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Process intensification and modeling of a hybrid air stripping-biofilter (ASBF) system for removal of benzene from produced water

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ABSTRACT

In this work, a novel concept of combining air stripping followed by biofilter technology (ASBF) is introduced and through a mathematical model, the performance of the system for the removal of benzene as a representative pollutant is evaluated. The system performance is investigated for a flow rate range of 8.5×10^{-3} – 17×10^{-3} m³/s and benzene concentrations of 45×10^{-3} and 75×10^{-3} kg/m³. The removal efficiency ranged from 29.2 to 90.3% for the air stripping section of the system at varying packing types and sizes while the biofilter section had a removal efficiency range of 34–99% for varying empty bed residence times (EBRTs). The overall removal efficiency for the ASBF system ranged from 92 to 97%. The simulation results also showed that the ASBF system can be effectively used to remove benzene from polluted industrial wastewater. Parameter perturbation study that was performed using factorial design showed influent benzene concentration, stripping factor, air stripping column packing size, packing type, and biofilter EBRT are the main factors that influence the removal of benzene.

Keywords: Air stripping; Benzene; Biofilter; Empty bed residence time; Process intensification; VOC; Wastewater; Process modeling; Process simulation

1. Introduction

One of the industrial processes that generate wastewater containing soluble volatile organic compounds (VOCs) is the fossil fuel extraction process. The water that is extracted along with the oil and gas from the shale rocks is called produced water. Produced water, also known as formation water contains a range of soluble organic and inorganic chemicals and salts, thereby necessitating treatment before discharging or recycling. Recently, major oil and gas companies are incorporating environmental sustainability in their operational goals. Due to its potential threat to human health, benzene was ranked sixth in the priority list of hazardous compounds [1].

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Short-term exposure to benzene may cause health conditions such as irritation of the skin, eyes, and upper respiratory tracts. It may also cause blister on the skin. Long-term exposure to benzene may cause blood disorders, reproductive and developmental disorder, and cancer [2]. The United States Environmental Protection Agency [3] classified benzene as "Group A," known human carcinogen for all routes of exposure. The maximum concentration threshold set by USEPA for benzene in drinking water is $5 \,\mu g/L$.

The necessity to meet stringent environmental regulations has led to the development and improvement of treatment processes available for benzene removal from industrial wastewater. Recovery treatment processes and mineralization treatment processes are the two groups that encompass the available treatment methods for benzene removal from wastewater. The recovery processes, as the name implies, extracts the benzene from the contaminated fluid while the mineralization processes decomposes the benzene into carbon dioxide and water. The preference of one removal process over the other is usually governed by several important factors which include the physicochemical properties and concentration, cost and availability of suitable equipment to achieve the desired removal objectives, and the legal environmental regulations.

The recovery processes include technologies such as adsorption (activated carbon-based and silica-based sorbent), condensation, membrane techniques (pervaporation, reverse osmosis, electrodialysis, and dialysis), and absorption (water scrubber). The mineralization processes include processes such as biofiltration, thermal oxidation, catalytic oxidation, and plasma methods. Several studies have shown that these control technologies can be effective in removing benzene from industrial waste gasses [4,5] but to effectively apply such technologies to an industrial wastewater containing VOCs such as benzene, the VOCs need to be stripped from the aqueous phase into a gas phase before either recovering or mineralizing them. Therefore, an innovative removal technology for VOCs removal from industrial wastewater system is essential. In order for the proposed removal process to be competitive with existing treatment methods, it must meet certain conditions. First, it should have the capability to handle continuous operation. Second, the size of the treatment reactors must be comparable to other VOC removal processes, and lastly, the system should be able to easily adjust to fluctuations in influent VOCs concentrations. To satisfy these requirements, in this work, a hybrid system of air stripping-biofilter (ASBF) is proposed.

Air stripping involves the use of air to enhance the volatilization of VOCs from aqueous phase into gas phase. It is very useful in the treatment of industrial wastewater laden with VOCs. Air stripping technology could be applied either through a stripping basin or a stripping tower. However, when high efficiency is required, stripping tower is preferred over stripping basin [6]. Typically, a stripping tower involves a countercurrent flow pattern between the air and the wastewater through specific packings which are designed to enhance the contact between the water and the gas phase. Previous studies have reported approximately 99.9% removal of most VOCs using air stripping technology [5]. Air stripping technology is merely the transfer of VOCs from an aqueous phase into a gaseous phase, therefore, the exiting air requires some form of treatment and our proposed alternative strategy is to treat the air-benzene mixture exiting the air stripping column with a biofilter. Biofiltration is useful for processing fluctuating influent VOCs concentrations and it performs well in mineralizing a wide range of VOCs [7,8]. In addition, it has been demonstrated that varying concentrations of benzene [9] could be successfully removed by biofiltration.

The objective of this study is to develop a mathematical model to illustrate the operation of the proposed ASBF system and to compare the predicted performance with the other treatment technologies. The second goal is to investigate the effects of important operational parameters on the performance of the ASBF system. Benzene was selected as the model VOC due to its prevalence in industrial wastewater. Its biodegradability and water solubility also makes it a suitable compound for this study. Literature on techniques that combines air stripping and biofilter technology for benzene removal is scarce, therefore this study will contribute to the knowledge of techniques available for benzene removal from industrial wastewater.

2. Materials and methods

2.1. Air stripping-biofilter system (ASBF)

Mathematical modeling was used to model the performance of the proposed ASBF system using benzene as a model VOC pollutant. The schematic flow diagram of the proposed system is presented in Fig. 1. In the proposed system, air stripping unit has dual purpose of stripping VOCs from water as well as humidifying the influent air to the biofilter unit. This novel arrangement eliminates the use of a humidifier as compared to a conventional biofilter system; thus capital equipment cost will be reduced. In conventional biofilter system, humidifier is needed to keep biofilter media moist. Some of the underlying assumptions made during the development of the model include (a) wastewater is assumed to be dilute with



Fig. 1. Schematic diagram of proposed air strippingbiofilter system.

respect to the soluble VOC, (b) the biofilter is assumed to behave as a plug flow reactor, (c) two-film theory is assumed to describe the relationship between the liquid-phase resistance and the gas-phase resistance in the air stripping column, (d) pH and temperature are constant, (e) microbial degradation of benzene is described by Monod kinetics as per reference [10].

2.2. Air stripper model equations

The process model and design equations guiding the operation of an air stripping unit have been previously developed [6,11]. The equation that relates to the height of a stripping tower is as follows:

$$Z = HTU \times NTU \tag{1}$$

where Z = height of stripping tower packing, m; HTU = height of a transfer unit, m; NTU = number of transfer units.

Height of a transfer unit HTU is defined as:

$$HTU = \frac{L}{\rho K_{La}}$$
(2)

where L =liquid loading rate, kg/(m² s) defined as:

$$L = \frac{Q_{\rm w}\rho}{A} \tag{3}$$

where Q_w = water flow rate, m³/s; ρ = density of water, kg/m³; A = cross sectional area of tower, m².

The overall mass transfer rate constant (K_{La}), which determines the rate at which the VOCs are transferred from water to air, was estimated using Sherwood and Holloway empirical correlations [6]. This estimate is applicable due to the low concentrations of VOCs in wastewater.

$$K_{\rm La} = \propto \times D_{\rm L} \times \left(0.305 \frac{L}{\mu}\right)^{1-n} \left(\frac{\mu}{\rho D_{\rm L}}\right)^{0.5} \tag{4}$$

where α , n = constants based on packing type (for example, α = 330 and n = 0.22 for 25 mm Raschig ring packing); $D_{\rm L}$ = VOC diffusion coefficient in water (m²/s).

 μ = viscosity of water (1.002 × 10⁻³ Pa s at 20°C); ρ = density of water (1,000 kg/m³ at 20°C).

The HTU is a measure of the mass transfer characteristics of the packing medium. The number of transfer unit is defined as:

$$NTU = \left(\frac{R}{R-1}\right) \ln\left(\frac{(C_{in}/C_{out})(R-1)+1}{R}\right)$$
(5)

where R = stripping factor and is defined as:

$$R = H' \times \left(\frac{Q_{\rm a}}{Q_{\rm w}}\right) \tag{6}$$

where $Q_a = air$ flow rate, m^3/s ; H' = dimensionless henry's law constant; $C_{in} = influent$ concentration of VOC in liquid phase, kg/m³; $C_{out} = effluent$ concentration of VOC in liquid phase, kg/m³.

Concentration of VOC in exiting gas phase,

$$A_{\rm out} = (C_{\rm in} - C_{\rm out}) \times Q_{\rm w}/Q_{\rm a} \tag{7}$$

The exiting air laden with VOCs from the stripping tower is the inlet air to the biofilter.

2.3. Biofilter model equations

The biofilter can be divided into subsections of volume $(A_b \times dz)$ and taking mass balance around each section (*j*) gives:

$$Q_{a}C_{j} = Q_{a}C_{j+1} + r_{j}A_{b} \times dz$$
(8)

where $Q_a = \text{air flow rate } (\text{m}^3/\text{s})$; C_j and $C_{j+1} = \text{concentration}$ (kg/m³) in section *j* and (*j* + 1), respectively;

dz = subsection height of biofilter, m; A_b = biofilter cross sectional area, m²; r_j = volumetric biodegradation rate in subsection j (kg/ $m_{\text{biofilter}}^3$ /s).

For biological systems, the rate can be expressed in terms of Monod kinetics:

$$r_j = V_{\max,j} \frac{C_j/H'}{K_m + C_i/H'} \tag{9}$$

where C_j/H' = the liquid phase concentration (kg/m³) of VOCs expressed in terms of gas-phase concentration at the interphase between liquid and air related by Henry's law; H' = dimensionless Henry's law constant; K_m = Monod constant (kg/m³); $V_{max,j}$ is represented as Eq. (10) as shown in Ref. [12].

$$V_{\max,j} = \left[1 - \frac{a}{\mu_{\max}} \left(1 + \frac{K_{\mathrm{m}}}{C_j/H'}\right)\right] V_{\max,\max}$$
(10)

where $V_{\text{max,max}} = \text{maximum } V_{\text{max}}$, kg _{VOC}/ $m_{\text{biofilter}}^3$ /s; $\mu_{\text{max}} = \text{maximum specific growth rate, s}^{-1}$, a = bacteria decay rate, s $^{-1}$.

Re-writing Eq. (8) as a differential equation and incorporating Eqs. (9) and 10 yields:

$$\frac{\mathrm{d}C}{\mathrm{d}Z} = \left[\frac{a}{\mu_{\mathrm{max}}} - \frac{C/H'}{K_{\mathrm{m}} + C/H'}\right] \times V_{\mathrm{max,max}} \times \frac{A_{\mathrm{b}}}{Q_{\mathrm{a}}} \tag{11}$$

Eq. (11) can be solved using the boundary condition $C = C_0$ (at the inlet) at z = 0.

A set of dimensionless variables is chosen for making the model Eq. (11) dimensionless These variables are:

$$u = \frac{C}{C_{\rm o}} \tag{12}$$

$$h = \frac{z}{H} \tag{13}$$

$$\alpha = \frac{a}{\mu_{\max}} \tag{14}$$

$$\beta = \frac{K_{\rm m}H'}{C_{\rm o}} \tag{15}$$

$$\tau = \frac{V_{\max,\max}}{C_{o}} \times \text{EBRT}$$
(16)

where $EBRT = \frac{H \times A_b}{Q_a}$ and H = biofilter height.

The model Eq. (11) can be reduced to:

$$\frac{\mathrm{d}u}{\mathrm{d}h} = \left[\alpha - \frac{u}{u+\beta}\right] \times \tau \tag{17}$$

Subject to the boundary conditions: h = 0, u = 1.

3. Results and discussion

3.1. Performance of ASBF system

The performance of ASBF system was initially compared with a hybrid system which combines a biofilter with a suspended activated sludge bioreactor [12]. In our simulation study, the benzene laden wastewater at a concentration of 45×10^{-3} kg/m³ was assumed to be flowing into the hybrid bioreactor at the water flow rate of 5.6×10^{-7} m³/s. The air flow rate was set at 1.7×10^{-5} m³/s and $V_{\text{max,max}}$ was chosen as 5.6×10^{-5} kg_{benzene}/ $m_{\text{biofilter}}^3$ /s based on the estimated value of V_{max} . The parameters used to solve the model and source of references for these parameters are presented in Table 1. A MATLAB code ode45 was used to solve the model equations.

The result of the performance of the ASBF for benzene removal is presented in Fig. 2. The removal efficiency was calculated as the ratio of removed benzene to influent benzene expressed in percentages, while the removal rate was calculated as the ratio of removed benzene to the residence time. The air-towater ratio is a function of the stripping factor and manipulating this parameter could enhance the benzene removal efficiency of the system. Therefore, performance of the ASBF system in terms of removal efficiency and removal rate was investigated at varying stripping factors and benzene concentrations. Increasing the air flow rate which is a function of the stripping factor improves benzene removal efficiency at constant benzene concentration and water flow rate. However, increasing the air flow rate beyond the column design pressure threshold could lead to flooding [6].

Parameters used for solving ASBF model equations for the removal of benzene from industrial wastewater

Table 1

Parameters	Value	Unit	Refs.
$ \frac{\mu_{\max}}{K_{m}} $ $ a $ $ D_{L} $ $ H' $	$\begin{array}{c} 2\times10^{-4}\\ 269\times10^{-6}\\ 4.7\times10^{-7}\\ 8.99\times10^{-10}\\ 0.185\end{array}$	1/s kg/m ³ 1/s m ² /s Dimensionless	[10] [14] [15] [16] [16]



Fig. 2. Effect of stripping factor on biofilter removal rate (A) and removal efficiency (B).

The removal rate of the biofilter is an important factor to consider when assessing the performance of the ASBF system. The upper plot (A) in Fig. 2 shows that as stripping factor increases at constant water flow rate, the removal rate is being driven by the inlet benzene concentration and at high stripping factor (or high air flow rate) removal rate drops. The lower plot (B) of Fig. 2 shows that the optimal stripping factor was 10, beyond which there is no further advantage in the removal efficiency. The optimal removal rate (or elimination capacity) lies between 1.29×10^{-9} and 1.33×10^{-9} kg/m³/s (or 4.64–4.79 mg/L/h) as seen in Fig. 2.

3.2. Comparison with other novel benzene removal technologies

Comparison of the ASBF system with other innovative benzene removal technologies in the literature (Table 2) shows that the ASBF treatment performance is comparable (>99%) for the parameters used in our simulation study. Table 2 shows two other studies with rotating biological reactor with the efficiency of 98% and a hybrid bioreactor unit with efficiency of 79.6%. The design parameters for the simulation of ASBF system based on the influent wastewater benzene concentrations are presented in Table 3. Table 4 shows the impact of air flow rate and empty bed residence time (EBRT) on the performance of ASBF system. Thus, the simulation data shows that only 72 s are enough to have >99% removal of benzene using ASBF system.

An advantage of the ASBF over other treatment methods is that packed column air stripper can operate effectively over varying air flow rates. Another advantage of the ASBF system over other benzene treatment method is the low energy usage and the complete elimination of the benzene pollutant without transferring from one phase to another. The implication of these advantages is the reduction in operational cost when ASBF system is used as compared to the other systems.

3.3. Influence of operating conditions on overall removal efficiency

The efficient operation of the ASBF is dependent on several operational factors. One of these factors is the air flow rate through the air stripping column. The air flow rate is an important operating parameter in an air stripping column because it affects the overall removal efficiency of the system. Fig. 3 shows the effect of varying stripping factor on benzene concentration along the biofilter of the ASBF system. To investigate the effect of stripping factor in the air stripping column on the concentration profile in the biofilter, the water flow rate in the stripping column was kept constant at $0.56 \times 10^{-6} \text{ m}^3/\text{s}$ while varying the inlet air flow rate from $8.3 \times 10^{-6} \text{ m}^3/\text{s}$ to $68.3 \times 10^{-6} \text{ m}^3/\text{s}$. As the stripping factor increases, relative benzene concentration also decreases. Thus, proper selection of stripping factor is critical in determining the size of biofilter.

Increasing the air flow rate in an air stripping column also increases the removal efficiency of the column so long the pressure drop is between 200 and

Table 2

Comparison between ASBF system and other benzene removal technologies

Treatment process	Max. removal %	Refs.
Air stripping-biofilter (ASBF) system	99.9	This Study
Hybrid bioreactor consisting of a combination of biofilter and suspended activated sludge	79.6	[13]
Rotating biological contactor with biofilm promoting mats	98	[17]

Table 3

ASBF design parameters for the removal of benzene from industrial wastewater

Air stripping tower	
Height of packing	0.75 m
Number of transfer unit	3.46
Height of transfer unit	0.22 m
Packing type	25 mm Rashig ring
Stripping factor	2.8
Biofilter	
Height of packing	0.6 m
EBRI	72 s

Table 4

Impact of air flow rate and EBRT on the performance of ASBF

EBRT (s)	Air flow rate $(m^3/s) \times 10^{-6}$	Biofilter efficiency (%)		
18	8.3	34.44		
	17	66.51		
25	8.3	48.1		
	17	90.29		
36	8.3	68.34		
	17	99.99		
72	8.3	99.99		
	17	99.99		

Note: Influent benzene concentration = 45×10^{-3} kg/m³.

 400 N/m^2 per meter of tower height [6]. Increasing the air flow rate in a biofilter could lead to sloughing off of the thick biofilm layer, thereby enhancing mass transfer into the biofilm, thereby causing an improvement in the removal efficiency of the system. At low air flow rate, the stripping efficiency of the air stripping column decreases [4,18].

Moisture content of biofilter packing material is another factor that affects the performance of a biofilter. It determines the efficiency of biodegradation in the biofilter [19]. Due to the absence of an aqueous phase in biofilter, the presence of adequate amount of moisture influences the diffusion of the organic compound into the bacteria for biodegradation to occur. In addition, the presence of moisture in the biofilter helps to prevent dryness of the biofilter due to evaporation. Evaporation of moisture occurs in biofilter through the generation of heat from the biodegradation reaction [20]. The ASBF system eliminates the



Fig. 3. Biofilter performance at varying stripping factor.

need to pre-humidify the air before passing into the biofilter because the air exiting the air stripping column would carry enough moisture. This implies a reduction in the number of unit operations required, thus operational and capital cost savings.

The effects of the design and operating parameters of the air stripping section of the ASBF on benzene removal efficiency was examined in order to determine the sensitivity of the operational parameters. Inlet concentrations of benzene and air flow rate affect the removal efficiency of benzene from the ASBF (Table 5). Table 5 also shows a reduction in removal efficiency when the benzene inlet concentration increased from 45×10^{-3} kg/m³ to 75×10^{-3} kg/m³ at constant air flow rates. To maintain high removal efficiency, increase in inlet concentration of benzene needs to be followed with an increase in air flow rate.

The packing size and type also affects the height of the packing required to achieve the desired removal efficiency. Table 5 also shows that the height of packing necessary to maintain the same removal efficiency increases from using 12 mm Raschig ring to 12 mm Berl saddle and 75 mm Tile. Therefore, the optimum packing type and size required to achieve the shortest column height is 12 mm Raschig ring for our study. Although, increase in air flow rate increases the stripping efficiency, the air stripping column should be operated below the flooding point. Fig. 4 shows that increasing the air-to-water ratio (stripping factor) from 20 to 140 reduces the height of packing required to achieve the same removal efficiency. Parameter perturbation performed on the ASBF system using factorial design revealed that the removal rate is influenced by the stripping factor of the air stripping column, air stripping column packing size, packing type, and biofilter EBRT. Thus, optimum balance between these parameters is needed to achieve desired removal efficiencies at minimum capital and operational cost.

Table 5

Effect	of	inlet	benzene	concentration,	air flo	w rate,	packing	type	and	packing	size	on	benzene	removal	efficiency	7 in the
biofilt	er s	sectio	n of ASB	F												

Concentration $(kg/m^3) \times 10^{-3}$	Air flow rate $(m^3/s) \times 10^{-6}$	12 mm Raschig ring Height of packing (n	12 mm Berl Saddle n)	75 mm Tile	Biofiler efficiency (%)
45	8.5	40	126	172	68
	17	34	106	145	99.9
75	8.5	40	126	172	42
	17	34	106	145	81

Notes: Water flow rate = $0.56 \times 10^{-6} \text{ m}^3/\text{s}$; Packing-Tile = 75 mm; Temperature = 20°C ; Inlet concentration = 45 ppm; Pressure = 101.3 kPa; Liquid loading rate = $2 \times 10^{-3} \text{ kg/m}^2/\text{s}$.



Fig. 4. Effect of varying air-to-water ratio on height of packing.

4. Concluding remarks

In this work, a novel process based combining air stripping followed by biofilter technology (ASBF) is proposed. The performance of ASBF system for the removal of benzene as a representative pollutant is evaluated using a mathematical model. The overall removal efficiency for the ASBF system ranged from 92 to 97%. The simulation results showed that the airstripping-biofilter system effectively removes benzene from polluted industrial wastewater and the system is comparable to other removal technologies in the literature. The effects of selected operational parameters of the performance of ASBF treatment method in terms of benzene removal from industrial wastewater were examined. Parameter perturbation study that was performed using factorial design showed influent benzene concentration, stripping factor, air stripping column packing size, packing type and biofilter EBRT are the main factors that influence the removal of benzene. Experimental verification of the ASBF system is needed for further verification of the results presented in this work.

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