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Removal of volatile fatty acid in landfill leachate by the microwave-hydrothermal method

Lie Yang, Zhulei Chen*, Jinfeng Yang, Yong Liu, Jia Wang, Yingjian Yu, Xiaoming Gao

School of Environmental Science & Engineering, Huazhong University of Science and Technology, Wuhan 430074, P.R. China
Tel./Fax: +86 27 87792151; email: czlgroup@sina.com

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ABSTRACT

In this study, we report on the rising concern with landfill facility is the safe disposal of leachate and high volatile fatty acid (VFA) concentration in leachate provokes the inhibition of biochemical treatment of landfill leachate to the elimination of these serious problems, a microwave-hydrothermal (MH) method has been employed. This new process, named MH method, was developed for the removal of VFA from leachate. The effects of pH, microwave (MW) power, radiation time, and aeration on the removal were investigated. pH and MW radiation time presented significant influence on the removal of VFA. The optimal removal obtained with 625 W power at initial pH $\approx$ 7.2 in 8 min. Aeration presented slight enhancement on the removal. With the optimal operating parameters, the removal of VFA in landfill leachate could reach $\approx$ 30.7% with aeration. The mechanism of VFA removal was proposed as the evaporation of molecular VFA by MW radiation. Compared with heating with an electric oven, MW radiation obtained higher VFA removal. The results shows both thermal and nonthermal effects contributed to the removal of VFA and thermal effects played a more significant role on the removal. It could be proposed that MW radiation could significantly alleviate negative effects of VFA on further treatment of leachate and act as an effective pretreatment method of leachate.

Keywords: Microwave radiation; Volatile fatty acid; Landfill leachate; Thermal effect

1. Introduction

Landfill leachates consist of a complex mixture of organic and inorganic compounds that can be contain toxic and hazardous contaminants [1]. A rising concern with landfill facilities is the collection and safe disposal of leachate. Leachate may contain large amounts of organic matter (biodegradable, but also refractory to biodegradation), where humic-type constituents consist of an important group, as well as ammonia/nitrogen, heavy metals, chlorinated organic and inorganic salts [2]. The removal of organic material based on chemical oxygen demand (COD), biochemical oxygen demand (BOD), and ammonium from leachate are the usual prerequisite before discharging the leachates into natural waters and a large amount of related literatures are found in recent years [2]. However, studies on volatile fatty acid (VFA) removal in leachate were rarely reported. The main components of VFA are formic acid, acetic acid,

*Corresponding author.

propionic acid, butyric acid, valeric acid, etc. It was reported that high VFA concentrations in the system cause the inhibition of the hydrolysis process and even when process pH is optimal, the accumulation of VFA may contribute to a reduced rate of hydrolysis of the solid organic substrate [3]. Borzacconi et al. [4] demonstrated VFA inhibition in acidogenic phase of MSW anaerobic degradation.

Moreover, a problem with research on inhibition of microbial processes by VFA is that a VFA concentration increase results in a pH decrease [5]. Hence, there is an urgent demand for develop an effective pretreatment technology for VFA removal in leachate before biochemical treatment.

In recent years, microwave (MW) radiation has a great deal of attention due to the molecular-level heating, which leads to homogeneous and quick thermal reactions [6]. Researchers have attempted the utilization of MW radiation in wastewater treatment, such as removal of ammonia nitrogen in coke-plant wastewater [7], degradation of remazol golden yellow dye wastewater [8], and boron removal and recovery from concentrated wastewater [9]. Concerning the mechanism of MW radiation, it has been generally assumed thermal and nonthermal effects in MW radiation [10]. Despite the large number of publications, there are still many conflicting results on thermal and nonthermal effects in MW field [7]. Therefore, it is emergent to figure out the removal mechanism of selected pollutants in MW field.

According to our knowledge, there are no reports on the removal of VFA from landfill leachate by MW radiation. As we know, VFA consists of a series of weak acids including formic acid, acetic acid, propionic acid etc. All the compositions of VFA are characterized by volatility.

Therefore, there is great potential application on VFA removal with MW radiation. In this study, we attempt to adopt MW radiation to remove VFA from real landfill leachate. The authors are now initiating some experimental investigations to achieve the following objectives: (1) to optimize the operating conditions for the removal of VFA from leachate by MW radiation; and (2) to explore the removal mechanism of VFA by MW radiation.

2. Experimental details

2.1. Chemicals and landfill leachate

Landfill leachate was obtained from Wuhan Changshankou Landfill, China, which receives domestic solid waste of 2,000 ton per day and acts as the biggest municipal sanitary landfills in the central region of China. Deionized water was used for the preparation of solutions. All other reagents used in this study were above analytical grade (Kermel, 97%). Typical chemical characteristics of landfill leachate are demonstrated in Table 1. All parameters was measured based on the standard method [11].

2.2. Procedures and equipments

The experimental equipment was designed according to Lin et al. [7], and its schematic diagram is demonstrated in Fig. 1. A modified domestic MW oven (800 W, 2,450 MHz, Haier Co., China) with different power setting was used as MW source. A 250 mL glass column was placed in the MW oven. The column was filled with 100 mL of leachate and radiated by MW under different conditions. The initial pH of the solution was adjusted by NaOH (2.5 mol/L) and HCl (2.5 mol/L). The aeration rate was maintained at 0.5 L/min by an air compressor. Temperatures were measured by using a thermometer at the end of trials. The generated contaminant vapor passed through two bottles containing 100 mL of NaOH solution (2.0 mol/L),

Table 1
Typical chemical characteristics of landfill leachate

pH	7.21
SS, mg/L	10,730
CODcr, mg/L	28,200
BOD_5, mg/L	5,270
VFA, mg/L	2,140
NH_4^+-N, mg/L	1,460
Total Cr, mg/L	1.45
Cr^{6+}, mg/L	0.24
Hg, μg/L	1.17
As, mg/L	0.37

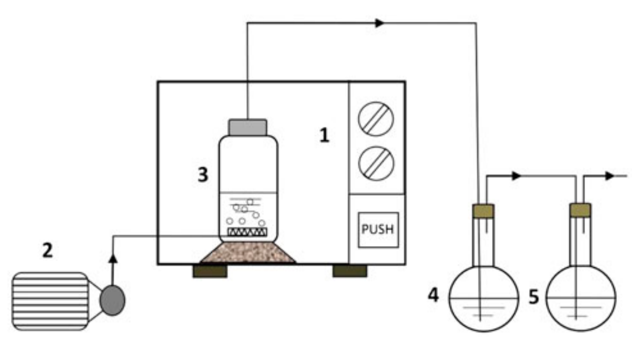

Fig. 1. Schematic diagram of the microwave reactor system. (1) MW oven, (2) air compressor, (3) glass column, (4) and (5) absorption vessel filled with NaOH solution.

respectively. Each experiment was performed in duplicate. An eclectic oven (1,000 W, 50 Hz, Shanghai Jiangren Experimental Equipment Co. Ltd., China) was used to heat leachate as the comparison. Temperatures were measured at the end of trials as mentioned previously. The removal efficiencies of VFA were compared with those obtained by MW radiation. In addition, a built-in thermometer in MW oven was used for temperature measurement.

2.3. Analysis

In the MW radiation process, the volume of leachate significantly decreased with the evaporation of water. When the leachate was cooled to room temperature at the end of experiment, deionized water was added into the column to keep the same initial volume of the leachate. VFA concentration was measured by means of a Varian CP-3800 gas chromatograph using a flame ionization detector and HP-FFAP capillary column (inner diameter of 0.25 mm and length of 25 m). The distribution of weak acids in original leachate was acetic acid (44.9%), propionic acid (25.4%), butyric acid (18.1%), valeric acid (5.2%), and others (6.4%). pH was measured with a pH meter (pHS-25, Shanghai Leici Instrument Factory, China).

3. Results and discussion

3.1. Optimization of operation parameters

In order to achieve the ideal removal of VFA from leachate by MW radiation, the operation conditions were first optimized. Four factors, including initial pH, MW power, MW radiation time, and aeration were investigated.

3.1.1. Effect of pH

Fig. 2 illustrates the removal of VFA at different pH. The optimal pH was found to be the initial value of 7.2, which resulted in 25% VFA removal. At the end of radiation, pH of the leachate increased from 7.2 to 8.1 due to escape of molecular VFA. As mentioned in Section 1, VFA consists of a series of weak acids including formic acid, acetic acid, propionic acid, etc. We can deduce that there is always a pH driving the equilibrium between soluble weak-acid radical ion and dissolved molecular VFA in leachate. In acidic and neutral media, VFA was presented as dissolved molecules. In basic solution, volatile molecules were converted to nonvolatile soluble weak-acid radical ion. Low pH favors VFA volatilization by driving the equilibrium between weak-acid radical ion and dissolved molecules to volatile molecules in leachate.

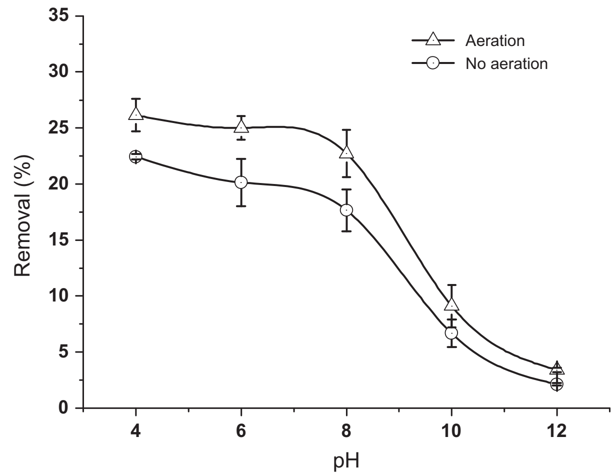

Fig. 2. Influence of pH (500 W MW power, 8 min radiation time) on the removal of VFA by MW with and without aeration.

$$R-COOH \rightleftharpoons R-COO^{-} + H^{+} \quad (1)$$

Fig. 2 shows that the removal of VFA reached 25% at pH 7.2 with aeration. It implied that a certain percentage of the volatile molecular VFA was removed from the solution by MW radiation. No significant increase in VFA removal was observed when solution pH was further decreased. When the wastewater was radiated by MW, the polar molecules in solution rotated fleetly (2,450 million times/s), which resulted in rapid heating of the solution [12]. Consequently, the molecular movement in wastewater was greatly enhanced, which was highly advantageous for the evaporation of volatile molecular VFA from liquid to gas. When the temperature of leachate rose to boiling point, molecular VFA could be stripped from the solution by the gas bubbles produced. The additional aeration was beneficial for the escape of volatile molecular VFA. Besides, MW radiation might reduce the activation energy of reaction system and weaken the intensity of molecular chemical bond [13]. This effect was also advantageous for the removal of VFA. This would be discussed in the later section. Besides, it has been established that there was a good correlation between VFA concentrations and pH values [14].

As we know, there is no report on VFA removal with similar chemical and physical methods. Bories et al. [15] investigated the biological method for the prevention of VFA production at pH 3.5 and achieved to significantly decrease the production of VFA in the winery wastewater discharged into ponds at pH 4.2.

In our experiments, considering the removal efficiency and the minimal dosage of acid added and further biological treatment or the reverse osmosis treatment, we chose initial pH 7.2 as the optimal value.

3.1.2. Effect of MW power

Fig. 3 demonstrates that the removal of VFA increased with radiation power and attained a plateau of 31.82% after 8 min. More heat could be generated with higher radiation power. Generally, the efficiency of MW system increases gradually with increase in MW power and irradiation time [13]. Thus, the solution temperature increased much faster in heating phase and induced more impetuous and rapid molecular motion of large majority of ions polar and clusters [16X1]. This is of great benefits for the escape of VFA from leachate. In addition, aeration contributed to the larger removal of volatile molecular VFA. As mentioned previously, no similar studies about VFA removal with MW was found. Some researchers have applied MW radiation to remove other polar pollutants in wastewater, especially for ammonia [7,17]. Lin et al. [7] investigated the removal of ammonia from wastewater by MW and demonstrated that 750 W was needed to achieve 98.2% removal at pH 11. In Fig. 3, it is observed that the VFA removal had already attained 30.68% after 8 min with 625 W MW power and aeration. Higher MW power from 625 to 800 W slightly increased the VFA removal and was not economical in this study. Hence, 625 W was considered to be the optimal radiation power.

3.1.3. Effect of MW radiation time

The effect of radiation time on the removal of VFA by MW radiation is presented in Fig. 4. It could be seen that the removal increased significantly with increase in radiation time until 8 min. More radiation could be generated with longer MW radiation time. Therefore, the leachate temperature became higher, which induced more impetuous and rapid molecular motion. Besides, aeration contributed to the larger removal of volatile molecular VFA in initial phase before 8 min. And then, the removal increase very slowly with radiation time. Since no other similar studies on VFA were found, we chose reports about MW application on other pollutant removal. Lin et al. [7] demonstrated that 3 min was the optimal radiation time for ammonia removal by MW radiation. Herbert et al. [18] found 60 min was optimal extraction time for the determination of semi-volatile priority pollutants in landfill leachates. Tsai and Lo [9] investigated the removal of boron in concentrated wastewater a MW hydrothermal method and found boron recovery efficiency reached 90% within 10 min. In this study, considering the removal and energy efficiency, we chose 8 min as the optimal radiation time for VFA removal. Moreover, through the MH method has been possible to obtain new phases oxides short time and with low power consumption [19–21].

3.1.4. Effect of aeration

The effect of aeration on the removal of VFA by MW radiation is presented in Figs. 2–4. It has been observed that the removal was enhanced by aeration

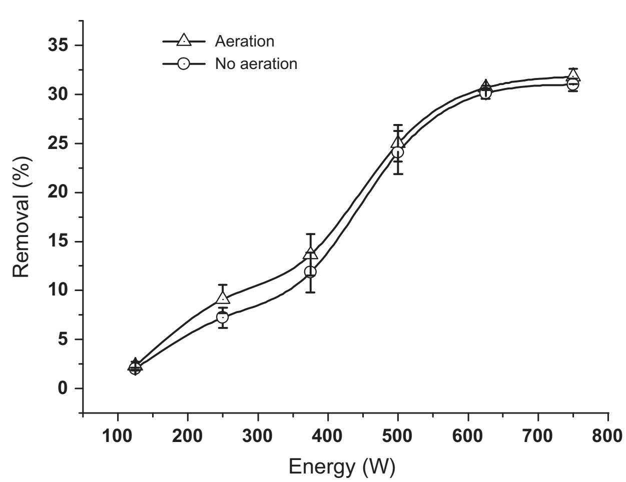

Fig. 3. Influence of MW power (pH 7.21, 8 min radiation time) on the removal of VFA by MW with and without aeration.

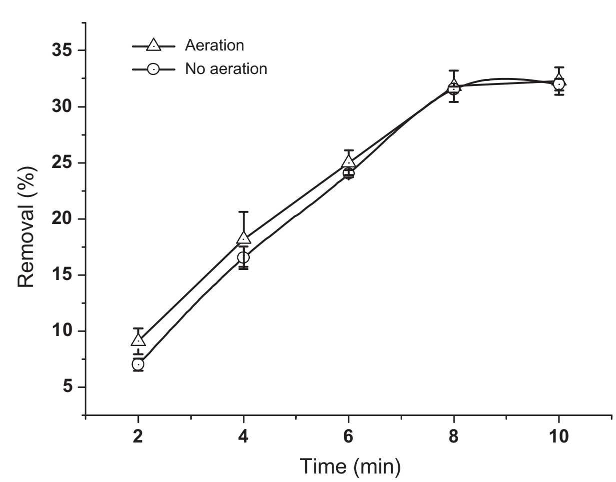

Fig. 4. Influence of MW radiation time (pH 7.21, 625 W radiation power) on the removal of VFA by MW with and without aeration.

to some extent. A lot of air bubbles were brought into the solution by aeration. This might result in turbulence and agitation [22]. Thus, the mass transfer of VFA in leachate was enhanced, which benefited the volatile of molecular VFA. Single aeration method used by us to remove VFA in leachate, the aeration rate was 0.5 L/min, and aeration time was 8 min. We found that 2–4% VFA was removed from leachate. There have been several studies about ammonia removal by using aeration. Kim et al. [23] reported that the ammonia molecule at pH 12 could be air-stripped from solution by fine gas bubbles produced during electrolysis. Because the electrolysis only produced small quantities of gas bubbles, only 9–11% ammonia was removed. Liao et al. [24] attempted removal of nitrogen from swine manure wastewaters by ammonia stripping and significant removal was observed. Similarly, the mechanism could also be used for VFA removal. In the present study, large quantities of gas bubbles were produced by aeration, which attributed to VFA removal.

3.2. Removal mechanism of VFA

3.2.1. Contribution of thermal and nonthermal effects

The removal rates of individual weak acids under the optimal removal parameters are demonstrated in Table 2. All of the main constituents of VFA decreased after MW treatment. Acetic acid had the highest removal probably because of its lower boiling point (117.9℃). The MW effects are mainly classified as thermal or nonthermal effects. Thermal effect is related to the heat generated by the absorption of MW energy by water and other polar molecules, both characterized by a permanent or induced polarization [25]. While none-thermal effect changes the chemical, biochemical, or physical behaviors of systems, while temperature and other parameters remain unaltered [7]. Many studies have been conducted to investigate the nonthermal effect associated with MW (e.g., [26–28]). Ahn et al. [26] demonstrated that MW irradiation increased the solubilization degree of municipal secondary sludge. Li et al. [27] attempted to use MW radiation to enhance reaction between sulfaminic acid and nitrite in a short time, in which nitrite in wastewater was completely converted into nitrogen gas without leaving any sludge and secondary pollutants. However, Shazman et al. [10] and Zhang et al. [13] reported that no nonthermal effects could be found in their works. Critics of the nonthermal effect often claimed that differences of the effect could be attributed to poor temperature measurement and control of experimental conditions that resulted in systematic error. The existence or not of nonthermal effects continued to be an area of considerable debate and research.

Table 2
The removal of individual weak acids in VFA

Constituents of VFA	Percent
Acetic acid	34.4
Propionic acid	28.4
Butyric acid	25.5
Valeric acid	31.1
Other	28.3

In order to figure out whether nonthermal effect was attributable to VFA removal, an electric oven was used to heat the leachate. The leachate was heated from the same initial temperature to the same final temperature by MW radiation and electric oven. The removal of VFA by MW radiation and electric oven was demonstrated in Fig. 5. It could be seen that larger removal of VFA was obtained by MW radiation compared with electric oven. MW energy induced molecular motion by the rotation of dipoles and migration of ions [29]. Leachate consisted of a large amount of molecular H_2O and VFA. Molecular H_2O and VFA were both polar molecules and could easily be polarized by MW radiation. It was reported that MW caused dipoles to rotate and line up rapidly (2,450 million times/s) [30]. It was clear that thermal effect could not lead to the molecular rotation or vibration. The rapid heating and attainment of high temperatures in MW chemistry indicates that most reported rate enhancements could be attributed to simple thermal or kinetic effects. As a result, it could

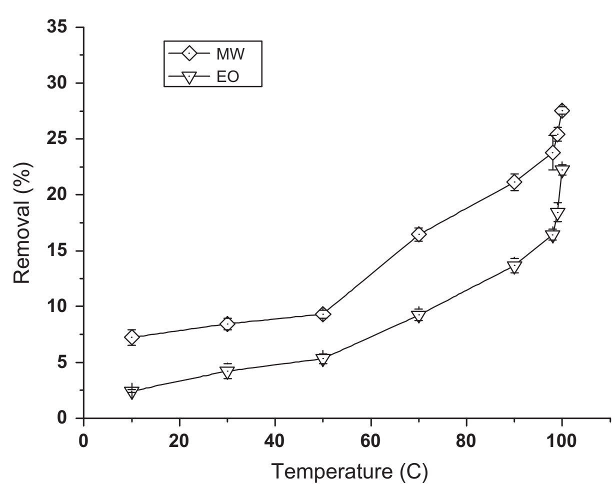

Fig. 5. Comparison of VFA removal by MW heating (625 W) and electric oven (EO) (1,000 W) (pH 7.21 and no aeration).

be concluded that thermal effect played a key role on the removal of VFA by MW radiation, and meanwhile, nonthermal effect increased the removal to some extent. In fact, the removal of VFA reached a plateau of 31.8%, which was still far from complete removal, probably because pH increased in the radiation process due to escape of VFA or part of VFA was difficult to be removed from leachate by MW alone. A complete treatment of more complex wastewater with multiple pollutants or the removal of highly biorefractory pollutant like pentachlorophenol (PCP) is very difficult with MW alone [30]. Further work is still needed to combine MW with oxidants, catalysts or advanced oxidation processes for removal promotion. In spite of above limitations, MW radiation could significantly eliminate negative effects of VFA on further treatment of leachate and act as an effective pretreatment technology of leachate.

3.2.2. Mass balance of VFA

The mass balance of VFA in the MW radiation process is demonstrated in Table 3. The initial mass of VFA in leachate was 214 mg. The VFA removed was completely collected in the bottles filled with NaOH solution. The mass of VFA remained in leachate was also determined. Table 3 demonstrated that there were no increase in both volatile VFA molecules and nonvolatile weak-acid ion in the MW process. It implied that VFA did not convert to other compounds, and no VFA was produced in the MW process. In fact, the VFA in leachate was removed by volatilization from the solution to gas. We believe that the removal mechanism was similar to that of ammonia in the MW process [30], which converted nonvolatile weak-acid ion to volatile VFA molecules in acid or neutral solutions and then stripped the molecule from liquid to gas. The difference was probably that VFA has bigger molecules and is more difficult to be removed from solution.

Table 3
Mass of VFA after the MW radiation

	Radiation time (min)				
	2	4	6	8	10
VFA in leachate	194.76	175.09	161.4	145.11	142.9
VFA in NaOH solution	19.45	39.41	53.67	68.59	69.94
Sum	214.21	214.50	215.07	213.70	212.84

Note: 2,140 mg/L initial VFA in 100 mL solution, 625 W MW power, pH 7.21, 8 min radiation time and with aeration.

4. Conclusions

A fundamental research had been conducted to explore the removal of VFA in landfill leachate by MW radiation. The effects of operating parameters and the mechanism of VFA removal were investigated. MW radiation was effective to remove VFA in leachate. Significant removal was observed in a short time. pH, MW power, and MW radiation time showed notable influence on VFA removal, whereas aeration presented slight effect. Meanwhile, lower pH, higher power, and longer radiation time resulted in higher removal. When the MW power was 625 W, pH was 7.21, and the radiation time was 8 min, the removal of VFA in landfill leachate could reach 30.68% with aeration. Further work is still needed to combine MW with oxidants, catalysts, or advanced oxidation processes for removal promotion. In addition, the mechanism of VFA removal by MW radiation was the volatilization of the molecular VFA (R-COOH), which was produced from weak-acid ion ($R\text{-}COO^-$) at low pH. Both thermal and nonthermal effects contributed to the removal of VFA. Thermal effects played a key role on the removal, and nonthermal effect enhanced the removal to some extent.

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