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Ammonia removal using 1.7 MHz high frequency ultrasound in batch and novel dam–weir falling systems

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ABSTRACT

In the present work, an experimental study was carried out to investigate the ammonia removal using high frequency ultrasound in two sonoreactors, a batch and a new proposed dam–weir falling reactor. Both of the reactors equipped with high frequency (1.7 MHz) piezoelectric transducers to induce ultrasound waves into the solution. The effects of initial concentration and ultrasound irradiation on ammonia removal with and without aeration were studied. A calorimetric study was done and it was found that 4 cm liquid height on the surface of piezoelectric is an optimum height for ultrasound irritation. Moreover, it was found that aeration has a significant influence on ammonia removal. It is found that after 120 min ultrasound irradiation in the presence of air, ammonia removal up to 80 and 49% can be obtained in the dam–weir and batch systems, respectively.

Keywords: Atomization; Ultrasonic irradiations; Ammonia removal; High frequency ultrasound wave

1. Introduction

Nowadays, amount of ammonia in domestic, agricultural, and industrial wastewaters has been increased significantly. Ammonia release in the environment can cause serious and irreparable damages, such as contaminates surface and underground water resources, prevent the growth of plants, toxic to the most fish species, emits a nasty smell, and is carcinogenic. So, the most important negative impact will be on the human health [1–5]. Regarding to the above reasons, removal of ammonia from wastewater become a vital and necessary issue. Therefore, in order to reduce the risk of contamination, an effective treatment method should be employed. To date, in order to remove ammonia, numerous beneficial physicochemical and biological treatment methods have been reported. Examples on various common treatment processes are: biological nitrification [6–9], advanced oxidation processes [10,11], air stripping [5,12,13], ion exchange [14–19], chemical precipitation [20,21], membrane separation processes [22–25]. Each of these techniques has some limitations and defects. For example, the drawbacks of ion exchange and chlorination processes are their high cost and requirement to chemicals which leads to hard

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maintenance. In addition, a biological treatment method cannot be suitable for high ammonia concentration. On the other hand, this approach is limited by undesirable environmental issues and slow bioconversion [22,26]. The effectiveness of air stripping and membrane separation processes is restricted by high pH values and fouling, respectively [4,5].

Compared with conventional treatment methods, ultrasound provides considerable advantages from yield and performance points of view. The principal benefit of using ultrasound is no need for extra chemicals. Therefore, ultrasound prevents the release of excess toxic compounds into the environment and diminishes the environmental associated concerns. Moreover, the operating cost will be reduced. This technique is not only appropriate from the economical viewpoint, but also it is easy to apply [26–29]. Studies presented in literature showed that ultrasound techniques have been widely used in biotechnology, chemical synthesis, polymer chemistry [30,31], catalysis [32,33], emulsification [34,35], atomization [36,37], crystallization [38,39], extraction [40,41], cleaning [32,42], etc. Because of the above-mentioned advantages, ultrasound has attracted considerable attention as a novel technique for treatment of variety of contamination in water and wastewater over the past few years [32]. The effect of ultrasound waves on the wastewater causes the acoustic cavitation phenomenon. This phenomenon includes formation, growth, and collapse of bubbles accompanied by the production of high localized temperatures and pressures, highly reactive radicals ($H^{\bullet}$, $HO_2^{\bullet}$ and $OH^{\bullet}$), and hydrogen peroxide (H_2O_2). These hot spots have a temperature of about 5,000 K and a pressure of about 100 MPa, which lead to breakdown of volatile substance and molecules present inside the cavitation bubbles. The reactive radicals are generated by sonolysis of water molecules as solvent within the collapsing cavitation bubbles. The generated radicals are also able to destroy organic contaminations by facilitating chemical reactions. In addition, hydrogen peroxide can be formed by recombination of primary radicals of sonolysis [43–49]. Acoustic streaming is also detected during sonication of wastewater in the range of MHz. High frequency ultrasound wave propagation through wastewater causes local turbulence and circulation flow besides the cavitation effects. This leads to provide macro- and micro-mixing, which was realized as a main reason of uniform distribution of ultrasound energy. Consequently, the efficiency of the heat and mass transfer processes within wastewater will be enhanced remarkably [50,51]. Li et al. [52] used a combination of power ultrasound radiation and hydrogen peroxide for degradation of organic matter and ammonia nitrogen in the studied wastewater. They demonstrated that initial high pH value enhance efficiency of COD reduction and ammonia removal.

Chen and Huang [53], studied degradation of dinitrotoluenes and trinitrotoluene in the wastewater by using 20 kHz ultrasound coupled with TiO_2. They revealed that ultrasonic irradiation is a beneficial technique for direct treatment of nitrotoluenes wastewater. The influence of ultrasound waves on degradation of Acid Blue 25 (AB25) in aqueous solutions was studied by Ghodbane and Hamdaoudi [54]. According to their study, combination of high frequency irradiation (1.7 MHz) with Fe(II) or H_2O_2 could be promising for the degradation of the above-mentioned pollutants. Matouq and Al-Anber [55], used 1.7 and 2.4 MHz frequencies of ultrasound to degrade ammonia from solution with three different concentrations. They found that ultrasound device waves can degrade ammonia effectively at the lowest concentration. In other research, the application of ultrasonic wave irradiation to remove some hydrophobic polyaromatic hydrocarbons (PAHs) from petrochemical industry wastewater was examined by Sponza and Ozetekin [28]. According to their results, ultrasonic irradiations can be effective for removing hydrophobic PAHs from wastewaters. In another study, Matouq et al. [56] applied 1.7 MHz ultrasound wave for removing diazinon pesticide from water. They reported that by increasing solution volume, diazinon removal reduced. Moreover, the ability of diazinon removal was decreased by increasing the diazinon concentration. In addition, Li et al. [57] used a combined 20 kHz ultrasound and Fenton process to treat wastewater. They claimed that US-Fenton process is the most useful for treatment of polluted wastewater in comparison with photolysis (UV) and Fenton techniques.

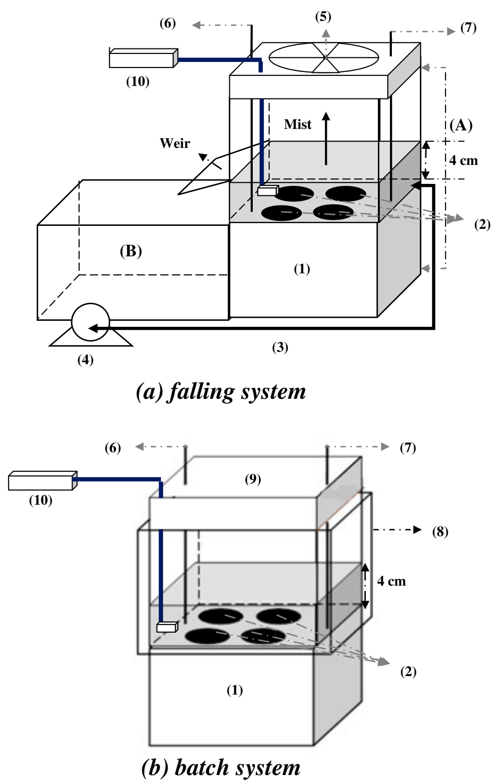
Many researches deals with, using the falling film system for enhancing evaporation [58,59], absorption [60,61], freeze concentration [62,63], etc. The main issue in using falling film is to increase the mass transfer (or heat transfer) due to short diffusion distance in this type of film. Here, this idea was used to give more opportunities for escaping and stripping of ammonia molecules from treated solution. In the other hand, aeration, as a well-known technique, was used for different purposes in the literature [64–67]. In aeration, air bubbles are injected into liquid. According to the previous experience in the reported studies [46,52], by coupling of aeration and sonication, the cavitation bubbles are broken into smaller bubbles. This can cause a better stripping and pyrolysis. Therefore, in this study, aeration was coupled with sonication to enhance the ammonia removal. Between studies in the field of wastewater treatment, there are only a limited number of research in which high frequency sonication in the range of MHz were used [55,56].

The novelty of the present study is the removal of ammonia using high frequency ultrasound waves (1.7 MHz) combined with air aeration. For this purpose, two systems were designed; a dam–weir falling and batch systems, both equipped with piezoelectric transducers. In the first stage, the optimal height of solution on the piezoelectric surface was determined by calorimetric study. Then, the effect of various operating conditions such as initial ammonia concentration, sonication, aeration, and combination of them on removal efficiency of ammonia was examined.

2. Experimental work

2.1. Experimental setup

In this work, in order to remove ammonia from solution by ultrasound waves, two experimental setups (batch and semi-falling systems) were designed. The schematic views of the experimental setups are shown in Fig. 1. The falling system included two reservoirs; upper reservoir A with a length of 15 cm, width of 11, height of 12 cm; and lower reservoir B with a length of 15 cm, width of 12, and height of 14 cm. Reservoir A was equipped with four piezoelectric transducers installed at the vessel bottom in a way that the fluid to be in direct contact with them. The solution, after passing through the surface of the piezoelectric transducers and weir in reservoir A collected in reservoir B and circulated back to reservoir A by a circulation pump. The upper side of the setup is open to the air, and a small electrical fan installed on it to draw off all mist produced during experiments. The batch system includes a reservoir with the specification of the reservoir A in the falling system. The reservoir at the upper side was kept close in all experiments. The temperature of the solution was maintained constant at 27 ± 2°C with a cold bath in this setup. The piezoelectric transducer with a diameter of 1.5 cm and a frequency of 1.7 MHz with input power of 9.5 W (Model ANN-2517GRL, Annon Piezo Technology Co. Ltd, China) was placed at the bottom of the reservoir A. In order to measure the solution pH, the pH meter probe (827 pH Lab Metrohm) was immersed into the liquid solution. The temperature of the solution was measured using a thermometer (Lutron, BTM-4208SD) before and after the experiment. For aeration, air was supplied by an air compressor.



(a) falling system

(b) batch system

Fig. 1. Schematic diagram of experimental setup, (1) ultrasonic inducer circuit, (2) piezoelectric transducers, (3) recycle, (4) pump, (5) fan, (6) digital thermometer, (7) digital pH meter, (8) cold bath, (9) cover, (10) aeration, (a) falling system, (b) batch system.

2.2. Materials and methods

Ammonium solution with a purity of %25 and Nessler's reagent were provided by Merck Inc. In order to prepare solutions containing 400, 600, 800, and 1,200 ppm of ammonium ion, the deionized water was used. In falling system, reservoir B was filled with a specified concentration of ammonia in 1,500 mL of sample solutions and then solutions were pumped into the reservoir A. This was done to create circulating stream leads to better mixing and enhance contact surface with ultrasound waves generated by piezoelectric transducers.

The batch system was filled with a 580 mL ammonia solution at specified concentration and irradiated with ultrasound waves. A solution pH was measured every 20 min and the concentrations of ammonia were determined using Nessler's reagent. In this method, the ammonia reacts with Nessler's reagent to form a colored complex that can change from yellow to deep amber. The ammonia absorption was measured using a UV-spectrophotometer manufactured by the UNIC

Company at 440 nm [68]. Changes in ammonia solution concentrations were evaluated during 2 h.

3. Results and discussion

In the present study, the effect of initial concentration of solution, sonication, and aeration on ammonia removal efficiency have been investigated.

3.1. Energy efficiency studies

In the first stage, in order to find the optimum operating volume of system, which is condition with maximum power density, the amount of actual energy dissipated in the bulk liquid was determined by recording the liquid temperature. Then, the energy efficiency, which is available energy for the cavitation phenomena, was estimated using Eq. (1) [69]:

$$\text{Energy efficiency}\,(\%) = \frac{m \times C_p \Delta T}{E \times t} \times 100 \quad (1)$$

In which, m is mass of liquid (kg), C_p is specific heat capacity (J/kg K), ΔT is rise in temperature of water (K), and E is electric energy supplied (W) for the time interval of t (s).

The ultrasound power density was obtained by dividing the calorimetry power, p (W), to sample volume, V (L) as follows:

$$\text{Power density} = \frac{p}{V} \quad (2)$$

The characterization of the optimum operating capacity, where the power density is the maximum, is quite important. It is strongly dependent on the amount of power density available for the cavitation phenomena [70,71]. Moreover, the experimental result of ultrasonic atomization using ammonia–water solution depend on the ammonia concentration and capacity used [55]. Five different liquid volumes of 0.58, 0.825, 0.99, 1.155, and 1.320 L (these are exactly equivalent to 4, 5, 6, 7, and 8 cm height) were chosen. No mist was observed with using more volume of sample (more than 8 cm height).

Energy efficiency was estimated for various amounts of water in constant power input of 38 W. The obtained results are given in Fig. 2.

The results show that as the reaction volume increases, energy efficiency in the bulk of liquid increases and then decreases. Fig. 3 illustrates the variation of power density with the operating volume. This figure indicates that the use of about 990 mL of solution (6 cm layer) shows almost the same results of power density as 580 mL. When 990 mL liquid solution was used, it was observed that little atomization occurred in comparison with 580 mL liquid solution. This may be due to the fact that the capacity of the transducer for producing energy to evaporate ammonia will be less when the liquid height is higher. This phenomenon was discussed clearly by Kawase et al. [72]. They reported that there is a strong relation between the liquid height and the consumed power

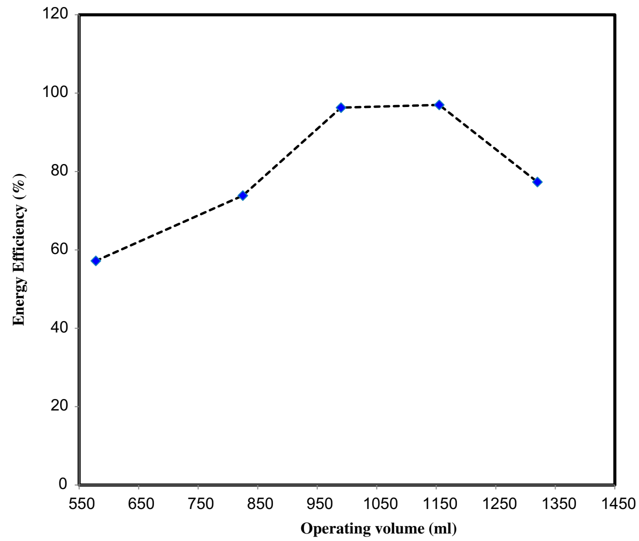


Fig. 2. Variation of energy efficiency with different operating volume at constant supplied power of 38 W and constant 1.7 MHz frequency of irradiation.

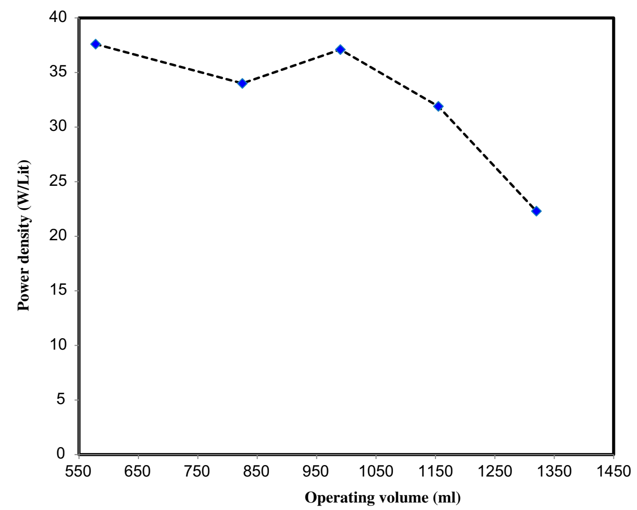


Fig. 3. Variation of power density with different operating volume at constant supplied power of 38 W and constant frequency of irradiation at 1.7 MHz.

for producing mist. Thus, it is better to use 580 mL, which is equivalent to 4 cm height liquid, as the operating capacity and hence, further experiments were done using this optimum operating capacity. The obtained power density of piezoelectric transducers was 37.6% for 580 mL of sample.

3.2. The effect of initial concentration

The effect of initial ammonia concentration on the ammonia removal in the two above-mentioned systems are investigated. Four different initial concentrations of ammonia solution, including 400, 600, 800, 1,200 ppm, were chosen. The removal ratio is defined as follows (Eq. (2)):

$$\text{Removal}\,(\%) = \frac{C_{Ao} - C_{Af}}{C_{Ao}} \times 100 \quad (3)$$

where C_{Ao} is the initial concentration of ammonia (mg/L) and C_{Af} is the final concentration of ammonia (mg/L) after applying ultrasound. The measurements were done during 2 h.

Fig. 4 reveals the relationship between ammonia removals at various initial concentrations for both batch and falling dam–weir layouts. In the first part, experiments were carried out in the batch system. The optimum operating height, 580 mL, which is equivalent to 4 cm height inside the reservoir, was used. The results presented in this figure show that using high frequency ultrasound wave had a significant effect on ammonia removal at various concentrations. This can be due to the pyrolysis reaction (thermal decomposition) in cavitation bubbles or reaction at nearby interfacial region. In addition, from this figure one can find that with increase in ammonia concentration, the degradation capability decreased. The effect of ultrasound waves decreases with increasing of initial ammonia concentration and the best removal of ammonia was obtained at 400 ppm sample concentration. This might indicate that with increase in ammonia concentration, there are more ammonia molecules in the solution, so that the number of ammonia molecules entering cavitation bubbles increase, and diminishes the transient high temperature inside the cavitation bubble. Thus, the removal rate of the ammonia from the solution by ultrasound was decreased [73].

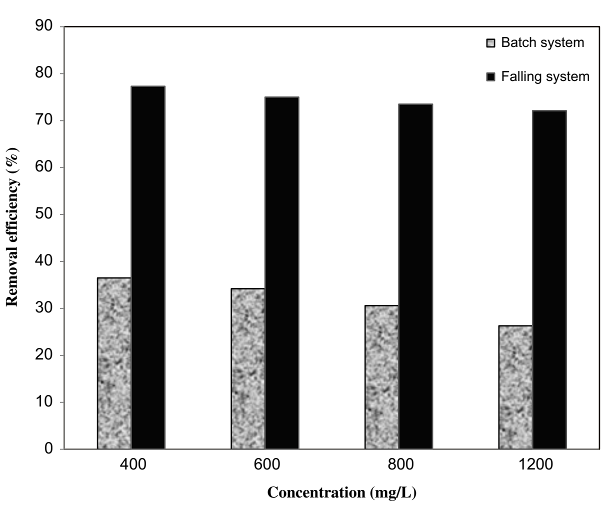


Fig. 4. Comparison between removal efficiency in the batch and falling systems.

Turning to falling system, in order to use the most capacity of 1.7 MHz transducer for producing energy to atomize ammonia, 4 cm was considered for weir height. This is the height of liquid on the surface of the piezoelectric transducers. The value of sample solutions in falling system is 1,500 mL. As illustrated in Fig. 4, the results show that the ability of ultrasound waves in the removal of ammonia in this setup is significantly more than that of the closed batch setup. The obtained removal efficiency for the falling system is nearly 50% more than that of the batch system in the different initial concentrations. It is noticed that in this new setup, besides thermal decomposition due to cavitation bubbles explosion, the ultrasonic atomization is quite important. The ultrasonic atomization is a process, which change the liquid to small droplets such as a mist in the gas phase [49]. Therefore, using weir in the falling system, which can contribute to create a thin liquid film, improves performance of ultrasonic atomization and as a result ammonia stripping. In addition, fluid recycling can enhance macro- and micro-mixing in the system, which helps stripping. It is evident from the results of this new layout that the main mechanism for ammonia removal by ultrasound waves is ammonia ultrasonic atomization and pyrolysis in cavitation bubbles is the secondary mechanism [55,56]. As far as in the batch layout ammonia removed by pyrolysis, the difference between the ammonia removal efficiency of falling and batch layouts can be related to ultrasonic atomization.

Fig. 5 shows the concentration profiles changing with time at various initial ammonia concentrations of both systems. It is obvious that increase in the ammonia removal rate is related to time of exposing to ultrasound waves in different initial ammonia concentration. According to this figure, it can be noticed that the more efficient sonication was obtained when the initial ammonia concentration was 400 ppm. The pH of this concentrated solution in batch and fall-

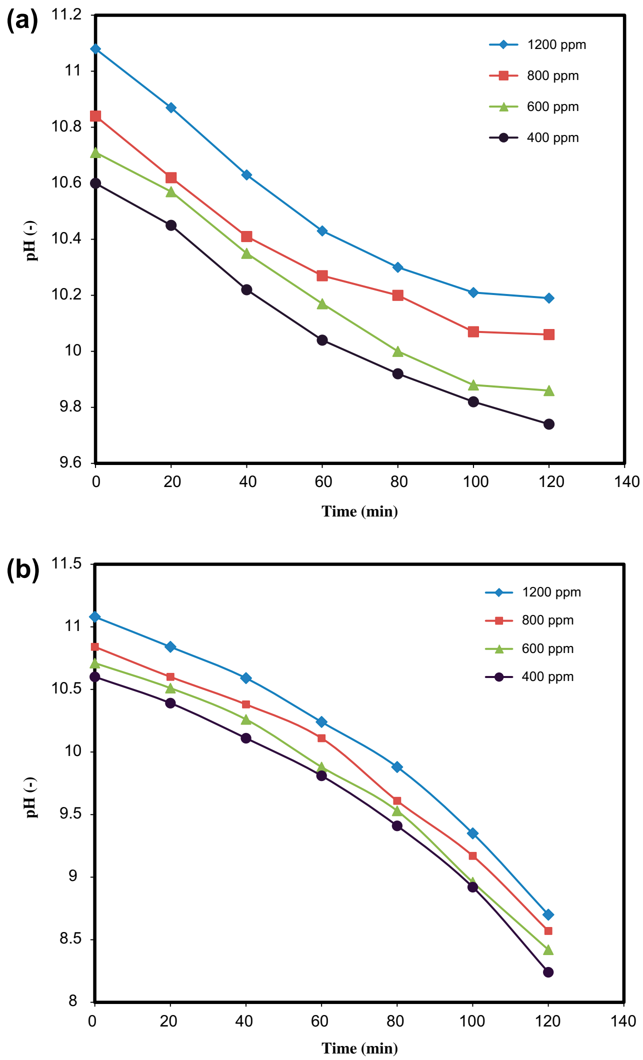


Fig. 5. Concentration profiles for ammonia removal at different concentration of solution: (a) batch system, (b) falling system.

ing systems decreases to 9.74 and 8.24 for batch and falling systems, respectively.

Finally, a comparison between removal efficiency ammonia by ultrasonic technique in the both systems and energy consumption, ε (W/kg), reported in Table 1. It was found that, with lower energy consumption, the obtained removal efficiency for falling system is approximately 40% more than batch system, which means the falling dam–weir type sonoreactor has outstanding performance. In the falling system, the solution circulated back into the vessel by a circulation pump. This application can promote the oxygen transfer on the liquid phase and thereby increase the cavitation phenomenon. Therefore, It can be argued that falling system uses both mechanisms of ammonia removal, including; decompositions with pyrolysis reaction (thermal decomposition) and evaporation of ammonia by atomization.

3.3. The effect of aeration

In the present work, in both setups, the influence of aeration on the ammonia removal efficiency was studied. The removal efficiency of ammonia was investigated in the presence of air with and without ultrasonic irradiation and the combination of them. In Fig. 6, the ammonia removal performances as a function of irradiation time in the batch system are illustrated. The results indicate that the removal efficiency in the presence of air with ultrasound irradiation was higher than other conditions. Therefore, it can be concluded that the presence of air into the systems can enhance the ammonia removal with increasing number of bubbles and accelerating cavitation and pyrolysis in the solution. According to previous experience in the reported studies [55], in presence of air, the cavitation bubbles are broken into smaller bubbles with very small diameter about of 0.2–0.3 μm. This can increase the bubbles surface area to more than 103–104 times of system without aeration. Therefore, the interfacial area between water and air was enhanced, and ammonia was removed better. The figure shows that after 60 min, for both layouts, with and without aeration, the slope of ammonia removal performances decreased. The main reason may include the collapse change of micro-bubbles in cavitation and the diminution of cavitation efficiency by continues ultrasound propagation, which leads to formation of degassing bubbles. These degassing bubbles are ineffective for sonolysis reaction, due to which they prevent the propagation of sound by absorption and spreading of

Table 1
Comparison between removal efficiency ammonia related to energy consumption (C_{NH3} = 400 ppm)

Setup	Removal (%) ultrasound	Removal (%) ultrasound + aeration	ε (W/kg)
Falling system	77.3	83	14.533
Batch system	36.5	49.1	37.6

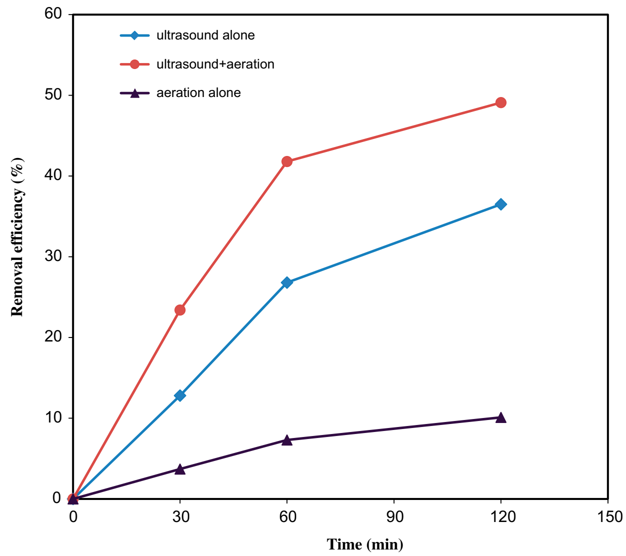


Fig. 6. Removal efficiencies of ammonia vs. time by ultrasound and aeration of air and their combination in batch system for $C_{Ao} = 400$ ppm.

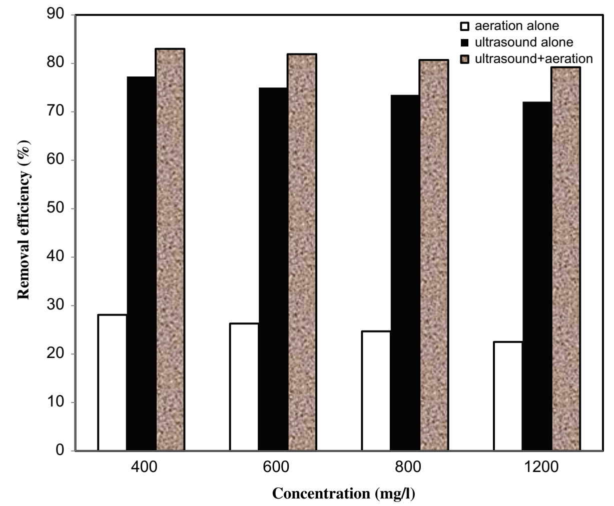


Fig. 8. Removal efficiencies of ammonia by ultrasound and aeration of air and their combination in falling system (total of time = 120 min).

the sound [74]. Therefore, reduction in the ability of the ultrasound wave for ammonia removal is logical.

Fig. 7 reveals the effect of initial concentration on the removal efficiency of ammonia in the presence of ultrasound irradiation and its combination with air in the batch system. It should be noticed that the removal efficiency of the combined system is higher than that of plain sonication for all concentrations. In addition, with increase in ammonia initial concentration the ammonia removal has diminished for both systems.

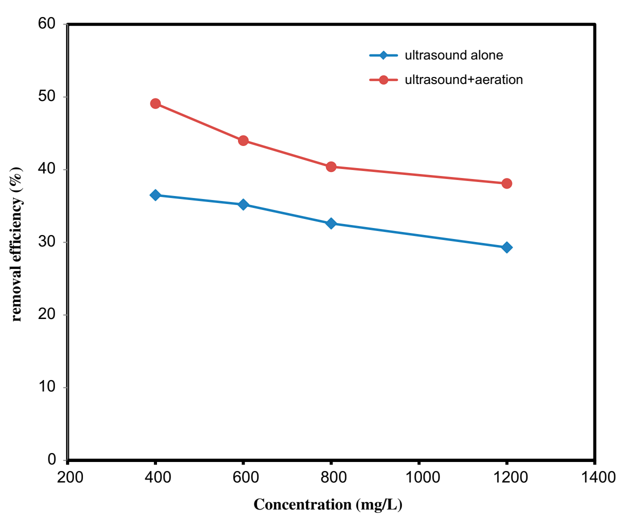


Fig. 7. The effect of initial concentration on the removal efficiency of ammonia in the presence of ultrasound irradiation and its combination with air in the batch system ($C_{Ao} = 400$ ppm; total of time = 120 min).

Fig. 8 indicates the removal efficiencies of ammonia at various conditions with different initial concentration in the falling system. Removal efficiency for three layouts including; plain aeration, plain ultrasonication, and combination of aeration and sonication are illustrated in this figure. The obtained results demonstrate that the combination of ultrasound irradiation and aeration resulted more ammonia removal compared with ultrasound irradiation and aeration alone. The obtained removal efficiency of ultrasound and aeration combination in the 400, 600, 800, and 1,200 mg/L are 7.4, 9.2, 9.7, and 10% more than those of plain ultrasonic irradiation. This elucidates a low impact of aeration on ammonia removal in the falling system. However, aeration is more effective in higher ammonia concentrations.

4. Conclusion

The important feature of this work is to introduce the use of ultrasound technique in the range of MHz for removal or decomposition substance in the field of clean technology for environmental protection. The new ultrasonic technique in comparison with the other treatment methods (such as biological, chemical absorption, and ion exchange processes) has a high potential to remove and destruct organic pollutants. Some advantages of employing the ultrasound waves are saving and optimization of power consumption, no need for chemicals, no generated waste, and treatment time reduction.

In this study, ultrasound wave with 1.7 MHz frequency was propagated directly into a batch and new proposed falling systems in order to investigate its effect on ammonia removal efficiency. In the presence (or absence) of aeration, the effects of initial ammonia concentration on ammonia removal were examined in the systems irritated by ultrasound waves. It was found that with decreasing the initial ammonia concentration, the efficiency of removal was increased in the batch system. However, its effect was not significant in the falling system. The ultrasound irradiation alone could enhance removal efficiency up to 70 and 30% in the falling and batch systems, respectively. The results showed that the combination of ultrasound irradiation and aeration can enhance removal efficiency up to 80 and 49% in the falling and batch systems, respectively. Therefore, this shows in the falling system, both mechanisms of ammonia removal, which are pyrolysis (or the thermal decomposition) and atomization occurred. However, in the batch system the pyrolysis phenomenon is the main mechanism.

From this study, it can be concluded that falling sonoreactor can be a suitable and innovative substitute for industrial pools of ammonia removal.

Nomenclature

C_{Af} — the final concentration of ammonia after the reaction, mg/L
C_{Ao} — the initial concentration of ammonia before the reaction, mg/L
Cp — specific heat capacity, J/g/ K
E — input power, W
m — mass of liquid, kg
p — calorimetry power, W
t — time interval, s
ΔT — rise in temperature of water, K
V — sample solution, L

Greek letters

ε — energy consumption, W/kg

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