oxygen demand (COD), biological oxygen demand (BOD$_5$), pH, conductivity, humic acid. All these analyses were performed in accordance with standard methods for the examination of water and wastewaters [12]. Conductivity measured by conductometer (DDS-11A); pH measured by microprocessor pH meter, 211. VFA concentrations in leachate samples were measured using Anderson and Yang method [34].

3. Results and discussion

3.1. pH and VFA

Fig. 1 showed that there was a steady increase in pH for control reactor (reactor A), while there was a sudden rise to a maximum
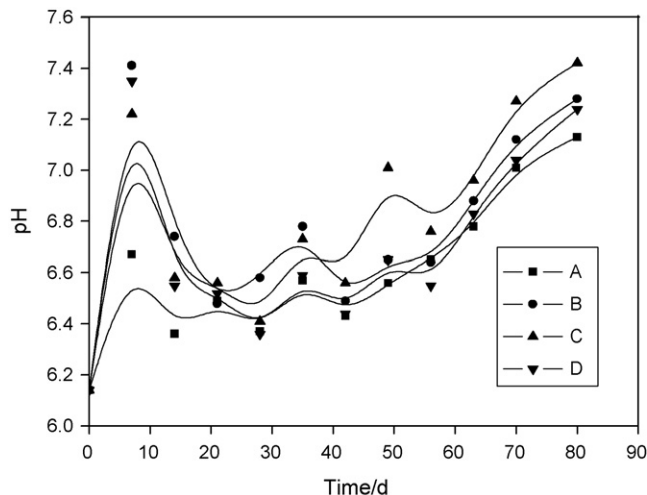

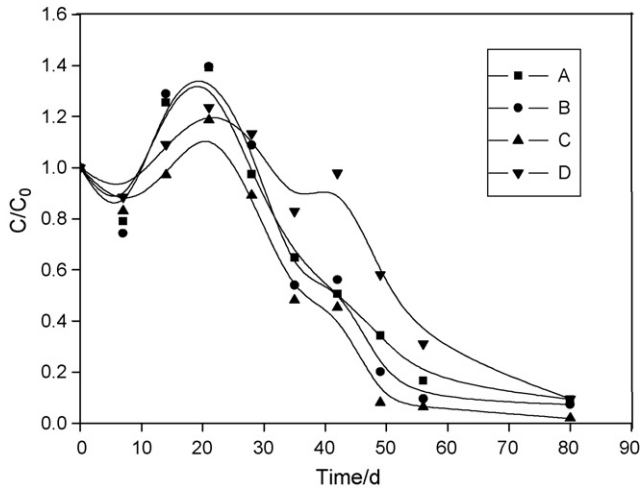**Fig. 1.** Variations of pH over time in reactors.

Fig. 2. Variations of VFA over time in reactors.

Fig. 3. Variations of COD over time in reactors.

of 7.41, 7.22 and 7.35 for reactors B, C and D in 7 d, respectively, this may be attributed to addition of alkalinity. The pH decline shown from the 7 d to 21 d may be attributed to the accumulation of metabolic intermediary products (VFAs, see Fig. 2, and H_2CO_3), the low alkalinity production and the rapid utilization of alkalinity, which is not enough for maintaining the neutral pH and buffering the VFAs produced. With the increase in alkalinity and acceleration of VFAs utilization rate, the pH of reactors began to increase after 28 d. Methanogens can be activated under favorable environmental conditions, which were nearly neutral pH, and the accepted optimum range is approximately 6.5–8.2 in general [23,34]. The final pH was up to 7.13, 7.28, 7.42 and 7.24 in four reactors, respectively. The pH in the control reactor was lower compared with those of alkaline added reactors (reactor B, C and D). The low pH would extend the time required for the stabilization of organic fraction of the waste to stabilize since methane forming anaerobes are known to be very sensitive to low pH [35]. Therefore, the alkalinity addition has positive impact on pH buffering, which could alleviate inhibition of methanogenesis.

The production of intermediate VFAs lies on continuous hydrolysis of solid waste and microbial conversion of biodegradable organic substrate. Fig. 2 showed that the initial concentration of VFAs was 114 mg L^{-1}. The variation trend of VFAs concentration in the landfill leachate for all reactors was similar. From day 0 to day 7, VFAs concentration decreased because of the buffering capacity of MSW. After day 7, VFAs concentration started increasing because the organic fraction of MSW hydrolyzed to intermediate organics and VFAs. Peak concentration was reached after 21 d, after which a decline caused due to VFAs was hydrolyzed and fermented to carbon dioxide and methane in subsequent steps. The final concentration of VFAs was 10.64, 8.4, 2.2 and 10.8 mg L^{-1} in reactor A, B, C and D, respectively.

3.2. COD and BOD_5

Organic matter was released from landfill by the lechate recycle. Leachate COD concentrations in the reactors showed a similar trend to VFAs concentrations (Fig. 3). Due to rapid release and hydrolysis of organics from the MSW into the leachate at the initial stage, the COD concentration in the reactor A, B, C and D increased from 12,265.51 mg L^{-1} to the peak concentration of 15,933.54, 15,701.18, 13,792.47 and 16,987.48 mg L^{-1} in 7, 21, 7 and

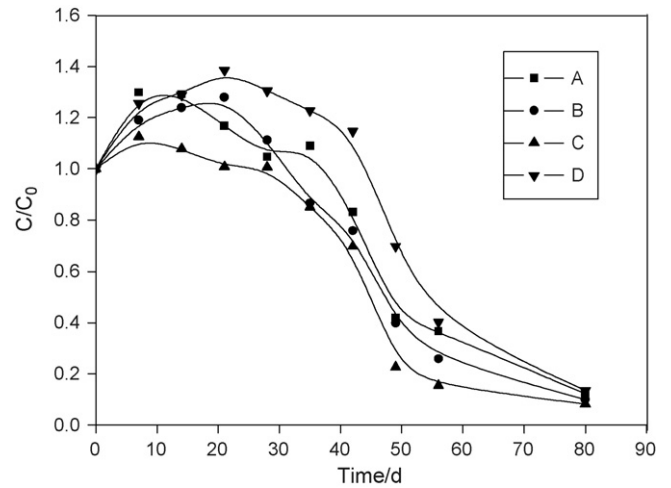21 d, respectively; this was followed by a drop in COD concentration. Concentrations of COD for reactors B and C were lower than that of reactor A, this illustrated that addition of HCO_3^- and CO_3^- have positive effects on anaerobic degradation of MSW, and bacteria is favorable to use HCO_3^- and CO_3^-. Concentration of COD for reactor D was higher than that of reactor A, which indicated that addition of OH^- has little contribution in acceleration of COD degradation. COD removal efficiency for reactors A, B, C and D were 90.56%, 92.21%, 92.74%, 90.29%, while COD degradation rates were 103.48, 125.43, 142.85, 102.74 mg L^{-1} d^{-1}, respectively. Fig. 4 showed that trend of BOD_5 change was similar to that of COD.

BOD_5/COD ratio indicates the amount of biodegradable compounds in the leachate. In the initial stage, BOD_5/COD ratio was increase over time due to BOD_5 released from MSW more than that consumed by microbes; whereas the BOD_5/COD ratio was decreasing. Fig. 5 showed that BOD_5/COD ratio for reactor B, C and D were lower than that of reactor A after 21 d; this indicated that the addition of HCO_3^-, CO_3^{2-}, and OH^- has positive impact on the stabilization of MSW. BOD_5/COD ratio of reactor A, B, C and D was 0.262, 0.104, 0.124 and 0.143 in 80 d, respectively.
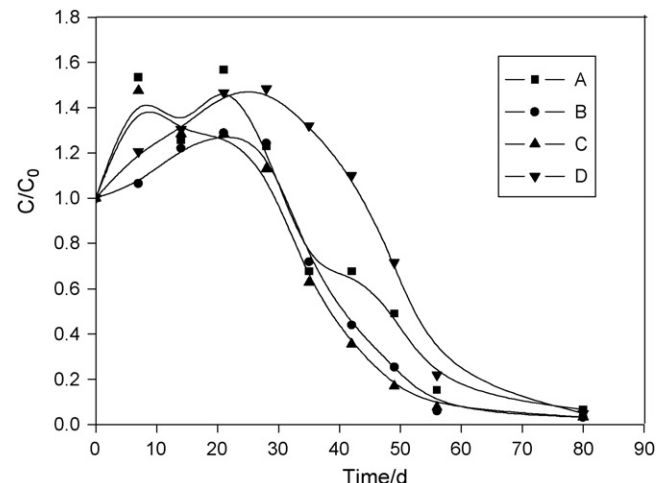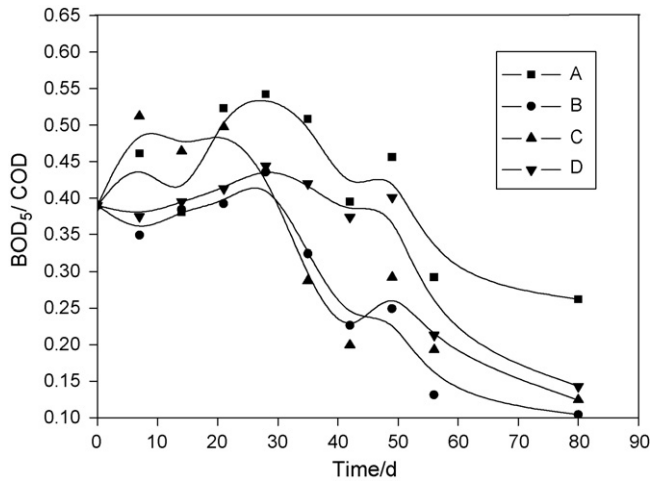

Fig. 4. Variations of BOD_5 over time in reactors.

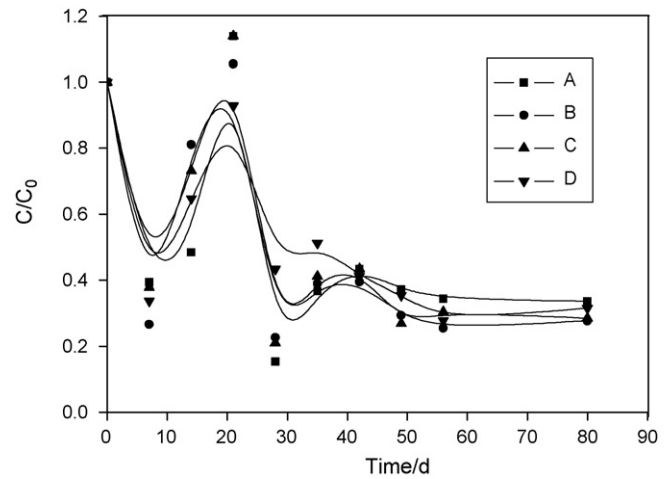**Fig. 5.** Variations of BOD$_5$/COD over time in reactors.

Fig. 7. Variations of NO$_3^-$-N over time in reactors.

3.3. Nitrogen

3.3.1. Ammonium nitrogen (NH$_4^+$-N)

In anaerobic bioreactor, ammonium nitrogen released through degradation of proteinic and nitrogenous compounds. Fig. 6 showed that NH$_4^+$-N concentration of four reactors decreased over time. The decreases of NH$_4^+$-N released from the nitrogenous organic substances could be attributed to ammonium bicarbonate alkalinity, which in turn maintained a pH close to neutral in the anaerobic solid waste bioreactor [23,24,27]. Fig. 6 showed that peak NH$_4^+$-N concentration in reactor A appeared in 7 d; this might be attributed to the leachate recycling which washed out NH$_4^+$-N adsorbed in solid waste, and after the 7 d, a decrease followed. On the contrary, NH$_4^+$-N in reactor B, C, and D decreased in first 7 d, and then increased to peak concentration in 14, 14 and 28 d, respectively, and which followed by a decline. This may be attributed to alkalinity production by ammonium bicarbonate [23,24,27] and the alkalinity addition which maintained a pH close to neutral in the anaerobic solid waste bioreactor. The loss of NH$_4$-N through stripping was considered to be negligible because the pH in the reactors was below 8 [24,29,30]. Though a part of NH$_4^+$-N was consumed by the growth of microorganisms, the NH$_4^+$-N removal was insignificant in the bioreactor landfill system. The final NH$_4$-N removal efficiency of reactor A, B, C and D was up to 65.27%, 56.22%, 60.03%
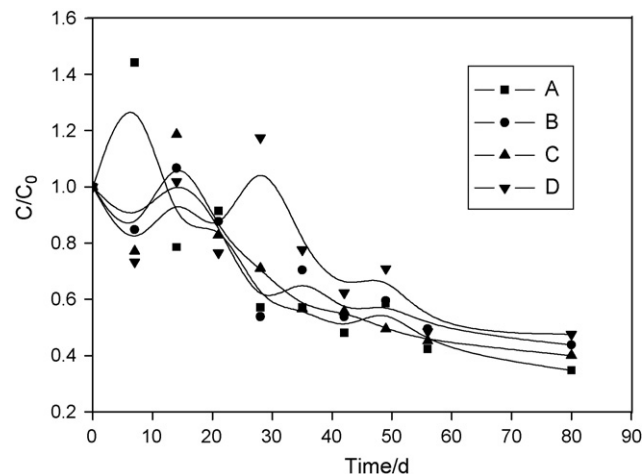
and 52.41%, respectively; so alkalinity addition does not have strong influences on the NH$_4$-N removal.

3.3.2. Nitrate nitrogen (NO$_3^-$-N)

Fig. 7 showed variations of NO$_3^-$-N in the four reactors. Variation trend for NO$_3^-$-N in the four reactors was similar. The drop in concentrations might be the result of the adsorption process in MSW. It was observed that NO$_3^-$-N released from MSW when leachate was recycled attaining a peak concentration of 9.29, 8.60, 9.32 and 7.57 mg L^{-1} in reactor A, B, C and D, respectively. Removal efficiency of reactor A, B, C and D reached 66.45%, 72.38%, 68.62% and 68.44%, respectively. This showed that alkalinity addition have a slight positive influences on NO$_3^-$-N removal.

3.3.3. Nitrite nitrogen (NO$_2^-$-N)

Fig. 8 showed the variation of NO$_2^-$-N concentration over time. The variation trend of NO$_2^-$-N concentration was similar in the 4 reactors. NO$_2^-$-N concentration decrease drastically until the 56 d. The removal efficiency of NO$_2^-$-N in reactors A, B, C and D was 96.5%, 98.75%, 97.75% and 98%, respectively. Though the removal efficiency of 4 reactors for nitrite nitrogen was high, compared with reactor A, addition of alkalinity (reactor B, C, and D) has a slight positive effect on removal of nitrite nitrogen.
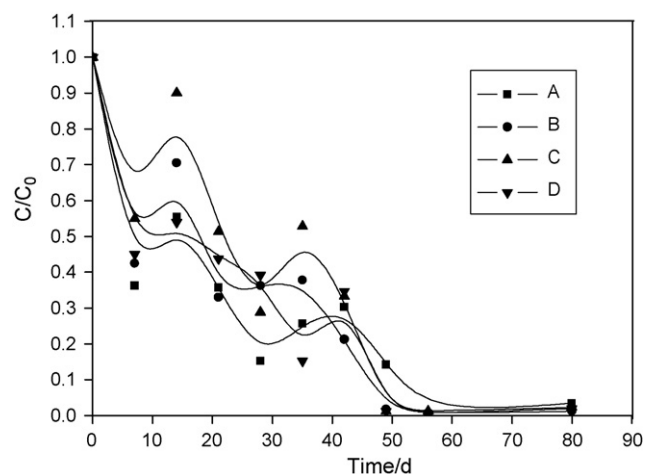

Fig. 6. Variations of NH$_4^+$-N over time in reactors.

Fig. 8. Variations of NO$_2^-$-N over time in reactors.
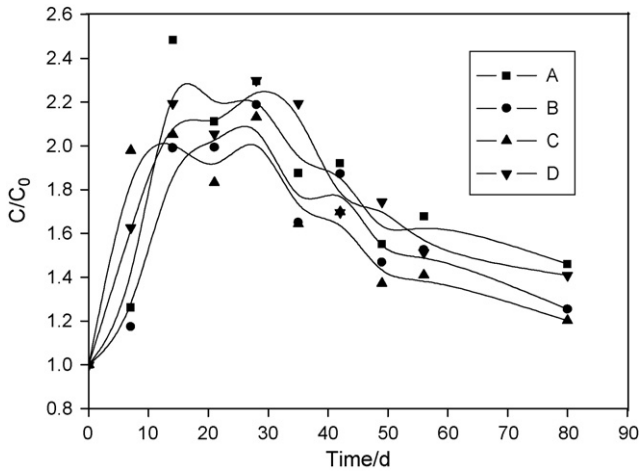
Fig. 9. Variations of TN over time in reactors.

3.3.4. Total nitrogen (TN)

Fig. 9 showed the variation of TN over time. In the initial stage (from 0 to 28 d), TN concentration increased drastically to peak concentrations of 1759.88, 1551.00, 1510.08 and 1603.67 mg L^{-1} for reactors A, B, C and D, respectively. In this stage, the protein in the MSW was converted to nitrogenous and released to leachate. After 28 d, the TN concentration begins to decline which might attributed to the nitrogen removal by biological process [12,3,31] and other attenuation processes, e.g., sulfur compounds [12,32,33]. Fig. 9 showed that the addition of alkalinity has positive influence on the removal of total nitrogen. Furthermore, CO_3^{2-} and HCO_3^- addition have stronger impact on nitrogen removal than OH^- did. The final TN concentration of reactor A, B, C and D were 1035.36, 889.35, 851.37 and 998.64 mg L^{-1}, respectively.

3.4. Total alkalinity (TALK)

Fig. 10 showed the variation of TALK over time. The TALK in the control reactor remained the lowest level in the whole process among four reactors. From 0 to 28 d, the TALK in reactor B (HCO_3^- added) was the highest and followed by that of reactor C (CO_3^{2-} added) and D (OH^- added); after 28 d, the TALK in reactor D was the highest and followed by reactor C and B. This indicated that HCO_3^- could be easily utilized by microbes, and CO_3^{2-} and OH^- followed.
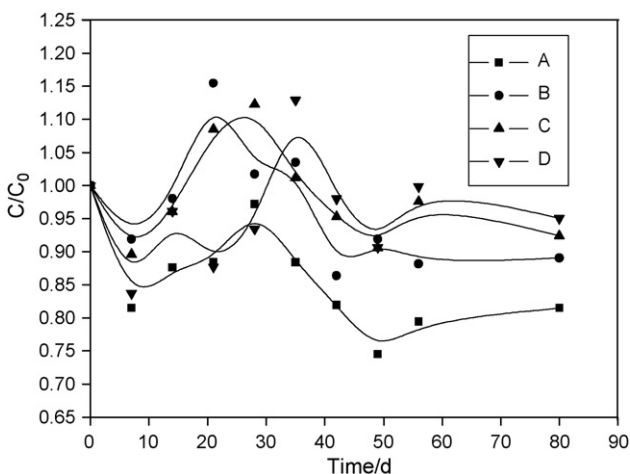
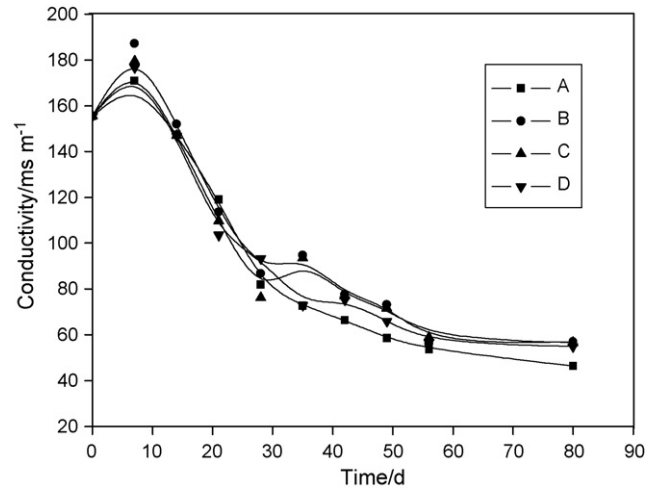

Fig. 10. Variations of TALK over time in reactors.

Fig. 11. Variations of conductivity over time in reactors.

3.5. Conductivity

The conductivity of leachate reflects its total concentration of ionic solutes and is a measure of the solution's ability to convey an electric current. Fig. 11 illustrated that conductivity increased in the first week. The maximum conductivity of reactors A, B, C and D were 170.8, 187.2, 179.4 and 176.8 ms m^{-1}, respectively. This may be attributed to the ions washed out by the leachate recirculation. After the first week, conductivity started decreasing to a final conductivity of 46.4, 57.0, 56.6 and 55.0 ms m^{-1}, for reactors A, B, C and D, respectively. The conductivities of reactors B, C and D were higher than that of reactor A in the whole process; this indicated that addition of alkalinity increased the conductivity.

3.6. Humic acid

Fig. 12 showed that humic acid content in the leachate increase first; this might result from humic acid released from MSW by leachate recirculation. Its maximum content was 3.25, 2.56, 2.73 and 3.13 g L^{-1} for reactor A, B, C and D, respectively. This was then followed by a drastic drop until the 24 d. From the 24 d, humic acid content in reactor A and D remained steady, while in reactor B and C there was a decrease. The final content of humic acid were 0.867, 0114, 0.271 and 0.569 g L^{-1} for reactor A, B, C and D, respectively.
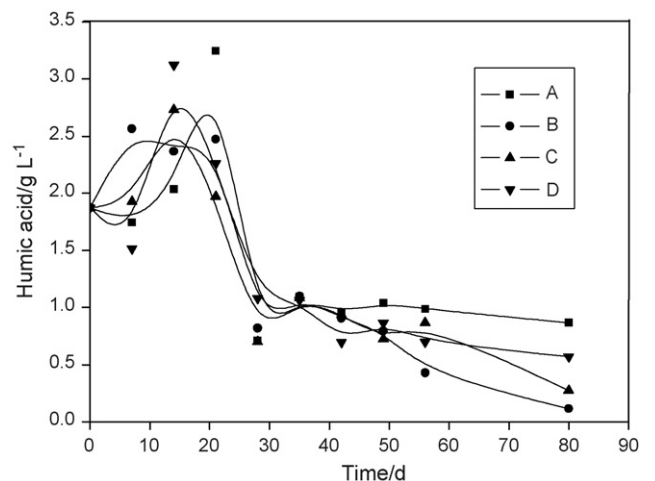

Fig. 12. Variations of humic acid over time in reactors.

This indicated that the addition of alkalinity could accelerate the stabilization of MSW. Moreover, the effect of alkalinity addition was in the order $HCO_3^- > CO_3^{2-} > OH^-$.

4. Conclusions

Alkalinity addition had positive effects on the stabilization of MSW. It could accelerate the removal of leachate pollutants and the MSW stabilization. Alkalinity addition could enhance pH-buffering capacity of MSW, which could alleviate inhibition of methanogenesis. The buffering capacity of the CO_3^{2-} was the highest, HCO_3^- and OH^- followed. HCO_3^- was easily consumed by microorganisms, while OH^- was least.

Alkalinity addition could accelerate degradation rate of pollutants. The final BOD_5/COD was 0.262, 0.104, 0.124 and 0.143; COD degradation rates were 103.48, 125.43, 142.85 and 102.74 $mg\,L^{-1}\,d^{-1}$; the COD removal efficiency reached 90.56%, 92.21%, 92.74% and 90.29% in control, CO_3^{2-}, HCO_3^- and OH^- addition reactor, respectively.

Though alkalinity addition has no clear effects on removal of NH_4^+-N, NO_3^--N and NO_2^--N, it has positive impacts on the transformation of nitrogen and the TN removal.

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