Pamukkale Univ Muh Bilim Derg, 28(6), 863-868, 2022

Pamukkale Üniversitesi Mühendislik Bilimleri Dergisi

Pamukkale University Journal of Engineering Sciences

Air stripping of ammonia using a gas-liquid contactor: Effect of pH, temperature, airflow rate, and initial ammonia concentration

Bir gaz-sıvı kontaktör kullanarak amonyağın hava ile sıyırılması: pH, sıcaklık, hava debisi ve başlangıç amonyak konsantrasyonunun etkisi

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Received/Geliş Tarihi: 05.04.2021 Revision/Düzeltme Tarihi: 20.10.2021 doi: 10.5505/pajes.2021.19540
Accepted/Kabul Tarihi: 19.11.2021 Research Article/Araştırma Makalesi

Abstract

Ammonia is one of the most important parameters to be considered in the treatment of domestic and industrial wastewater. If the ammonia discharged to receiver media (river, lake, and sea) is not treated, it may cause environmental problems. The present study examines the impact of pH, airflow rate, initial ammonia concentration, and temperature on ammonia stripping by using air stripping with a gas-liquid contactor. In the system operated in batch mode, the operations were conducted with fixed pH throughout the stripping period (360 min.) and the optimum pH level was determined to be 11. It was shown that high airflow rate and temperature have significant effects on ammonia stripping efficiency and overall volumetric mass transfer coefficient (K_La). When removing ammonia by altering the initial ammonia concentration, no significant changes were observed in ammonia stripping efficiency and K_La. With pH of 11, initial ammonia concentration of 100 mg/L, airflow rate of 20 L/min, and temperature of 55 °C, it was determined that removal of ammonia took approximately 90 min. and the highest overall volumetric mass transfer coefficient for this setting was found to be 0.0462 min^{-1}.

Keywords: Ammonia removal, Gas stripping, Volumetric mass transfer coefficient.

Öz

Amonyak, evsel ve endüstriyel atıksuların arıtılmasında giderilmesi gereken en önemli parametrelerden biridir. Alıcı ortamlara (nehir, göl ve deniz) deşarj edilen amonyak arıtılmazsa çevresel problemlere neden olabilir. Bu çalışma kapsamında bir gaz-sıvı kontaktör kullanılarak hava sıyırma ile amonyak giderimi üzerine pH, hava debisi, sıcaklık ve başlangıç amonyak konsantrasyonunun etkisi araştırılmıştır. Kesikli işletilen sistemde sıyırma süresi boyunca (360 dk.) sabit pH değerlerinde çalışılmış ve optimum pH değeri 11 olarak belirlenmiştir. Yüksek hava debisi ve sıcaklığın amonyağın giderim verimliliği ve genel hacimsel kütle transfer katsayısı (K_La) üzerinde önemli etkilere sahip olduğu gösterilmiştir. Başlangıç amonyak konsantrasyonunun değiştirilmesiyle amonyak gideriminde ve K_La üzerinde dikkat çekici bir değişim gözlenmemiştir. pH 11, başlangıç amonyak konsantrasyonu 100 mg/L, hava debisi 20 L/dk. ve 55 °C'de amonyağın tamamen giderilmesi için yaklaşık 90 dk. gerekli olduğu belirlenmiş ve en yüksek genel hacimsel kütle transfer katsayısı bu işletme değerlerinde 0.0462 dk^{-1} olarak belirlenmiştir.

Anahtar kelimeler: Amonyak giderimi, Gaz sıyırma, Hacimsel kütle transfer katsayısı.

1 Introduction

Mainly having two forms namely ionized (NH_4^+) or unionized (NH_3) forms, ammonia is one of the most important pollutants that may be found in freshwater sources and wastewaters. Ammonia may create a toxic effect on sensitive aquatic biota, decrease dissolved oxygen concentration, and cause eutrophication [1]-[3]. On the other hand, it cannot be replaced in agriculture and industry [4]. Ammonia is widely found in many domestic and industrial wastewaters such as leachate from urban solid waste facilities, wastewaters from fertilizer industry, wastewaters from oil refineries, metallurgical wastewaters, coke plant wastewater, and domestic sources [5]. In the treatment of ammonia wastewaters, the methods of nitrification/denitrification [6], breakpoint chlorination [7], reverse osmosis [8], vacuum UV-based process [9], adsorption [10], electrochemical oxidation [11], chemical precipitation [12],[13] and air stripping [14] are used. Although biological treatment (nitrification/denitrification) is an affordable and environment-friendly method, its disadvantages such as long retention period, effects of climatic conditions, need for large spatial areas, and high effluent concentration after treatment of wastewater (especially industrial) containing high concentrations of ammonia limit the use of this system [15]. Chemical precipitation process is affected by the wastewater composition and may produce secondary pollutant [7]. Breakpoint chlorination creates many toxic and harmful disinfection byproducts and necessitates a high amount of chlorine to reach the breaking point [3]. On the contrary with the aforementioned methods, the air stripping method is rather a simple physical process and it is also suitable specially to remove the high ammonia concentration.

Ammonia is an important commercially produced chemical due to its wide use in numerous technical procedures [16]. Nowadays, the Haber-Bosch process which is a traditional method, has great importance in the synthesis of ammonia. This method consumes a high amount of energy and creates a huge amount of CO_2 during ammonia production [17]. Together with the increase in global demand, the recovery of ammonia gained importance. In the process of ammonia stripping, ammonia in wastewater is switched to the gas phase and then ammonia in the gas phase is transformed into ammonium salts by making use of an acid solution, so ammonia is recovered [4],[18]. When compared to other methods, the process of ammonia stripping

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is used in the treatment of ammonia wastewaters thanks to its advantages such as relatively cheaper, easier operation, creation of less sludge, and higher ammonia stripping efficiency [19]. The efficiency of the air stripping method is affected mainly from four factors: pH, temperature, air-water ratio, and liquid's characteristics [20]. If the air temperature and pH are held constant, the process of ammonia stripping becomes simple and wastewater fluctuations and toxic matters do not affect the removal performance [21].

Many studies have been conducted on the air-stripping of ammonia and complete-stirred gas-liquid contractors such as jet loop reactor [22], water sparged aerocyclone [5], hydrodynamic cavitation [19], rotating packed bed [15] have been used for this purpose. Within the scope of the present study, ammonia stripping was performed by using a gas-liquid contactor (column) having a simple structure with no liquid recirculation and no moving part inside. The effects of pH, temperature, airflow rate, and initial ammonia concentration among the parameters affecting the stripping process were examined. After the optimization of these parameters, the coefficients of overall volumetric mass transfer were calculated by using a mathematical model.

2 Materials and methods

2.1 Experimental setup

Schematic presentation of the experimental setup is illustrated in Figure 1. The gas-liquid contactor used here was made of cylindrical (6 cm diameter and 115 cm height) transparent acrylic material. The liquid volume used in the reactor was 2 L. Air compressor (KULETAS) was used in order to provide the air needed by the system. The volume of air given to the system was adjusted using the air-flow meter connected to the air line. Air was given to the reactor by using a sintered glass diffusor with a pore size of 16-40 micron and a diameter of 6 cm. pH controller (EUTECH alpha-pH2000P) was used in keeping pH constant. Temperature control was performed using a cartridge heater attached to the NOVUS N1030 model controller.

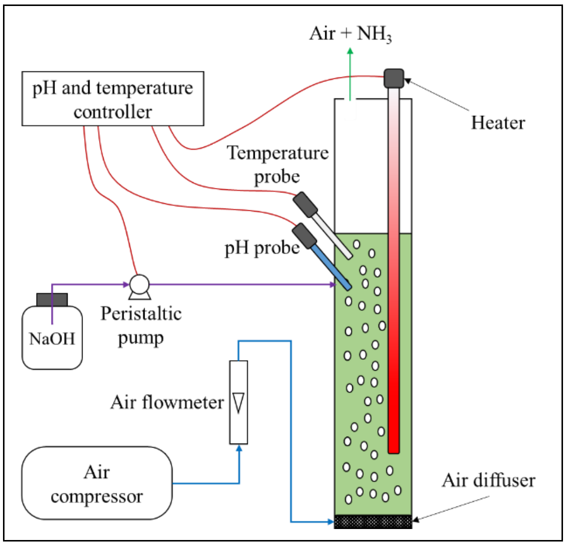


Figure 1. Experimental setup.

Synthetic wastewater was used to determine the optimum operating conditions. NH_4Cl and distilled water were used to prepare the synthetic wastewater. The wastewater prepared at the desired concentration was placed in the gas-liquid contactor shown in Figure 1 and the pH value was adjusted using NaOH.

2.2 Analytical method

Ammonia was determined by the Nessler reagent method. The chemical reagents used in this study were procured from HackLange Company. Ammonia concentrations were measured at 425 nm wavelength by using a spectrophotometer (HachLange DR6000).

2.3 Mathematically modeling the ammonia stripping procedure

Ammonia stripping in gas-liquid contactors is based on the Two-Film Theory. The overall volumetric mass transfer rate of ammonia in the batch operated reactor is calculated using a mathematical model developed by Matter-Müller et al. [23] and expressed with the following equation:

$$-ln\frac{C}{C_0}=\frac{GK_H}{V}\left[1-exp\left(-\frac{K_LaV}{K_HG}\right)\right]t \qquad (1)$$

where C_0 is initial NH_3 concentration (mg/L), C is NH_3 concentration (mg/L) at any time, G is airflow given to the system in a unit time (L/min), K_H is dimensionless Henry's coefficient, t is stripping time (min), V is the total volume of water in the system (L), and K_La is the overall volumetric mass transfer coefficient (min^{-1}).

Since the air used in stripping the ammonia remains in the reactor for a short time, the ammonia in the air released from the reactor would be far away from the saturation level. In this case, $(K_LaV)/(K_HG) \ll 1$ and Equation 1 turns into Equation 2.

$$-ln\frac{C}{C_0}=K_Lat \qquad (2)$$

The coefficient of overall volumetric mass transfer can be calculated using experimental data and Equation 2.

2.4 Calculation of ammonia stripping efficiency

Ammonia stripping efficiency was calculated using Equation 3:

$$Ammonia\ removal, (\%) = [(C_0 - C)/C_0] \times 100 \qquad (3)$$

Where, C_0 and C (mg/L) refer to the initial ammonia concentration and the concentration at another time, respectively.

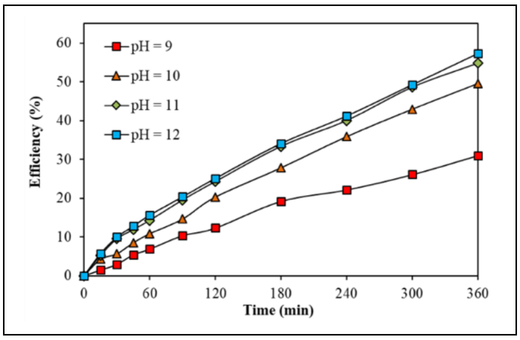
3 Results and discussion

3.1 Effect of pH on ammonia stripping

The transition of ammonia ion (NH_4^+) to free ammonia (NH_3) is the most important factor for a successful stripping process [24]. In the ammonia stripping procedure, pH is a parameter determining the balance of ammonium and ammonia (Equation 4).

$$NH_4^+ + OH^- \leftrightarrow NH_3 + H_2O \qquad (4)$$

In studies examining the effect of pH on stripping of ammonia, various experiments were carried out at different pH levels (9, 10, 11, and 12) by keeping initial ammonia concentration (100 mg/L), airflow rate (5 L/min), and temperature (25 °C) constant. In order to keep pH constant in experiments, a solution containing 2 M NaOH solution was given into the reactor by using a peristaltic pump and a pH controller. The ammonia removal efficiencies obtained at different pH levels are presented in Figure 2.



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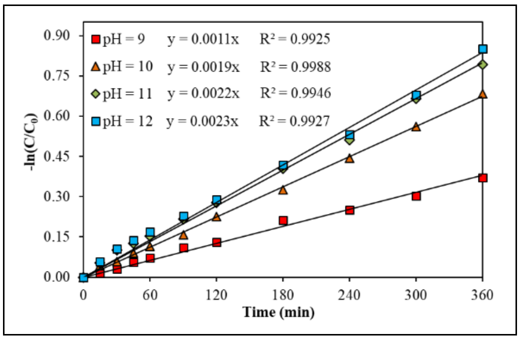

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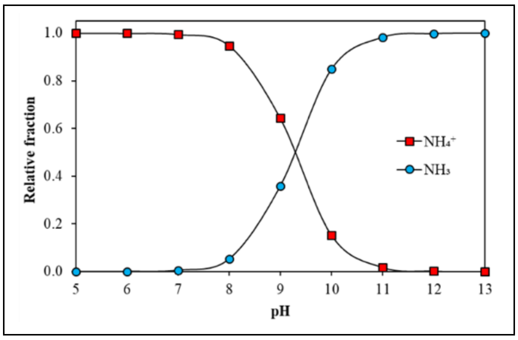

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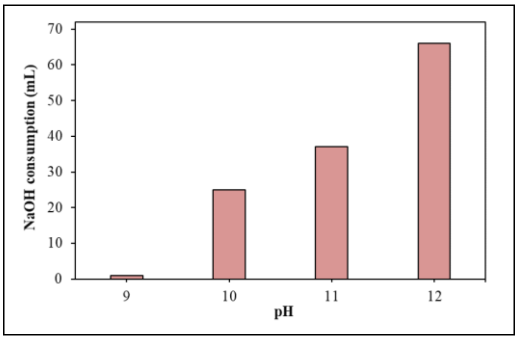


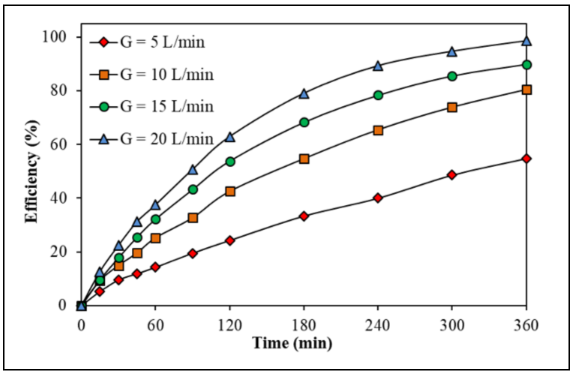
(d)

Figure 2. Changes of (a): Aammonia stripping efficiency, (b): Overall mass transfer coefficients, (c): Ammonium ion and ammonia transformation, and (d): Consumed NaOH at different pH levels (Experimental conditions: T=25 °C, NH_3=100 mg/L, G=5 L/min).

As seen in Figure 2(a) and Figure 2(b), both ammonia stripping efficiency and overall volumetric mass transfer coefficient increase with the pH level of the solution. Gas-form ammonia (NH_3) and ammonium ion (NH_4^+) are in equilibrium at pH level of approximately 9.25. At pH level of 9, ammonium ions are dominant (Figure 2 (c)). For this reason, ammonia stripping efficiency was found to be 31% at the end of 360 min. period at pH level of 9. At the pH level of 10, ammonia stripping efficiency increased and reached the level of 49%, whereas the ammonia stripping efficiency reached 55% at pH level of 11 and 57% at pH level of 12. As seen in Figure 2(a), the ammonia stripping efficiencies increased with increasing pH and there was no significant difference between ammonia stripping efficiencies at pH 11 and pH 12. Calculated using Equation 2 and illustrated in Figure 2(b), overall volumetric mass transfer coefficients increased from 0.0011 min^{-1} to 0.0023 min^{-1} with pH increasing from 9 to 12. As seen in Figure 2(b), at pH levels of 11 and 12, overall volumetric mass transfer coefficients are as close to each other as ammonia stripping efficiencies. In Figure 2(c), it can be seen that all the ammonium is in ammonia form at the pH levels of 11 and 12. It explains why there is no significant difference between ammonia stripping efficiencies at pH levels of 11 and 12. Figure 2(d) shows the amounts of NaOH consumed during the experiment. As can be seen, NaOH consumption increases with increasing pH. NaOH consumption at pH level of 12 is almost twice that of pH 11. However, there is almost no difference between ammonia stripping efficiencies and between overall volumetric mass transfer coefficients. Considering the NaOH costs, overall volumetric mass transfer coefficients, and ammonia stripping efficiencies, the optimum pH level was chosen to be 11. In their study, Lin et al. also reported similar results [25].

3.2 Effect of airflow rate on ammonia stripping

Airflow rate is of great importance parameter in ammonia stripping process. To determine the impact of airflow rate on the ammonia stripping, the experiments were carried on at airflow rates of 5, 10, 15, and 20 L/min and the results are presented in Figure 3. In these experiments, pH and temperature were kept at 11 and 25 °C, respectively. Figure 3(a) illustrates that ammonia stripping efficiencies at the end of 360 min. stripping period at airflow rates of 5, 10, 15, and 20 L/min were found to be 57%, 80%, 90%, and 99%, respectively. It was determined that ammonia stripping efficiency increased with increasing airflow rate. It suggests that the time needed to ensure a specific level of ammonia stripping efficiency decreased with increasing airflow rate. As seen in Figure 3(b), overall volumetric mass transfer coefficient ranged between 0.0022 and 0.0092 min^{-1} depending on the airflow rate. It can be stated that K_La values linearly increased with increasing airflow rate. It was reported in previous studies that increasing airflow rate significantly affected various parameters such as coefficient of overall volumetric mass transfer, stirring behavior, and gas-liquid interface area and positively contributed to the transition of liquid ammonia to gas form [3],[24],[26]. In ammonia stripping process, mass transfer resistance occurs in the gas film side due to the high solubility of ammonia in liquid [5]. It is possible to reduce the mass transfer resistance with increasing airflow rate. Thus, stripping of ammonia from the liquid phase is accelerated. Mass transfer resistance in the gas film can be reduced by increasing the airflow rate and stripping of ammonia from the liquid phase is promoted. In conclusion, the higher airflow rate increases the ammonia stripping efficiency, besides the K_La. Considering the removal efficiencies and overall volumetric mass transfer coefficients, the airflow rate for the next experiments was chosen to be 20 L/min.



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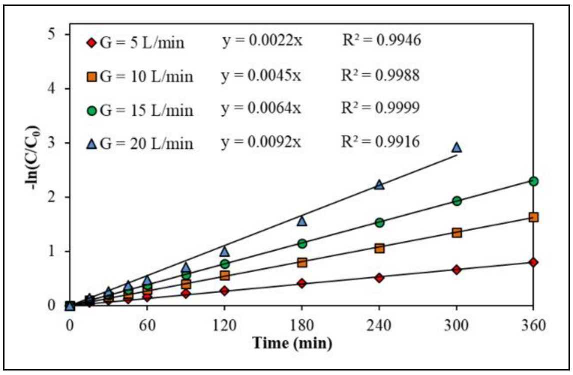

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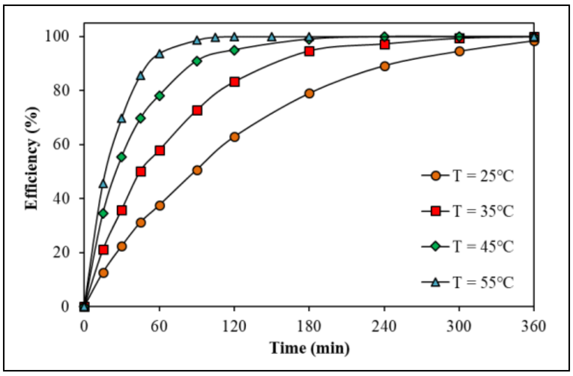
Figure 3. Changes of (a): Ammonia stripping efficiency and (b): and overall volumetric mass transfer coefficients at different airflow rates (Experimental conditions: T=25 °C, pH=11, NH_3=100 mg/L).

3.3 Effect of temperature on ammonia stripping

Temperature is another important factor affecting ammonia stripping [22],[27]. With experimental conditions of pH level of 11, initial ammonia concentration of 100 mg/L, and airflow rate of 20 L/min, experiments were performed at 25, 35, 45 and 55 °C in order to determine the effects of temperature and the results are presented in Figure 4. At the end of 360 min. period, 98% ammonia stripping was achieved at 25 °C, whereas all the ammonia was stripped at higher temperatures. Comparing the temperature-related ammonia stripping efficiencies at the end of 60 min. period, ammonia stripping efficiency was found to increase from 37% to 94% with the increasing temperature. In this process, 98% ammonia stripping efficiency could be achieved in 360 min. at 25 °C, 300 min. at 35 °C, 180 min. at 45 °C, and 90 min. at 55 °C (Figure 4(a). As a result, with increasing temperature, the stripping time required to reach the desired ammonia concentration decreased. In this case, the higher the temperature of the wastewater, the higher the ammonia removal. It can be clearly seen in Figure 4(b) that ammonia stripping occurred faster with increasing temperature. Overall volumetric mass transfer coefficients at 25, 35, 45, and 55 °C were found to be 0.0092, 0.0153, 0.0258, and 0.0462 min^{-1}, respectively. Increasing the temperature increases Henry's law coefficient and molecular diffusion coefficient of ammonia and, thus, it decreases the water solubility of ammonia. It results in an increase in ammonia's liquid-to-gas mass transfer coefficient [24],[28]. If possible, the process should be carried out at a higher temperature in air-stripping the ammonia. For this reason, the optimum temperature value was chosen to be 55 °C.



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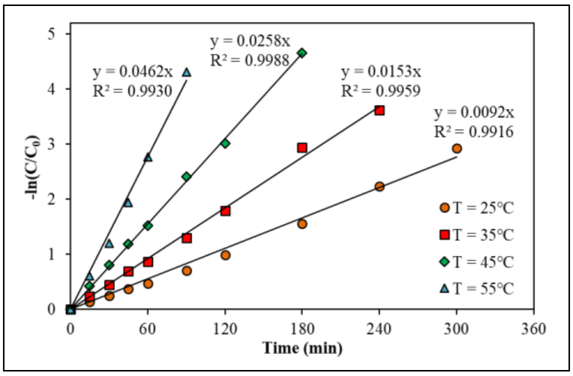

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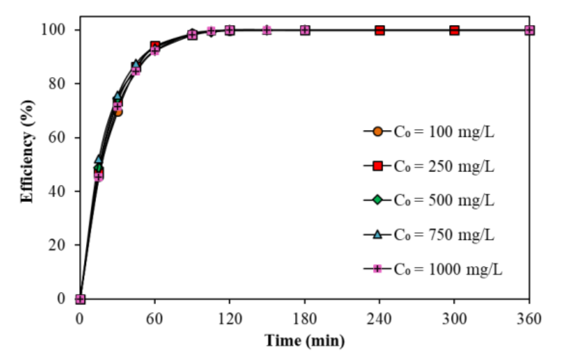
Figure 4. Changes of (a): Ammonia stripping efficiency and (b): Overall volumetric mass transfer coefficients at different temperatures (Experimental conditions: pH=11, NH_3=100 mg/L, G=20 L/min).

3.4 Effect of initial ammonia concentration on ammonia stripping

To determine the effect of initial ammonia concentration on the ammonia stripping, experiments were performed at pH level of 11, temperature of 55 °C, and airflow rate of 20 L/min. In order to simulate varying the ammonia concentrations varying depending on the type of wastewater, synthetic wastewaters were prepared with different initial concentrations (100, 250, 500, 750, and 1000 mg/L) and ammonia stripping efficiencies were investigated. The ammonia stripping efficiency results are presented in Figure 5. For all the initial ammonia concentrations, ammonia stripping efficiencies higher than 98% were achieved at the end of 90 min. stripping process. As seen in Figure 5(a), ammonia stripping efficiencies in the course of time are very close to each other. Thus, it can be concluded that ammonia stripping efficiency is independent of initial ammonia concentration. Similar results were also reported in different studies carried out using different reactors [5],[25]. It can be clearly seen in Figure 5(b) that initial ammonia concentration has a very small effect on overall volumetric mass transfer coefficients. Although ammonia concentration significantly changed (from 100 to 1000 mg/L), $K_L a$ values ranged between 0.0433 and 0.0469 min^{-1}. Considering that system is controlled through diffusion from gas film layer, it can be concluded that different initial concentrations did not affect the transition of ammonia to gas phase [19].



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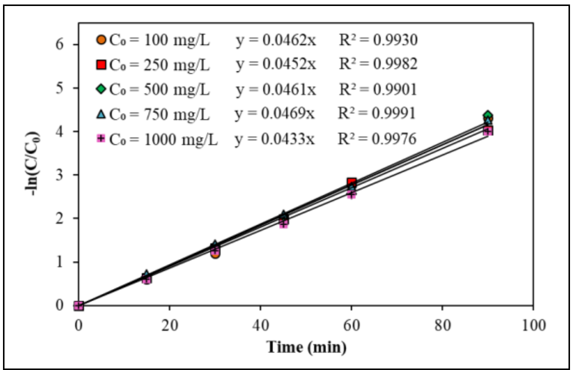

(b)

Figure 5. Changes of (a): Ammonia stripping efficiency and (b): Overall volumetric mass transfer coefficients at different initial concentrations (Experimental conditions: G=20 L/min, pH=11, T=55 °C).

4 Conclusions

In the present study, the effects of pH, temperature, airflow rate, and different initial ammonia concentrations among the parameters affecting the air-stripping of ammonia in the batch operated gas-liquid contactor were examined. The most important parameters in ammonia stripping procedure were determined to be pH, temperature, and airflow rate. The optimum values of these parameters were determined to be 11 for pH, 20 L/min for airflow rate, and 55 °C for temperature. When changing the initial ammonia concentration, no remarkable change was observed in ammonia stripping efficiency. It was concluded that conducting the stripping process at a higher airflow rate and temperatures might significantly decrease the operation time. Moreover, it was determined that even though operating the system at pH value of 12 might reduce the stripping time, the cost of chemical input might eliminate this advantage. In further studies, using different diffusor types might further decrease the ammonia stripping times.

5 Acknowledgments

This research has been supported by Kastamonu University Scientific Research Projects Coordination Department (Project Number: KÜ-BAP01/2017-9).

6 Author contribution statements

In the scope of this study, Gokce Didar DEGERMENCI contributed formation of the idea, design, supplying the materials, data collection, conducting the analyses, literature review, writing and assessment of the obtained results.

7 Ethics committee approval and conflict of interest statement

There is no need to obtain permission from the ethics committee for the article prepared.

There is no conflict of interest with any person/institution in the article prepared.

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