

# Kinetics and Modeling Study of Bofiltration of Toluene Vapour Using a Novel Packing Material

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Abstract :The biofilter performance was studied using the novel toluene as pollutant. The experiments were performed in a upflow mode biofilter for maintaining pollutant concentration and Empty Bed Contact Time (EBCT). Polyurethane foam as packing material with an inoculated with the paper industry sludge from waste water treatment plant. Toluene was treated effectively, with toluene influent concentrations maintained at nearly 0.2 g m<sup>-3</sup> and a total removal efficiency of over 96% achieved. The maximum elimination capacity of the bioreactor system was 93.8 g m<sup>-3</sup> h<sup>-1</sup>, which was higher than that obtained with the biofiltration column alone. Ottengraf–van den Oever model was tested and fitting demonstrated a good agreement between calculated and experimental data. The model showed a good agreement between calculated data and the physics of the process, so that it could represent a good mathematical mean for a preliminary process design

Keywords — Biofilter; Toluene, Elimination Capacity; EBCT; Removal Efficiency; Ottengraf-van den Oever model

# I. INTRODUCTION

Various wastewater streams generated in petroleum refinery process units, offsite storage area and utility areas are routed to Effluent Treatment Plant (ETP). In this process ETP receives different volatile organic compounds (VOCs) (namely benzene, ethyl benzene, toluene, xylene, methyl tertiary-butyl ether, naphthalene, phenol, styrene etc.) dissolved in wastewater and thereafter these VOCs are emitted to atmosphere due to their low boiling point and high vapor pressure at room conditions [1]. The VOCs released from ETP may show the hazardous and mutagenic effects on human and vegetation in the nearness of emission sources [2]. This VOCs released from the ETP plant cause issue in industry and government authorities major worldwide which in turn provoked the all industry to focus on treatment technologies to diminish these emissions.[3]

The sources of VOC emissions from all the industry include combustion, process operations and fugitive emissions. Also, VOCs that are leaked from the production or process and storage equipment's or pipelines contribute to the overall pollution [3]. Toluene is one of the 188 compounds regulated as a Hazardous Air Pollutant (HAP) under the 1990 Clean Air Act Amendments. This colorless liquid toluene has a sharp, sweet odor reminiscent of butterscotch and acetone. It is produced industrially on a large scale, and also occurs in trace amounts in nature. It is soluble in water and is commonly used as an industrial solvent. Toluene is an effective and common solvent and is used in processes involving gums, resins, cellulose acetate and nitrocellulose coatings and in vinyl films.

There are many different techniques available to control toluene emitted from the ETP plant. Some of the techniques are Oxidation, Adsorption, condensation, incineration and biological methods. Among these methods biofiltration is a cheap and effective alternative for VOCs elimination. [4]. Biofiltration is a process that involves a combination of different processes including adsorption, biodegradation and desorption of gas phase toxic pollutants. Biofilter also needs little nutrient addition for microbial growth with no hazardous secondary pollutants produced [5]. However, a biofilter with packing material is typically inoculated with microorganisms such as mixed bacteria and fungi strains [6],[7],[8],[9],[10].

The packing material is always selected based on the surface and pore structure and their characteristics are important for improving efficiency. The packing material may be sugarcane bagasse, pressmud, corn cob, compost, peat or peat/perlite mixture, wood chips, and other organic/inorganic commercial media materials [11],[12],[13],[14]. The biofilter performance is influenced



by a number of factors such as inlet load, air velocity, humidity, pressure drop, pore size distribution and pH of the filter bed.

Pearl millet stack is an agricultural residue generated from the separation of pearl millet and stack. This stack mainly used for animal feed and natural fertilizer. Although utilized in the many factories as fuel for the boilers, large quantities are accumulated in the mills, creating environmental problems. This paper the stack is used as raw material, which can be used as solid support. Pearl millet stack is a residue composed approximately of 50% cellulose, 25% hemicelluloses, and 25% lignin and therefore it is relatively resistant to biodegradation. In addition, the possibility of using a waste as packing material for off-gases treatment is particularly attractive.

Many researchers have reported high removal efficiencies in their lab scale biofilters for the removal BTEX under biofilter **Biofiltration** of thermophilic [15],[16]. ethylbenzene - xylene mixture was performed in a continuous biofilter employing mixed microbial culture [17]. BTEX degradation was evaluated as separate substrates and in mixtures, in liquid culture, and in packed biofilters with the filamentous fungus Paecilomyces variotii. [18]. Due to the prominence of toluene as a major VOC, many researchers have used toluene as the main carbon substrate in their biofiltration experiments [19], [20], [21]. Baskaran et al. 2016 studied the influence of substrate concentration, nutrients and temperature on the biodegradation of toluene in a compost biofilter reactor.

# **II. MATHEMATICAL MODELLING**

Biofiltration processes involve the passage of the pollutant from the gas to the water phase and then into the biofilm, and mathematical description of VOC biodegradation in a biofilter was to couple biofilm models with the equations describing the mass transfer Processes in the biofilter [22]. In early days, the biofilm models for gas phase biofilters were based on biofilm models for waste water treatment. Ottengraf and van de Oever (1983) developed a model based on the biodegradation of non-adsorbable substrates in submerged biofilms [23]. From the ottengraf model Devinny et al. (1999) presented a review on biofilter models. In gas-phase biofiltration process modeling, zeroorder, first-order, or Monod kinetic model were often used. Many factors could be incorporated in recent models. Alonso et al (1997,1998) incorporate the growth of biomass and the increase in biofilm thickness in vapor-phase biofilters. Nukuny et al 2005 also divided total biomass into active and inactive biomass. An empirical model was also reported to predict biofilter performance for toluene removal [7]. Saravanan et al., 2017 validate the two situations of the reaction. The aim of the experiment was to evaluate the feasibility of using polyurethane foam as packing materials for the biofiltration of toluene vapor. The obtained experimental results are also validated with the Ottengraf–van-den Oever and modified Ottengraf–van-den Oever model for various phases.

The main objective of the present work is to evaluate the performance of biofilter having polyurethane foam and berl saddles as media bed while treating toluene vapors at varied concentrations. The removal capacity and removal efficiency were studied and monitored under different operating situations. Further, the experimental values are compared with those predicted by the Ottengraf's model and modified ottengraf model.

# **III.** MATERIALS AND METHODS

# A. Microorganism and Culture Media Used

The microbial mixed culture obtained form a paper industry wastewater treatment plant was acclimatized with toluene as the carbon source in a mineral salt medium as shown in table 1. The pH of the mineral salt media was adjusted to 6.5 and the cultures were grown under ambient conditions in a rotary shaker.

<b>Table 1.</b> Shows the nutrient solution	Та	ble 1	. Shows	the	nutrient	solution
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	S.No	Nutrient	Weight (g/l)
E	AIV	K <sub>2</sub> HPO <sub>4</sub>	0.5
t	2	KH <sub>2</sub> PO <sub>4</sub>	9.0
,	3 Appli	MgSO <sub>4</sub> 7H <sub>2</sub> O	0.21
in Eng	gineerang m	NH <sub>4</sub> Cl	2.0

# B. Toluene

Toluene, a colorless and aromatic oily liquid used predominately in the petrochemical and polymer-processing industries, contributes to natural resource pollution via the release of styrene-contaminated effluents and off-gases. This compound is listed in the 129 priority pollutants by the Environmental Protection Agency in the USA, and was used as the target compound in this study.





# Fig. 1. The schematic diagram of the integrated bioreactor.

#### C. Biofilter system

In this study, the polyurethane foam was used as a packing medium. The packing media was cut along the length of the biofilter. The biofilter was made from a height of 1m cylindrical polymethylacrylate column with an inner diameter of 0.05 m, and filled to a height of 0.75m with the packing media inoculated with activated sludge as shown in figure 1. The activated sludge was placed for 20 min, and then the supernatant liquor was removed. The residual activated sludge suspension was used as inoculum. The volume amount of activated sludge suspension used depended on the final water content of packing media, and the water content of packing media was generally maintained at about 50%. Compressed air was passed first through an activated carbon filtration device to remove moisture, oil and particulate matter. The air filtered was split into two air fractions. The major portion of air was humidified in a water humidifier to ensure that the air relative humidity was more than 95%. The minor portion of air was allowed to bubble through liquid toluene container to generate the contaminated air stream. Then these two air streams were mixed in an air mixer, and fed to the bottom of the biofilters in upflow mode of operation. The flow rates were controlled by valves and metered by previously calibrated flowmeters to obtain the desired toluene inlet concentration and gas residence times in the filter bed. The nutrient solution was continuously sprayed with about 0.1 L  $\min^{-1}$  in biofilter for 30 min each day to ensure satisfactory

conditions of moisture and nutrients for microorganism's activity.

# D. Biofilter Operation

Experiments were performed for a period of 200 days. The experimental operation was divided into four periods (I, II, III and IV) according to Empty Bed Residence Time (EBRT). The operating conditions of each period were summarized in table 2. The inlet concentration of pollutant was varied from 0.2 to 1.2 gm<sup>-3</sup>. The EBRT was varied from 40 to 168 s.

Table 2.	Shows	the	experimental	plan
			1	1

Stagos	Days of	Gas flow	Inlet Toluene
Stages	operation	rate	Concentration
	1-10		0.2
	11-20		0.2
т	21-30	0.02	04
1	31-40	0.05	0.6
	41-50		0.8
	51-60		1.0
	61-70		0.2
	71-80		04
П	81-90	0.06	0.6
	91-100		0.8
	101-110		1.0
	111-120		0.2
	121-130		04
Ш	131- 140	0.09	0.6
	141-150		0.8
	151-160		1.0
	161-170		0.2
	0171-180		04
ĪV	181-190	0.12	0.6
1 pplica	191-200		0.8
	201-210		1.0

# E. Analytical methods

Toluene and carbon dioxide concentrations in the air stream were measured using a VOC and IR detector detector respectively.

# F. Biofilter Terminology

To describe the mechanisms of biofiltration clearly, general terminology pertinent to the field should be well defined. Biofiltration involves chemistry, microbiology, physics, fluid dynamics, and mathematics. The first works published on biofiltration establishes a common terminology facilitating communication and comparison among the various processes. These terminologies, with the most common units used, are defined in Table 3.

# IV. RESULTS AND DISCUSSION

#### A. Performance of the biofilter

The biofiltration of gas stream containing pure benzene is carried out for 200 days at various operating conditions in an up flow mode biofilter. The experiment has been operated in three stages. Each stage is divided in to three phases as shown in table 2. Various EBCT and concentration are maintained so that the corresponding loading rate could be maintained and regulated in the reactor to study the performance of the reactor. Table 2 shows the inlet concentration ( $C_{in}$ ), outlet concentration ( $C_{out}$ ), inlet gas flow rate (F). Volume of the reactor (V) supplied to the biofilter.

#### Table 3 Performance equation of biofilter

S.No	Term	Equation	Unit
1	Removal	$C_{in} - C_{Out \mathbf{V}_{100}}$	%
	Efficiency	$\frac{1}{C_{in}}$ X100	
2	Inlet Loading	QC <sub>in</sub>	g/m <sup>3</sup> h
	rate	V	
3	Elimination	$F(C_{in} - C_{Out})$	g/m <sup>3</sup> h
	Capacity	V	
4	EBCT	v	Mins
		$\overline{\mathbf{Q}}$	
		Inter	
100	EBRT - 2.81 EBR	T - 1.47 EBRT - 0.9	EBRT 0.7
90		man	
80			
(%) 70			0.80
cienevy 09	- imilati		R <sub>esearch</sub>
j 50 19 40	¥ Y		Ouceu
Semon 30		╵	0.4
20			e 0.2
10			(manual frances)
0	0 20 40 60 8	0 100 120 140 160	0 180 200
		Time(days)	
-	-■- Removal efficency -→- In	let toluene ——Outlet toluene conc	centration

# Fig. 2. Effect of removal efficiency toluene by varying different inlet concentration and different EBRTs

B. Performance of biofilter at an EBRT of 2.81 min

During the first 60 days, the EBCT is maintained at 2.81 min as shown in figure 2. In the first phase of stage - I, toluene inlet concentration is maintained at  $0.2 \pm 5 \%$  g/m<sup>3</sup>, so that the average loading rate of 4.163 g/m<sup>3</sup> h can be applied to the reactor. The corresponding gas flow rate is maintained at 0.03 m<sup>3</sup>/h. Continuous increase in removal efficiency is as shown in figure 2. It is clear from the figure

2 that a maximum removal efficiency of 96% removal is obtained after 8 days of operation. The results showed that the maximum removal efficiency of toluene is 95%.

In phase II, average loading rate has been increased by almost twofold from 4.163 to 8.143 g/m<sup>3</sup> h. The flow rate and EBRT is kept at 0.03 m<sup>3</sup>/h and 2.81 min respectively. The average inlet toluene concentration is sustained at  $0.4 \pm 5 \%$  g/m<sup>3</sup>. Due to this shock load there is a fall in the RE of toluene. A maximum RE of toluene is 88 %. In phase III, average loading rate is increased from 8.143 to 12.857 g/m<sup>3</sup> h and the flow rate of main stream and EBCT is 0.03m<sup>3</sup>/h and 2.81 min respectively. The inlet concentration is kept constant at 0.8 g/m<sup>3</sup>. With the sudden increase in the loading rate to the reactor, the RE decreased from 88% to 81%.

In phase IV, average loading rate is increased from 12.857 to 16.323 g/m<sup>3</sup> h and the flow rate of main stream and EBRT is  $0.03m^3/h$  and 2.81 min respectively. The inlet concentration is kept constant at 0.8 g/m<sup>3</sup>. With the sudden increase in the loading rate to the reactor, the RE decreased from 81% to 78%. In phase V, average loading rate is increased from 16.323 to 20.408 g/m<sup>3</sup> h and the flow rate of main stream and EBCT is  $0.03m^3/h$  and 2.81 min respectively. The inlet concentration is kept constant at 1.2 g/m<sup>3</sup>. With the sudden increase in the loading rate to the reactor, the RE decreased from 78% to 75%.

# C. Performance of biofilter at an EBRT of 1.47 min

During the second 50 days, the EBCT is maintained at 1.47 min as shown in figure 2. In the second phase of stage - I, toluene inlet concentration is maintained at  $0.2 \pm 5 \%$  g/m<sup>3</sup>, so that the average loading rate of 8.163  $g/m^3$  h can be applied to the reactor. The corresponding gas flow rate is maintained at 0.06 m<sup>3</sup>/h. Continuous increase in removal efficiency is as shown in figure 2. It is clear from the figure 2 that a maximum removal efficiency of 92% removal is obtained after 8 days of operation. The results showed that the maximum removal efficiency of pure toluene is 92%. In phase II, average loading rate has been increased by almost twofold from 8.143 to 16.323 g/m<sup>3</sup> h. The flow rate and EBCT is kept at 0.06  $\text{m}^3/\text{h}$  and 1.47 min respectively. The average inlet toluene concentration is sustained at 0.4  $\pm$  5 %  $g/m^3$ . Due to this shock load there is a fall in the RE of toluene. A maximum RE of toluene is 83 %.

In phase III, average loading rate is increased from 16.857 to 24.848 g/m<sup>3</sup> h and the flow rate of main stream and EBCT is  $0.06m^3/h$  and 1.47 min respectively. The inlet concentration is kept constant at 1.0 g/m<sup>3</sup>. With the sudden increase in the loading rate to the reactor, the RE decreased from 83% to 80%.

In phase IV, average loading rate is increased from 24.848 to 32.636 g/m<sup>3</sup> h and the flow rate of main stream and EBCT is  $0.03m^3/h$  and 2.81 min respectively. The inlet concentration is kept constant at 0.8 g/m<sup>3</sup>. With the sudden



increase in the loading rate to the reactor, the RE decreased from 80% to 76%. In phase V, average loading rate is increased from 32.636 to 48.818 g/m<sup>3</sup> h and the flow rate of main stream and EBCT is  $0.06m^3/h$  and 2.81 min respectively. The inlet concentration is kept constant at 1.2 g/m<sup>3</sup>. With the sudden increase in the loading rate to the reactor, the RE decreased from 75% to 72%.

#### D. Performance of biofilter at an EBRT of 0.9 min

During the third 50 days, the EBCT is maintained at 0.9 min as shown in figure 4. In the third phase of stage - I, toluene inlet concentration is maintained at  $0.2 \pm 5$  % g/m<sup>3</sup>, so that the average loading rate of 12.249 g/m<sup>3</sup> h can be applied to the reactor. The corresponding gas flow rate is maintained at 0.09 m<sup>3</sup>/h. Continuous increase in removal efficiency is as shown in figure 2. It is clear from the figure 2 that a maximum removal efficiency of 89% removal is obtained after 8 days of operation. The results showed that the maximum removal efficiency of pure toluene is 89%. In phase II, average loading rate has been increased by almost twofold from 12.249 to 24.49 g/m<sup>3</sup> h. The flow rate and EBCT is kept at 0.09 m<sup>3</sup>/h and 0.9 min respectively. The average inlet toluene concentration is sustained at  $0.4 \pm 5$  % g/m<sup>3</sup>. Due to this shock load there is a fall in the RE of toluene. A maximum RE of toluene is 85 %. In phase III, average loading rate is increased from 24.848 to 36.736  $g/m^3$  h and the flow rate of main stream and EBCT is 0.09m<sup>3</sup>/h and 0.9 min respectively. The inlet concentration is kept constant at 1.0 g/m<sup>3</sup>. With the sudden increase in the loading rate to the reactor, the RE decreased from 85% to 80%.

In phase IV, average loading rate is increased from 36.736 to  $48.78 \text{ g/m}^3$  h and the flow rate of main stream and EBCT is  $0.09\text{m}^3$ /h and 0.9 min respectively. The inlet concentration is kept constant at  $0.8 \text{ g/m}^3$ . With the sudden increase in the loading rate to the reactor, the RE decreased from 80% to 76%. In phase V, average loading rate is increased from 48.78 to  $61.24 \text{ g/m}^3$  h and the flow rate of main stream and EBCT is  $0.09\text{m}^3$ /h and 0.9 min respectively. The inlet concentration is kept constant at  $1.2 \text{ g/m}^3$ . With the sudden increase in the loading rate to the reactor, the RE decreased from 48.78 to  $61.24 \text{ g/m}^3$  h and the flow rate of main stream and EBCT is  $0.09\text{m}^3$ /h and 0.9 min respectively. The inlet concentration is kept constant at  $1.2 \text{ g/m}^3$ . With the sudden increase in the loading rate to the reactor, the RE decreased from 76% to 72%.

#### E. Performance of biofilter at an EBCT of 0.7 min

During the fourth 50 days, the EBCT is maintained at 0.7 min as shown in figure 2. In the fourth phase of stage - I, toluene inlet concentration is maintained at  $0.2 \pm 5 \%$  g/m<sup>3</sup>, so that the average loading rate of 16.323 g/m<sup>3</sup>h can be applied to the reactor. The corresponding gas flow rate is maintained at 0.12 m<sup>3</sup>/h. Continuous increase in removal efficiency is as shown in figure 1 It is clear from the figure 1 that a maximum removal efficiency of 84% removal is obtained after 8 days of operation. The results showed that the maximum removal efficiency of toluene is 84%. In phase II, average loading rate has been increased by almost

twofold from 16.323 to 32.656 g/m<sup>3</sup> h. The flow rate and EBCT is kept at 0.12 m<sup>3</sup>/h and 0.7 min respectively. The average inlet toluene concentration is sustained at  $0.4 \pm 5$  % g/m<sup>3</sup>. Due to this shock load there is a fall in the RE of toluene. A maximum RE of toluene is 82 %. In phase III, average loading rate is increased from 32.656 to 48.979 g/m<sup>3</sup> h and the flow rate of main stream and EBCT is 0.12 m<sup>3</sup>/h and 07 min respectively. The inlet concentration is kept constant at 1.0 g/m<sup>3</sup>. With the sudden increase in the loading rate to the reactor, the RE decreased from 82% to 78%.

In phase IV, average loading rate is increased from 48.979 to 65.30 g/m<sup>3</sup> h and the flow rate of main stream and EBCT is 0.12 m<sup>3</sup>/h and 0.78 min respectively. The inlet concentration is kept constant at 0.8 g/m<sup>3</sup>. With the sudden increase in the loading rate to the reactor, the RE decreased from 78 % to 74%. In phase V, average loading rate is increased from 65.30 to 81.6 g/m<sup>3</sup> h and the flow rate of main stream and EBCT is 0.12 m<sup>3</sup>/h and 0.7 min respectively. The inlet concentration is kept constant at 1.2 g/m<sup>3</sup>. With the sudden increase in the loading rate to the reactor, the RE decreased from 74% to 70%.

# F. Inlet load and Elimination Capacity effects

Pollutant elimination capacities for the different runs versus IL are presented in figure 3. A comparison of the EC of toluene as a function of inlet load for different EBCT of 2.8, 1.47, and 0.7 min is shown in Figures 3. A maximum EC of 49.33 g/m<sup>3</sup> h toluene is observed at a gas flow rate of 1 m<sup>3</sup>/ h (EBRT = 0.7 min). When increasing the gas flow rate the elimination capacity also shows positive trend. From the figures 3, it is observed that the polyurethane foam based biofilter tended to operate with slightly higher removal capacities. It suggests that, increase in inlet load enhances the transfer rate of toluene to the biofilm at constant gas flow rate. However, at higher inlet load, the elimination capacity curve deviated markedly from the 100% removal curve. Similar observation is obtained for the removal of toluene, ethylacetate and xylene vapour using perlite as porous material [25-27].



Fig. 3. Elimination capacity vs. Toluene inlet load for various gas flow rates

# *G.* Effect of Empty bed residence time on biofiltration operation

The influence of residence time on the removal efficiency, at different inlet load has been studied. All the results are shown in Figure 4. The maximum toluene elimination is observed at an EBCT of 2.81 min. A decrease in the EBCT from 2.81 to 0.7 min caused a decrease in the removal efficiency from 92% to 50%. But for residence times less than 1.47 min, a sharp linear decrease is observed, achieving a removal efficiency of 50% at 0.7 min. These results suggest that a minimum EBCT of about 1 min may be needed to reach adequate removal efficiencies, and an EBCT of 1.47 min could be considered safe to ensure the absence of limitations associated with contact time. Similar trend was observed for the different ratios of corn stack and pressmud [27]. A maximum removal efficiency of 98% is observed in pure pressmud based biofilter.

# V. APPLICATION OF THE THEORETICAL MODEL

The outlet concentration of toluene, in the situation of diffusion limitation can be described by the following equation:

(1)

$$\sqrt{C_i} = \sqrt{C_o} - k_1 \frac{V}{Q}$$

Hence, in the case of diffusion limitation, the validity of the theoretical model can be checked by plotting  $\sqrt{C_i}$  versus for the range of inlet concentrations ( $\sqrt{C_o}$ ) for which the EC is less than the k<sub>o</sub>. The reaction limitation behavior is attained at a level of pollutant load that corresponds, at a given gas flow rate, to the critical inlet concentration at which the biofilter behavior is in transition between the diffusion and the reaction limitation. Therefore, the critical concentration of toluene can be estimated from the following relationship

$$EC = \frac{Q}{V}C_{o,Crit}L\left(1 - \left(1 - k_1 \frac{V}{Q} \frac{1}{\sqrt{C_{o,crit}}}\right)^2\right) = k_o$$
 (2)

Hence,

$$C_{o,Crit} = \frac{1}{4} \left( \frac{k_o}{k_1} + \frac{k_1 V}{Q} \right)^2$$
 (3)

Whe model is tested for the biofiltration of toluene using poly urethane foam based biofilter. For this packing material, the plot has been displayed the increasing elimination capacity with toluene loading rate can be defined by the diffusion determining step, and at the reaction determining step make behaved. Thus, diffusion determining step is effective for low concentrations and theoretical reaction limitation model seems to be valid for high concentrations of toluene in the biofilter. The values of model parameters, kinetic constants and maximum EC for at different operating conditions were tabulated in Table 4.



Fig4 Effect of EBCT for removal efficiency of toluene

The biofilm thickness was also calculated for different phases by taking the values of effective diffusivity of biofilm (D) and Henry's constant (m) for toluene as 1.048 X  $10^{-6}$  m<sup>2</sup> h<sup>-1</sup> and 0.00258 respectively. The values of biofilm thickness were reported in Table 3. An increasing trend was observed for the biofilm thickness for different phases.

A. Modified Ottengraf model

In the Ottengraf model propose two regimes for two different conditions. One for reaction limitation area and the other for the diffusion limitation area; the transition between the two conditions is ruled by the Thiele number. This new model considers both diffusion and reaction limitations as a single equation.

Table 4.	Model	parameters	and	kinetic	constants	at
various o	perating	conditions				

GF	K <sub>1</sub>	K <sub>d</sub>	K <sub>0</sub>	С	IL	δ
( <b>m<sup>3</sup>h<sup>-</sup></b> <sup>1</sup> )	(gm <sup>-</sup> <sup>3</sup> h <sup>-1</sup> )	(gm <sup>-</sup> <sup>3</sup> h <sup>-1</sup> )	(gm <sup>-</sup> <sup>3</sup> h <sup>-1</sup> )	critical (gm <sup>-</sup> <sup>3</sup> )	critical (gm <sup>-</sup> <sup>3</sup> )	(µm)
0.03	0.771	0.292	17.21	0.945	23	272
0.06	0.745	0.345	31.45	0.942	42	300
0.09	0.731	0.281	69.2	0.91	69	321
0.12	0.727	0.291	72.9	0.9	80	399

# B. Fundamentals of the new model

The new model individuates two different phenomena, ruling and determining the rate of the biofiltration process.



At low load values, diffusion is the rate determining step and, in such conditions, the elimination capacity is given by the following equation:

$$EC_{dL} = L \left( 1 - \left( 1 - A_s \sqrt{\frac{k_0 D}{2m}} \sqrt{\frac{V}{QL}} \right)^2 \right)$$
(4)

where the index dl stands for diffusion determining step. Otherwise, at high loads, the removal of the toluene is mainly influenced by the biological reaction and the elimination capacity is load-independent.

$$EC_{rl} = EC_{max} = A_s k_0 \delta$$
 (5)

But, having the use of one equation, only that can continuously connect the different expression of  $EC_{dl}$  and  $EC_{rl}$  can be very useful for biofiltration design. The following equation can satisfy this condition

$$EC = EC_{max} + \frac{\left(EC_{dl} - EC_{max}\right)}{1 + \left(\frac{L}{L^*}\right)^p}$$
(6)

where  $L^*$  is the load at which the transition between reaction and diffusion limitation occurs. Parameter p was calculated by fitting of the experimental data. Its value agrees the rate at which the channel between the two different regimes conditions occurs.

$$\eta = \frac{C_{i} - C_{o}}{C_{i}} = \frac{EC}{L} = \left| EC_{max} + \frac{\left(EC_{dl} - EC_{max}\right)}{1 + \left(\frac{L}{L^{*}}\right)^{p}} \right| / L \quad (7)$$

1

With some arithmetical steps and using the definition of L and EC, it is also possible to write efficiency and  $C_0$  as a function of  $C_i$ :

where C\* is the inlet concentration at which load is equal to the L\*, at constant flow rate and volume. The new model as inlet load increases, limitations caused by diffusion reduce and the ones caused by reaction become stronger. The original Ottengraf's model and the modified model were compared with the experimental data and it was depicted in Figure. 5 and 6.

$$\eta = \frac{\left(A_{s}k_{o}\delta + \frac{\frac{C_{i}Q}{V}\left(1 - \left(1 - A_{s}\frac{V}{Q}\sqrt{\frac{k_{o}D}{2mC_{i}}}\right)^{2}\right)\right)}{1 + \left(\frac{C_{g,in}}{C_{g}^{*}}\right)^{p}}\right)}{C_{i}Q} \qquad (8)$$

$$C_{o} = C_{i} - \left[\frac{Q.A_{s}k_{o}\delta}{V} + \frac{C_{i}\left(1 - \left(1 - A_{s}\frac{V}{Q}\sqrt{\frac{k_{o}D}{2mC_{i}}}\right)^{2}\right) - A_{s}k_{o}\delta}{1 + \left(\frac{C_{i}}{C^{*}}\right)^{p}}\right] (9)$$



Fig. 5 Comparison of ottengraf model and modified ottengraf model with experimental values for toluene removal in a polyurethane foam based biofilter.



# Fig. 6 Comparison of experimental and model predicted values for RE of toluene using polyurethane foam based biofilter

Fitting was carried out for IL vs EC was shown in Figure. 5. Figure. 6, report the model fitting for RE vs inlet loading rate for the biofilter. It shows a good agreement between experimental and calculated data. The transition value between diffusion and reaction limitation (Critical Inlet Load) area were given in Table 4. In spite of all the limits encountered and discussed, the new model has a good agreement with the experimental data.

# **VI.** CONCLUSION

The polyurethane foam based biofilter was evaluated for the biofiltration of toluene vapours. The hgh toluene concentration values up to 1.2 g m<sup>-3</sup> have been treated, with a maximum elimination capacity of 95 g m<sup>-3</sup> h<sup>-1</sup> was achieved. The EBCT of 0.2 min has been established as the minimum operational. The polyurethane foam based biofilters exhibited a better performance in terms of elimination capacity and long-term stability. Ottengraf–van den Oever model was tested and fitting demonstrated a good agreement between calculated and experimental data.



The model showed a good agreement between calculated data and the physics of the process, so that it could represent a good mathematical mean for a preliminary process design

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