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Statistical analysis and optimization of ammonia removal from landfill leachate by sequential microwave/aeration process using factorial design and response surface methodology

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ABSTRACT

The application of microwave (MW) radiation followed by aeration (A) for the purpose of ammonia removal from both synthetic solutions and landfill leachate was investigated in this study. 100 mL of synthetic solution or landfill leachate was subjected to MW radiation for 30, 45, 60, 90 and 120 s under 50 and 100% power output level and a pH of 10, 10.5 and 11. The samples were then aerated for 10 min. The initial, after MW application and final total ammonia nitrogen (TAN) were measured. Results confirmed that the sequential microwave/aeration process was an effective approach for removal of ammonia from aqueous systems. Maximum ammonia removal of 81.7% for 100 mL synthetic solution and 70% for 100 mL landfill leachate was achieved by applying 78 KJ microwave energy output and 10 min aeration. Factorial design and response surface methodology were applied to evaluate and optimize the effects of pH, MW energy level and microwave power output. When applying the same energy output to the batch tests, the effect of varying MW power output is negligible. For 100 mL synthetic ammonia solution, the optimum pH and MW energy output level for ammonia removal were 11 and 78 KJ and the maximum ammonia removal efficiency predicted for the synthetic solution is 76.3%. R^2 of 0.941 indicates that the observed results fitted well with the model prediction.

1. Introduction

As one of the major inorganic pollutants in surface water, ammonia exists in aqueous solution in two forms: un-ionized ammonia (NH_3) and ionized ammonia (NH_4^+) [1]. It is common in aquatic chemistry to refer to and express the sum of NH_3 and NH_4^+ as simply ammonia or total ammonia nitrogen (TAN) [2]. Previous research has shown that toxicity is mainly due to NH_3 form [3,4]. The concentration distribution of the ionized and un-ionized ammonia depends on pH, temperature, and total ammonia concentration. Under low pH conditions, the majority of TAN is in the form of NH_4^+, while under high pH conditions, NH_3 becomes the dominant species [1]. Agricultural drainage and wastewaters from steel, fertilizer, petroleum, and meat-processing industries and landfill leachate are the main sources of ammonia in natural water bodies [5]. According to the EPA's fresh water criteria, ammonia is toxic to aquatic organisms even at low levels (1.9 mg/L as TAN at 20 °C and pH of 7) [6]. Also, discharge of wastewater with high ammonia concentration can increase nitrogen levels in natural aqueous systems, thus causing eutrophication [7].

With the increase of population and life quality globally, the generation of municipal solid waste (MSW) has increased rapidly over the past 30 years. EPA data shows that the total MSW generation in the US in 1980 was 151.6 million tons; this increased to 253.7 tons by the year 2005, and remained approximately at the same level until 2012 [8]. The most common MSW management approach is landfilling due to easy operation procedures and low cost. One of the main concerns associated with landfilling process is leachate generation, which is one kind of hazardous and severely polluted wastewater that contains large amounts of organic matters, ammonia nitrogen, heavy metals, chlorinated organic and inorganic salts [9]. Mature landfill leachate is usually characterized by a low BOD/COD ratio as low as 0.04, and high NH_3-N level up to 13,000 mg/L [10]. Ward et al. reported that the significant toxicity of leachate from a landfill in Florida was due to the presence of ammonia [11].

Traditional biological processes for treatment of ammonia incorporate nitrification and denitrification and they do not perform well to high concentration of ammonia in landfill leachate, which could inhibit microbial activities including the nitrification

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process. The inhibition of the nitrification process is reported to be due to NH_3 form while the presence of NH_4-N in the aqueous solution is reported to inhibit the NO_2-N oxidation process at concentrations of 20 mg/L, and also begins to inhibit the NH_4-N oxidation process at concentrations of 100 mg/L of [NH_4-N] [5]. Ammonia inhibition effects on microorganisms in biological treatments have also been reported in the range of 1500–5000 mg/L as TAN by other researchers [12,13].

For wastewaters with high ammonia concentration, physical and chemical methods have been reported to exhibit high removal efficiencies. However, physical and chemical methods usually come with high cost and some other issues such as the large volume of contaminated sludge generated from the flocculation and coagulation approach, as well as the long contact time during the stripping process [14,15].

Considering cost and removal efficiency, the application of chemical/physical methods as a pretreatment for the high ammonia concentration before biological processes could be an effective option. The biological processes require a proper C:N ratio around 20:1 for the aerobic treatment and 40:1 for the anaerobic process. Since high ammonia levels in the leachate usually result in a low C:N ratio in the system, the pretreatment could reduce the ammonia concentration while still retaining enough carbon sources for the biological process [16].

Microwave (MW) radiation has been widely applied in the environmental field for the sludge and wastewater treatments [17–20]. MW enhanced oxidation processes with oxidants such as hydrogen peroxide (H_2O_2) and persulfate ($S_2O_8^{2-}$) increased the degradation rate of COD and some micro-pollutants such as acid orange 7 (AO7), bromophenol blue, phenol and pharmaceutical wastewater [21].

MW radiation under high pH levels was reported to reduce ammonia significantly without affecting the organic compounds [22]. It was observed that, when applying 350 W microwave radiation, the MW radiation process (below the boiling point) was responsible for 40% of the ammonia removal. The remaining portion of the dissipated ammonia was removed in the following two minutes after the solution reached the boiling point, by a combination of MW and thermal processes. Meanwhile, when aeration process was applied concurrently with MW radiation process, the contribution of aeration decreased with longer radiation time. However, the continuous boiling process employed in the research by Lin et al. [22] could be a safety concern due to the high operation temperatures and may possibly generate unknown secondary pollutants when applied to real landfill leachate. Moreover, when MW radiation has been used as a pretreatment approach, the high effluent temperature can have a negative effect on the subsequent biological treatment processes.

Aeration is another effective physical treatment for the removal of ammonia without affecting carbon sources in the aqueous system. The main drawback of the aeration process is the energy expenditure, the need to raise pH and the contact time required [23]. Mattinen et al. reported 89% ammonia reduction at pH 11 with 24 h contact time [24]. In a similar research, the maximum ammonia removal efficiency was found at 85% for a landfill leachate with approximately 1000 mg/L ammonium nitrogen, at pH of 12 with 17 h contact time [14].

In this study, a sequential MW radiation followed by aeration process was systematically applied for ammonia removal from aqueous phase (synthetic solution) under different MW power output, pH and radiation time based on a factorial experimental design. Response surface methodology (RSM) was also applied to evaluate and optimize the effect of pH, MW power output, and radiation time. For both safety and economic concerns, the samples were maintained below the boiling point. The temperature of the samples was significantly increased during the MW radiation stage, and then decreased through subsequent aeration process. And lastly, the MW treatments with and without aeration processes were applied for the ammonia removal from a real landfill leachate.

2. Material and methods

2.1. Materials and equipment

The synthetic solution which contained 2700 mg/L total TAN was prepared by dissolving analytical grade ammonia chloride (Fisher Scientific) in distilled water. As mentioned above, ammonia inhibitory effects in biological treatments in the range of 1500–5000 mg/L as TAN have been reported [12,13]. As such, it was decided to select an initial concentration of 2700 mg/L total TAN which is within the inhibitory range. In each test, 100 mL of the fresh synthetic solution were prepared. The initial pH was around 5.7. The pH was adjusted to the desired value using 10 mol/L NaOH. The landfill leachate was obtained from a local landfill in Ottawa with the initial pH 9.0 ± 0.2 and TAN concentration of about 4000 mg/L.

The MW process was carried out by a Panasonic microwave oven (Model NN-S 750) with an operating frequency of 2450 MHz and power consumption of 1320 W. The maximum power output was 1300 W and could be adjusted from 10% to 100% in 10% intervals.

The aeration process was performed using a Model 200 MARINA pump with a total aeration rate of 110 L/h with 2 air diffusers. In each test, only one diffuser was placed in the batch reactor. For the tests carried out with the synthetic solution, a 150 mL beaker was used as the batch reactor for all MW with and without aeration processes. For the landfill leachate, a 1 L beaker was used as the batch reactor for the microwave process and a 4 L glass container was used for the aeration process.

2.2. Analytical methods

TNT 832 ammonia vials from HACH Company were used to test ammonia concentrations based on the Salicylate Method using a HACH DR5000 Spectrophotometer. The 10 mg/L ammonia nitrogen standard solution from HACH Company was used to check the spectrophotometer. The pH was measured using a glass electrode in combination with a Fisher Accumet® Model XL25 dual channel pH/ion meter.

2.3. Experimental design

For this study MW and aeration were performed in sequence. Experimental conditions for the tests conducted are summarized in Table 1. The pH range was selected based on the results of a preliminary set of experiments, which was conducted to evaluate the effect of pH on process efficiency. At a 100% power, MW radiation times greater than 90 s resulted in boiling of the samples

Table 1
Experimental design of MW processes for the synthetic solution.

MW time (s)		30		45		60		90		120	
Power output level (%)		50	100	50	100	50	100	50	100	50	100
pH	10	✓	✓	–	✓	✓	✓	✓	–	✓	–
	10.5	✓	✓	–	✓	✓	✓	✓	–	✓	–
	11	✓	✓	–	✓	✓	✓	✓	–	✓	–

✓ Experiments conducted under these conditions.
– No experiments conducted under these conditions.