A packed bed Reactor Combined with Membrane Unit for the Elimination of Toluene Vapours using a Novel Packing Material

B.Sundar, V.Saravanan, M.Rajasimman

Abstract: ***Toluene is a colorless and aromatic oily liquid primarily used in the petrochemical and polymer processing industries and has been used in this study as the target compound. Continuous experiments were performed in a biofilter on a laboratory scale, followed by a membrane reactor to monitor toluene as one single contaminant. This bioreactor device included a reactor with a packed bed and a membrane array. Pearl millet stacks and berl saddles have been used as packing material for the development of the attached microorganism. Toluene was efficiently treated, with toluene effluent concentrations held at less than 0.4 g m^{-3} and a Total Removal Efficiency (TRE) of more than 96 % achieved when fluctuating loads were faced by the packed bed reactor. The combined packed bed reactor system had a maximum RE of 93.8 g m^{-3} h^{-1}, which was higher than the one obtained with the packed bed reactor alone. In this work the influences on gas membrane separation were also explored in the combined bioreactor and membrane fouling.***

Keyword: Biofilter, Toluene, Membrane, Elimination Capacity

I. INTRODUCTION

Bioreactors are more cost-effective than other conventional methods of treating effluent-volatile organic compounds (VOCs), especially for large volumes with lower concentration emissions[1]. In recent years the removal of VOCs through biofiltration systems has gained increasing attention and focus on research[2]. Biological processes on different microbial species, with the biodegradation of organic loading to mineral products occurring over multiple steps and intermediate compound growth. Bioreactors are delicate to flow in VOC loadings for those details.

While biofiltration is done excellently at steady contaminant loading in waste gas streams for VOC control, variations are typical of real-life applications. One approach to decreasing the concentration of outlet containment is by installing multiple series biofilters to expand the effective volume of the reactor and increase the residence time[3]. Although studies have adjusted the success of this technology during transient contaminant loadings, large land requirement is a major disadvantage and limits its feasibility in areas where land acquisition costs are relatively high. (4)

Combined reactors, involving of a packed bed reactors followed by a chemical, physical, have been used to increase the RE of influent gas containing fairly higher concentrations of VOCs[5]. Catalytic oxidation of VOCs therefore requires higher working temperatures and adsorbents required to be restored next flooded adsorption, both expensive and energy-requirement processes. [2]

Attenuating the variation in input to biotic methods offers an operative result, particularly for causes where fluctuations in pollutant loading are normally found. Solo and dual activated carbon adsorption reactors were used as buffer reactor with unstable pollutant loads before a biofilter for the monitoring of VOC emissions[6]. Conversely, submissions of this knowledge are obstructive in respects to needed pollutant concentration. Because pollutants are mainly detected and stored in the adsorption unit, the biofilter is subjected to substrate starvation until the adsorption column shows a breakthrough. Substrate hunger significantly affects biological activity within the biofilter, and lengthier hunger sources slower acclimatization of biofilter presentation. Additionally, when significant contaminant loading fluctuations are observed, the buffer size of the adsorption unit is easily depleted. [6]

In recent years the separation of organic vapor from air streams to control VOC emissions has been extensively investigated. Gas membranes used for the separation of organic vapors have gained interest and acceptance since they selectively permeate organics over air[5]. The advantages of the gas separation membrane include simple, low energy consumption and small volume. A large amount of air flows at the feed level on one side of the membrane when the VOCs are removed and recovered from air streams at a certain point by membrane separation. When a vacuum is operating on the other side to create a partial pressure driving force, the VOCs selectively permeate through the membrane in preference to air, and the concentration of VOC in the air stream being treated is then sufficiently low to vent into the atmosphere. Vapor permeation through membranes offers significant energy savings and opportunities for recycling VOCs compared to traditional control processes, particularly when the VOC concentration is high[6]. Systems are lightweight and mobile, and can be designed to handle a wide variety of flow levels and compositions for feed streams. The separation of aromatic VOCs over specific gas membranes has been investigated[7]. There are, however, few studies on the use of gas membrane separation to maintain the performance of biofilters during transient VOC loads.[5] This paper meant to uphold continuing reliable presentation of biotic processes using a gas membrane separation module connected next a biofilter during transient VOC loadings. Separation arises when different molecules are moved at various levels through the membrane.

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The permeation of VOC via membranes in the gas membrane module was much quicker than the other off-gas components. Thus, the permeated stream becomes enriched in VOC, while the retentate stream turns into depleted air from VOC. The enriched-VOC was recharged back to the biofilter in the permeated stream, and the retentate stream can be emitted into the atmosphere depending on the extent of purification. The combination of gas separation and microbial processes resulted in increased RE, decreased sensitivity to the surge inlet load and minimized the bioreactor capacity.[2-3] This paper describes how the column for biofiltration and the module for the gas membrane operate under different conditions.

II. METHODS

Toluene, a colourless and ring oily substance primarily used in the petro - chemical and chemical manufacturing productions, subsidizes to the degradation of normal resources by the release of toluene -polluted waste and off-gases. This compound is identified by the U.S. Environmental Protection Agency in the 129 priority contaminants, and included in this analysis as the target compound.

A. *Combined Bioreactor system*

Constant experimentations were led in a packed bed reactor shadowed by a membrane reactor for monitoring toluene as a sole pollutant (Fig. 1). This bioreactor arrangement comprised a packed bed reactor and a membrane unit. Biological removal of toluene was agreed available in a acrylic packed bed computing 75cm long, and 5 cm diameter with an operative volume of 1.4 L. Pearl millet (1 cm^3) with the explicit area of 1.1 to 1.4 × 10^5 m^2 m^{-3} were worked as filling material for involved in microorganism development. Toluene was isolated from off-gas and spread over a gas membrane plate.

B. *Membrane materials*

The Tehnic Membrane System, Chennai, India, developed a commercially usable composite membrane. Polydimethylsiloxane (PDMS) was the water affinity thick upper layer , with an normal thickness of 0.3 m. Quality of the thick PDMS complex membranes developed by the Chennai, India, technical membrane system. A representation of the membrane packed bed reactor (MPBR) is exposed in Fig. 1. The MPBR, which was put at 23°C in an constant temperature chamber, was made up of two identical acyrilic compartments had four channels, 20 cm in length, 5 mm in width and 2 mm in height. The membrane was fastened in among the two sections, follow-on in a 40 cm^2 interaction region. The recirculation medium was continuously inserted via the thick membrane side, contrary to the air flow. A peristaltic pump (Miclins, India), changed the fluid flow rate to 60 cm^3 min^{-1}.

Tabulated in table 1 are the required macro and micronutrients integrated using a pH buffered nutrient solution. The flow rate of the mineral solution provided to the MPBR was balanced to retain concentrations of nitrogen > 20 mg L^{-1} in the solution for recirculation.

The individual layers of the membrane were detached by spacers. Gas feed flowed similar to the surface of the membrane, and the permeated stream entered the flow perpendicular to the membrane. The MPBR used for this procedure include polydimethylsiloxane (PDMS).

Table 1. Shows the nutrient solution

S.No	Nutrient	Weight (g/l)
1	K_2HPO_4	0.5
2	KH_2PO_4	9.0
3	$MgSO_4\,7H_2O$	0.21
4	NH_4Cl	2.0

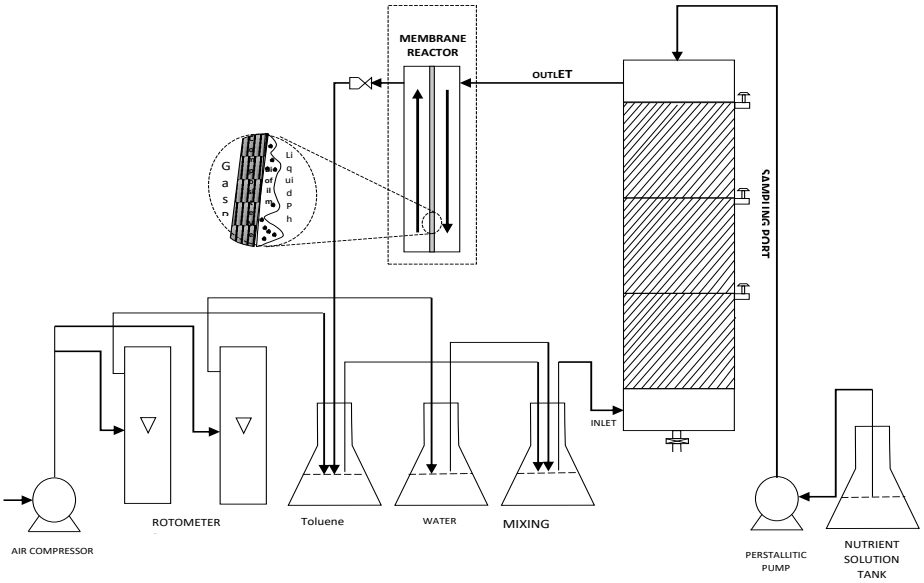

Fig. 1. Representation diagram of Membrane packed bed reactor

Firstly, toluene-containing off-gases reached the biofiltration column. Toluene was ingested by the microorganisms attached to the packaging material, and eventually biodegraded to carbon dioxide and soil. Residual toluene gasses were then fed into the module for the gas membrane. The toluene-rich stream impregnated a collection pipe through the membrane and was recharged from the permeate side by a vacuum pump back to the biofiltration column. The depleted residues of toluene were then discharged from the retentate side. Valves were placed on the permeate line so the pressure of the permeate would not reach the level of the feed. The permeated stream flow rate was 0.35 m^3h and the system air circulation rate was 30.4 percent (v: v).

The airflow had been provided by air compressors. The partial portion of air stream was bubbled through the conical flask holding pure pollutant, then mixed into the mixing chamber with a larger stream of air. This lead to in formation of pollutant desired concentration varying from 0.2 to 1.2 gm^{-3}. The wanted toluene concentration in the powerful stream was attained by adjusting the air flow speeds.

In order to find the toluene concentrations in the raw and treated source, specimen ports were positioned at the inlet, 25 cm, 50 cm, 75 cm and exit of the packed bed reactor and membrane reactor. In a laboratory, the experiment was performed with temperature variations from 23 to 32 ° C. In the biofiltration column, pH and relative humidity (RH) were measured periodically to confirm that they remained at optimum level.

Initially, the packed bed reactor was inoculated with aerobic microbial cultures obtained from a paper industry waste water treatment sludge. 10 grams of sludges was soaked in 1000 mL of nutrient solution as given in table 1. The mixture was shaked for 30 min. The concentrated sludge's solution were immunized onto the filing material (pearlmillet + polyurethane foam) of the packed bed reactor. The initial pollutant concentrations of toluene were in the range of 0.2 to 1.2g m^{-3}.

C. *Analytical methods*

The concentrations of toluene and carbon dioxide in the air stream were measured using a VOC and IR detector.

III. RESULTS AND DISCUSSION

A. *Performance of the combined packed bed reactor*

Several researches was led to test the toluene removal ability of the combined packed bed reactor system, in which the concentration of inlet toluene was wide-ranging over a duration of one year. The experiments were separated into 5 stages according to the biofilter unit's operating state as shown in table 2. The gas flow rate in the MPBR was 0.03 m^3h^{-1} for the first stage of biofilter operation.

Table 2. Shows the experimental plan

Stages	Days of operation	Gas flow rate	Inlet Toluene Concentration
I	1- 10	0.03	0.2
	11-20		0.2
	21-30		04
	31-40		0.6
	41-50		0.8
	51-60		1.0
II	61-70	0.06	0.2
	71-80		04
	81-90		0.6
	91-100		0.8
	101-110		1.0
III	111- 120	0.09	0.2
	121-130		04
	131- 140		0.6
	141-150		0.8
	151-160		1.0
IV	161-170	0.12	0.2
	171-180		04
	181-190		0.6
	191-200		0.8
	201-210		1.0

Removal of toluene in the packed bed reactor was performed in this analysis, and the membrane reactor was only worked when the column of biofiltration overwent. Illustration. 2 Shows variations in toluene concentrations in the inlet stream (C_{in}), the packed bed reactor (C_e) effluent concentration, and the membrane reactor retentate flow (Cr). Since the membrane reactor was mounted after the packed bed reactor, the packed bed reactor effluent source was also the membrane reactor feed supply. The packed bed reactor removal efficiencies (RB) and the complete removal efficiencies (R(T)) are shown in Fig. 3.

In the Figs. 2 and 3, the toluene exit concentrations varied with inlet concentrations and corresponding gas flow rate. As the microbial cultures attained from toluene-removing reactor were inoculated in the packed bed reactor, toluene RE gradually increased from 67.7 %to 89.3 %within 1 week. For the biofilter system, inoculation quickened toluene acclimation. Removal toluene output then continued at a unchanging level for the two months, i.e. the packed bed reactor was in a stable state (Stage II). In this case, the mean toluene concentration in the waste stream was 0.2 gm^{-3} while the toluene inlet concentration was just below 0.4 gm^{-3} and the mean efficiency of removal was 90.9%. When the toluene inlet concentration unexpectedly increased from 0.4 to 0.8 gm^{-3} at day 60, the toluene concentration in the biofilter unit effluent stream increased to 0.251 g m^3 and the average RE fell from 92 % to 79.8 %, suggesting that the packed bed reactor overload had exceeded its ability. In Stage III and IV, respectively, the normal concentrations of inlet toluene were held at 0.8 and 1.2 gm^{-3}.

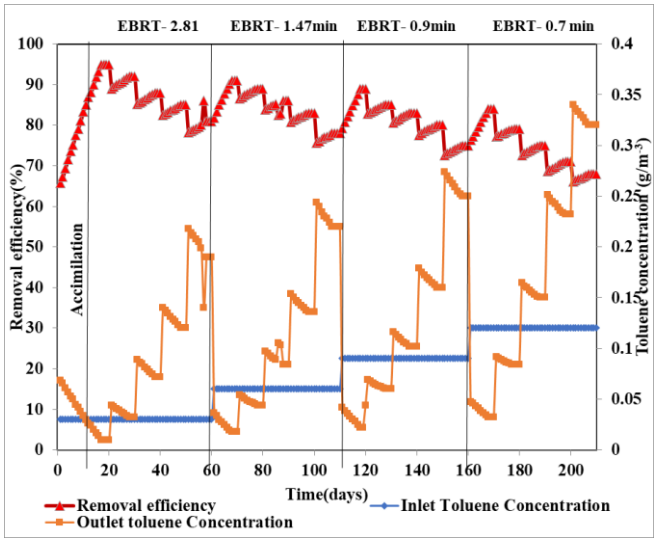

Fig. 2. Effect of inlet concentration of toluene on the Packed bed reactor.

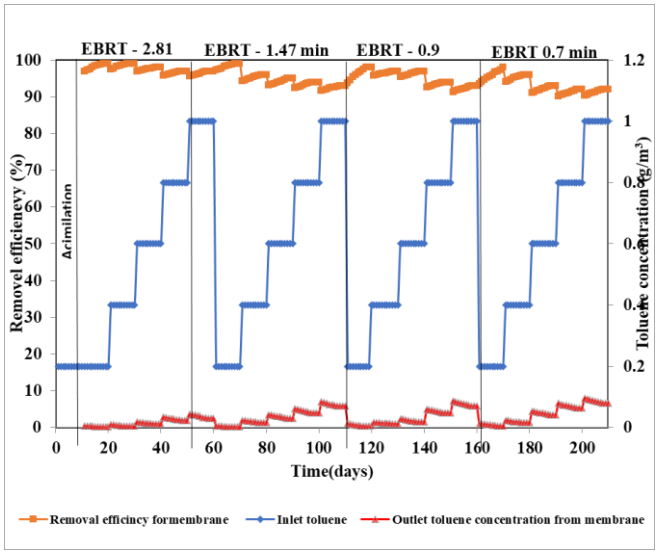

Fig. 3. Effect of inlet concentration on RE of packed bed reactor and TRE

The biofilter unit's mean RE was just 79.6 percent. Although it would be very useful to extend the volume of the biofiltration column to solve this problematic, land supplies and difficulty in service make this an antioption problem. The membrane reactor was used to diminish toluene concentration to ensure the RE. Outcomes in Figs. 2 and 3 show that the normal toluene concentration from the membrane reactor was as less as 0.4 gm^{-3} and that TRE in the MPBR was higher than 96 per cent. Residual toluene was not only isolated from, but also condensed in the gas membrane container. Most toluene in the remaining gas might be recycled to the packed bed reactor, resultant in longer processing time. Therefore MPBR with a packed bed reactor and a membrane reactor are a possible alternative to treat changing toluene loads without increasing the size of the biofilter. Such a method would lessen requirements of land dramatically if those experimental environments were extended to a large scale operation.

Packed bed reactor deletion capability was provided at Fig. 4, where the toluene removal capability versus toluene load was plotted at varying inlet concentrations. Results show that capacity to remove has been consistently increased with load. A steady and linear increase in the removal capacity was observed during Stage I, up to a toluene load of around 40 g/m^3h. Such actions suggested that a linear affiliation happened at low loading levels among elimination rate (ER) and loading rate, and that the contaminants had been almost totally removed. By more load increases during Stage III, the ER improved more gradually up to a breakthrough load and then endured steady, indicating the packed bed reactor had reached full elimination power. The packed bed reactor itself had a higher potential for toluene removal of 77.7 g $/m^3$ h. This dawn was found at 0.4gm^{-3}inlet concentration and 0.03 m3/h1 gas velocity. However, total ER risen steadily with loading rate, and a extreme value of 93.8 g/ m^3 h was found in the MPBR due to the separation of the membrane reactor.

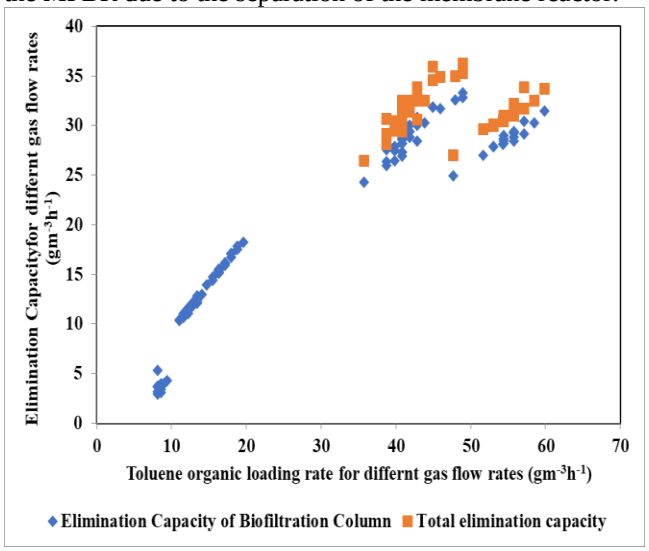

Fig. 4. ER vs. Toluene loading rate for numerous gas flow rates

The combination of packed bed reactor and membrane reactor meaningfully enhanced the RE, predominantly with higher contaminant loads. These findings are possibly useful for the manufacturing application of MPBER where membrane reactor can be mounted and operated when necessary followed by a biofilter. That, in effect, makes care as a whole much more economically feasible.

B. *Effect of bed height*

Tests suggest that 61 percent of the entire RE was linked to section 1 (the biofilter's first depth of 25 cm in bed). Sections 2 and 3 were linked to the remaining 39 percent. The RE values for sections 1, 2 and 3 were 61%, 25% and 14% respectively; because these sections were connected serially, the overall RE of the device was estimated more than 90%.

Fig. 5 Shows RE and EC values along biofilter device bed height. EC values decreased with an increase in bed height. The most significant cause of this phenomenon may be a decrease in the concentration of inlets around the biofilter surface. According to Equation (6), reduction in the concentration of the inlet induces a decrease in EC values[1-3].

This result, on the other hand, may be due to the higher microbial population in the first sections than in the other sections (it was discussed in the next section). In primary 25 cm of their biofilter under analysis.

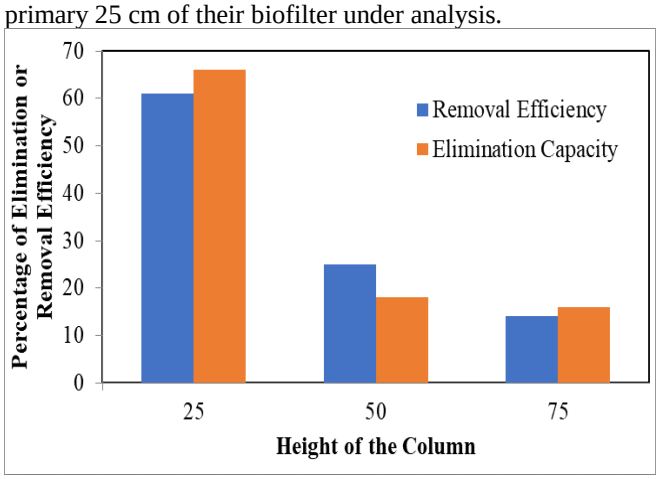

Fig.5 Effect of bed height on RC and EC

C. *Membrane reactor Influences on RE*

The efficiency of toluene parting in the membrane reactor was examined by varying concentration of inlet toluene in the feed stream (Cf) and alterations in pressure between feed and permeate sides (Pd). The toluene concentration ranged between 0.2 gm^{-3} and 0.6 gm^{-3}. The flow of feed gas (Qf), Pd and temperature were respectively 0.06 gm^{-3}, 5.0 kPa and 31°C. As shown by Fig. 6, Cr was held below 0.2 gm^{-3} if Cf was below 0.4 gm^{-3}. The membrane reactor RE was over 75 percent. However, with the rise in Cf, Cr increased significantly. Cr reached 0.2 gm^{-3} when Cf was above 0.6 gm^{-3} and RE was reduced to 55 percent. It is presumably due to the PDMS membrane's toluene permeability. Continuous increase of toluene concentration in permeated stream (Cp) under the same experimental complaint could be observed in this analysis, with the increase of Cf. Similar findings were reached in the preceding work[1-3]. Permeability of the gas is determined through a membrane by the solubility and diffusivity of the gas.

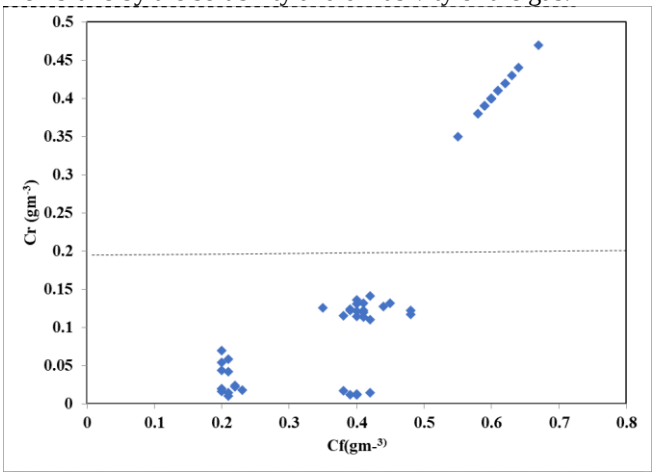

Fig. 6.Influence of toluene concentration in feed stream, Cr, concentrations of toluene in retentate stream

High toluene absorption can occur on organophilic polymers, including PDMS, which induced membrane swelling. Due to membrane swelling, the expanded interstitial spaces within the membrane between polymer chains not only support diffusion but also allow for high toluene solubility on the membranes. Hence an increase in toluene concentration in the feed stream would increase toluene permeability through the membrane.

IV. CONCLUSIONS

The Combined reactor system efficiently treated toluene charge with higher RE compared to a single biofilter. The membrane reactor improved the packed bed reactor flaws and reinforced the packed bed reactor by extending holding period and retaining ample toluene for the development of toluene degrading organisms when overcapacity happened. Following the packed bed reactor, membrane reactor can be mounted in sequence and therefore the best number of membrane reactor and variations in pressure might be approved giving to the toluene concentration from the packed bed reactor. This innovative packed bed reactor technique lays a basis for the combined regulator skill.

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