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Comparison of Agro Industries Packing Material for Treating MEK Vapor Using Biofilters: Kinetics and Modelling

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ABSTRACT

Packing materials play a key role in the performance of bioreactors for waste gas treatment and particularly in biofilter applications. In this work, the performance of two biofilters packed with different packing materials, operated in parallel for the treatment of relatively high inlet concentration of MEK was studied. The reactors were compared for determining the suitability of cornstack and press mud as packing materials for biofiltration of MEK. Biofilters achieved maximum removal efficiency (RE) of 98 and 95 % throughout its operation at an EBRT of 2.8 min for an inlet concentration of 0.2 gm^{-3} with press mud and cornstack based biofilter respectively, which is quite significant than the values reported in the literature. Elimination capacities of MEK increased with the increase in influent MEK loading, but an opposite trend was observed for the removal efficiency for the biofilter. In general, press mud exhibited a better performance than cornstack in terms of elimination capacity and removal efficiency. The experimental results for both packing materials were compared with the values obtained from the Ottengraf–Van Den Oever model for zero-order diffusion-controlled region. The critical inlet concentration, critical inlet load and biofilm thickness were estimated using the model predictions.

1. Introduction

Biofiltration is currently the most used biological gas treatment technology. It involves microorganisms immobilized in the form of a biofilm on a porous carrier, such as, peat, soil, compost, synthetic substances or combinations of them. The carrier provides to the microorganisms a favorable environment in terms of pH, temperature, moisture, nutrients and oxygen supply. As the polluted air stream passes through the filter bed, pollutants are transferred from the vapor phase to the biofilm developing on the organic substrate. The microorganisms metabolize the pollutants almost all organic compounds can be used as biofilter carrier [1]. Bohn listed 13 important physical, chemical and biological characteristics of the filter media with the most important being (i) large specific surface area, (ii) low bulk density, (iii) a high void fraction, (iv) large number of different bacteria naturally present in the carrier, (v) sufficient nutrients, i.e. N, P and K, (vi) large Water Holding Capacity and (vii) a neutral or alkaline pH as well as buffer capacity [1].

Press mud is an agricultural residue generated from industrial sugar extraction process. Although utilized in the sugar factories as fuel for the boilers, large quantities are accumulated in the mills, creating environmental problems. Recently, there is an increasing trend towards a large and inexpensive source of raw material, which can be used as solid support also in several biotechnological processes [2]. Press mud 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.

MEK is one of the 188 compounds regulated as a Hazardous Air Pollutant (HAP) under the 1990 Clean Air Act Amendments [3]. The U.S. EPA lists MEK as one of the top 20 chemicals in terms of largest total on-site and off-site releases in 1999, with 40,720,712 pounds in total releases (US EPA, 2001). Methyl ethyl ketone or MEK, also known as, butanone is an organic compound with the formula $CH_3C(O)CH_2CH_3$. This colorless liquid ketone 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 [4]. It is soluble in water and is commonly used as an industrial solvent. MEK is an effective and common solvent and is used in processes involving gums, resins, cellulose acetate and nitrocellulose coatings and in vinyl films [5]. For this reason it finds use in the manufacture of plastics, textiles, in the production of paraffin wax, and in household products such as lacquer, varnishes, paint remover, a denaturing agent for denatured alcohol, glues, and as a cleaning agent. It has similar solvent properties to acetone but boils at a higher temperature and has a significantly slower evaporation rate [3]. Butanone is also used in dry erase markers as the solvent of the erasable dye. Butanone is an irritant, causing irritation to the eyes and nose of humans [3]. Serious health effects in animals have been seen only at very high levels. These included skeletal birth defects and low birth weight in mice, when they inhaled MEK at the highest dose tested (3000 ppm for 7 hours/day).

Recently reported research using methyl ethyl ketone (MEK) as a model contaminant demonstrated that this approach can be used to achieve higher overall removal efficiency and higher minimum instantaneous removal efficiency than can be achieved by a continuous flow system. Deshusses *et al* studied the behavior of biofilters in the treatment of an air stream contaminated with MEK by equivolume mixture of compost and polystyrene spheres. Two EBRT's, 90 and 45 seconds, were tested over a loading rate range of 0 – 350 $gm^{-3}h^{-1}$[6].

Amanullah *et al* performed on MEK using two types of support media, compost and granular activated carbon (GAC), were evaluated. The experimental procedure used EBRT's ranging from 25 to 50 seconds and an MEK influent concentration 1.1869 g/m^3. Reported removal efficiencies ranged from 25 to 30% under the conditions tested [7]. Chou and Huang was used to study the two types of packing materials, polypropylene spheres and wood bars, were tested in reactors with a treatment volume of 0.141 m^3. Influent MEK concentrations ranging from 0.9 to 5 g/m^3 were tested. Removal efficiencies ranging from 40 to greater than 97% were reported [8].

1.1 Mathametical Modeling with Ottengraf–Van Den Oever Model

Most of the studies conducted on biofiltration utilize bacterial strains, either pure or that are isolated from the filtering media, suspended in liquid growth media. The drawbacks of these methods are that (1) they necessitate prior operations for the conditioning of the biomass; (2) they do not necessarily represent the real growth media (the solid bed pellets), which more likely contain consortia of interacting micro-organisms,

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among them the degrading species; and (3) they do not reflect the mass transfer constraints that exist in a biofilter. To date, a few works only have focused on the experimental protocols for application to solid growth media [9,10]. Since many different phenomena contribute to the effectiveness of a biofiltration process, a model has to be used which can comprehensively foresee bioreactor performance. Ottengraf and Van den Oever (1983) made first attempt to develop a model for the biofiltration of toluene. This model simply deals with conventional biofilter at stationary state [11]. In spite of its simplicity, this model has been widely used also by others [12]. Ottengraf's model considers the different phenomena ruling biofilter performance: mass transfer and biological reaction. At low inlet concentrations, the driving force ruling the mass transfer is limited. Therefore, the amount of pollutant which passes into the liquid phase is moderate and, as pollutant comes in contact with the biomass, it is completely degraded. In these conditions, diffusion is the rate determining step. With higher gas concentrations, mass transfer is inversely promoted. The amount of pollutant transferred in the aqueous phase is greater and biomass could not be able to completely degrade this amount. In such conditions, the reaction limits the process rate. Ottengraf proposed equations to represent what occurs in the water film in these two opposite situations.

1.2 Mass Balance

Pollutant concentration in the gas phase can be expressed by the following expression:

$$-U_g \frac{dU_g}{dh} = NA_s \quad (1)$$

where U_g is the superficial gas velocity (mh^{-1}), h is the reactor height (m), N is the flux of substrate from the gas to the solid ($gm^{-2}h^{-1}$) and A_s is the specific surface area (m^2m^{-3}).

Mass balance in the gas/biofilm can be written as follows:

$$D\frac{d^2C}{dx^2} - k_o = 0 \quad (2)$$

where D is the diffusion coefficient (m^2h^{-1}), x is the direction perpendicular to the gas-solid interface and k_0 the zero-order constant ($gm^{-3}h^{-1}$). Such equations can be solved considering the different boundary conditions in reaction limitation and diffusion limitation assumptions.

Zero – Order Kinetics with Reaction Limitation

In this condition, introducing 'm' as the dimensionless gas-solid partition coefficient, the following boundary conditions can be used:

$$x = 0, \quad C = C_g/m \quad (3)$$

$$x = \delta, \quad dC/dx = 0 \quad (4)$$

and equation (2) has the following solution:

$$\frac{C}{C_g/m} = 1 + \frac{1}{2}\frac{\Phi^2}{C_i/C_o}\left(\sigma^2 - 2\sigma\right) \quad (5)$$

Where Thiele number; σ = x/δ is the dimensionless length coordinate in the biolayer; and *m(* = $(C_g/C_l)_{equilibrium}$) is the distribution coefficient.

$$\phi = \sqrt{\frac{k_m}{D^{'}C_{go}}} \quad (6)$$

Then, N can be written as

$$N = \frac{-D^{'}}{\delta}\left(\frac{dC_l}{d\sigma}\right)_{\sigma=0} = k\delta \quad (7)$$

Substituting equation (7) into equation (1) using the boundary condition $C_g = C_g$, in for h= 0, the solution becomes:

$$\frac{C_o}{C_i} = 1 - \frac{A_s k_o \delta H}{C_i U_g} \quad (8)$$

where H is the height of the tower. Assuming $A_s k_o \delta$ = K to be constant, it follows that

$$\eta = 1 - \frac{C_o}{C_i} = \frac{A_s k_{o\delta} H}{C_{g,in} U_g} \quad (9)$$

Elaborating equation (8), and solving as function of the elimination capacity, the following expression can be obtained [12]

$$EC = EC_{max} = A_s k_o \delta \quad (10)$$

A critical point can be determined, supposing that C=0 at the water-solid interface, or when x = δ Substituting this value into equation (5), a critical Thiele number can be determined:

$$\phi = \delta\sqrt{\frac{k_o m}{DC_i}} = \sqrt{2} \quad (11)$$

When φ < Φ cr, reaction is the rate determining step of the process.

Zero- Order Kinetics with Diffusion Limitation

Mass balance into the (Air/biofilm) phase should be now solved using different boundary conditions. Defining ë as the distance from the interface gas/liquid at which C = 0, boundary condition (4) can be substituted by the following:

$$x = \lambda \quad dC/dx = 0 \quad (12)$$

Obtaining a new equation for the water phase:

$$\frac{C}{C_o/m} = 1 + \frac{1}{2}\frac{\Phi^2}{C_i/C_o}\left(\sigma^2 - 2\sigma\frac{\lambda}{\delta}\right) \quad (13)$$

$$\lambda = \sqrt{2\frac{D^{'}C_o}{km}} \quad (14)$$

λ can be easily determined with equation (13), fixing C=0 for ó = λ/δ: With this new condition, N = k_0λ and pollutant concentration in the gas phase can be calculated:

$$\frac{C_o}{C_i} = \left(1 - \frac{A_s H}{U_g}\sqrt{\frac{k_o D}{2C_i m}}\right) \quad (15)$$

EC is now a function of the mass loading rate and the correlation is represented by the following expression:

$$EC = L\left(1 - \left(1 - A_s\sqrt{\frac{k_o D}{2m}}\sqrt{\frac{V}{QL}}\right)^2\right) \quad (16)$$

The aim of the experiment was to evaluate the feasibility of using corn stack and press mud as packing materials for the biofiltration of MEK vapor. The obtained experimental results are also validated with the Ottengraf–van-den Oever and modified Ottengraf–van-den Oever model for various phases.

2. Experimental Methods

2.1 Microorganism and Culture Media Used

The microbial mixed culture obtained from a pharmaceuticals industry wastewater treatment plant was acclimatized with MEK as the carbon source in a mineral salt medium [13]. The pH of the mineral salt media was adjusted to 6.5 and the cultures were grown under ambient conditions in a rotary shaker.

2.2 Biofilter System

In this study, two different packing medium was used press mud and cornstack. The packing media was sterilized by autoclave before packing. The biofilter was made from a height of 1 m cylindrical polymethylacrylate column with an inner diameter of 0.05 m, and filled to a height of 0.75 m with the packing media inoculated with activated sludge as shown in Fig. 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

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water humidifier to ensure that the air relative humidity was more than 95%. The minor portion of air was allowed to bubble through liquid MEK 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 up flow mode of operation. The flow rates were controlled by valves and metered by previously calibrated flow meters to obtain the desired MEK 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.

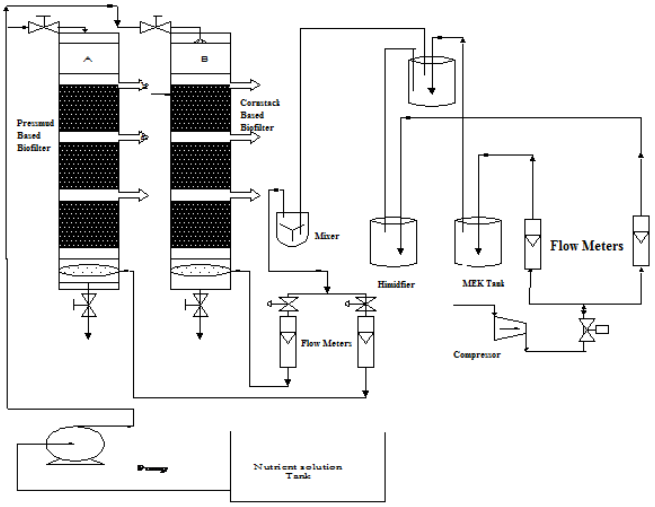

Fig. 1 Schematic diagram of biofilter

2.3 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 Fig. 2. The inlet concentration of pollutant was varied from 0.2 to 1.2 gm^{-3}. The EBRT was varied from 40 to 168 s.

2.4 Gas Analysis

The MEK concentration in the gas phase is analysed by using a PID Gas detector (model Gas Alert micro PID, BW technologies by Honey well, Canada).

2.5 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 [13].

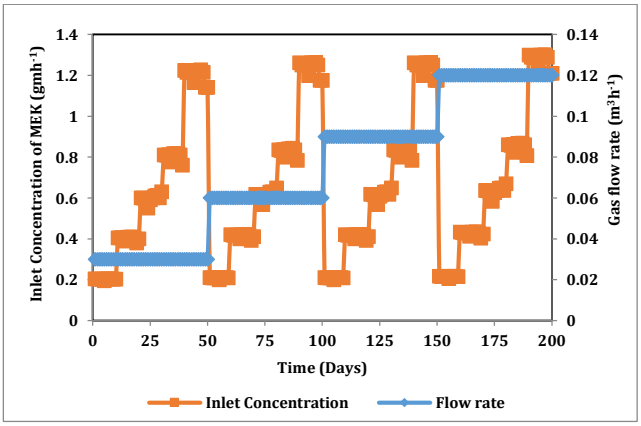

Fig. 2 Start-up of the biofilter for the removal of MEK using press mud based biofilter

3. Result and Discussion

3.1 Biofiltration of MEK using Press Mud and Cornstack as Packing Material

The biofiltration of gas stream containing MEK is carried out for 200 days at various operating conditions in an up flow mode press mud (BF1) and cornstack (BF2) based biofilters. Each biofilter had been operated in five stages. Each stage is divided into four phases as given in Fig. 2.

3.2 Effect of Gas Flow Rate and Inlet MEK Concentration

In the present work, the combined effect of the MEK inlet concentration and the gas flow rate on the biofilter performance is investigated by two packing materials BF1and BF2. Only the results obtained at steady state are discussed. Fig. 3 and Fig. 4 presents the RE and the outlet concentration of MEK for various inlet MEK concentration (0.2 – 1.2 gm^{-3}) and gas flow rates (0.03 - 0.12 $m^3 h^{-1}$) for BF1 and BF2 respectively. Fig. 5 for BF1 and Fig. 6 for BF2, presents the EC as function of the inlet load for each gas flow rates.

At a gas flow rate of 0.03 m^3h^{-1} and inlet concentration of 0.2 ± 10% gm^{-3}, the removal of MEK is 97.9% for BF1 and 93% for BF2. The RE decreases to 80% for BF1 and 75% for BF2 when the inlet concentration increased from 0.4 gm^{-3} to 1.2 gm^{-3}. The results are shown in Fig. 3 and Fig. 4. At this gas flow rate, for IL upto 8.023 $gm^{-3}h^{-1}$, corresponding to inlet concentrations of 0.4 gm^{-3}, it is found that EC increases with IL. For higher IL the EC decreases.

At a gas flow rate of 0.06 $m^3 h^{-1}$, with MEK concentrations varying from 0.2 $gm^{-3}$to 1.2 gm^{-3}, the RE decreases from 89% to 63% for BF1 and 85% to 59% for BF2. It is evident from the Fig. 3 and Fig. 4. At this gas flow rate, the EC of MEK increases upto an IL of 16 $gm^{-3} h^{-1}$ for BF1 and 30 $gm^{-3} h^{-1}$ for BF2 and then decreases as given in Fig. 5 and Fig. 6 for BF1 and BF2 respectively.

Similarly, at a gas flow rates of 0.09 m^3h^{-1}, the removal of MEK decreases for inlet concentrations ranging from 0.2 gm^{-3} to 1.2 gm^{-3} for BF1 and BF2 respectively as shown in Fig. 3 and Fig. 4. For loads smaller than 60 $gm^{-3} h^{-1}$, EC increases with IL to a maximum of 41 $gm^{-3} h^{-1}$ and 35 $gm^{-3} h^{-1}$ for BF1 and BF2 and decreases for higher MEK loads. Similar trend is observed in the gas flow rate of 0.12 $m^3 h^{-1}$.

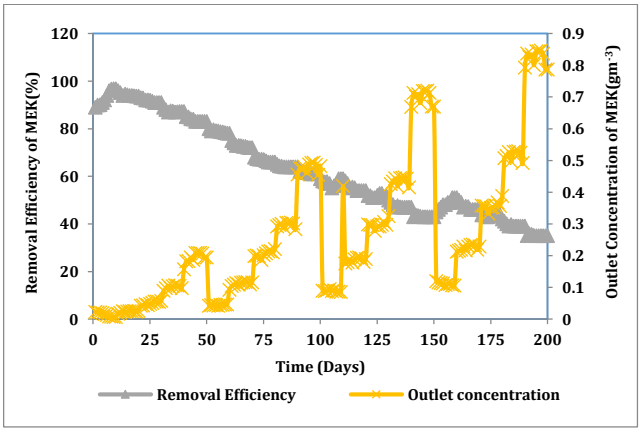

Fig. 3 Performance of biofilter for the removal of MEK using press mud based biofilter

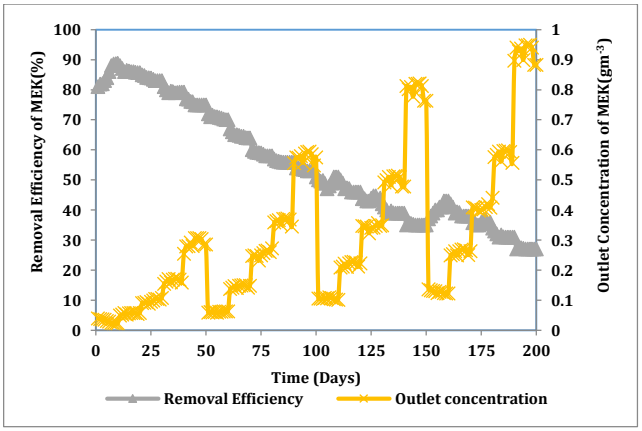

Fig. 4 Performance of biofilter for the removal of MEK using Press mud based biofilter

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A maximum elimination capacity of 64.4 gm^{-3} h^{-1} for BF1 and 57.7 gm^{-3} h^{-1} for BF2 is achieved at inlet concentration of 0.8 gm^{-3} and a gas flow rate of 0.12 m^3 h^{-1} as shown in Fig. 5 and Fig. 6 for BF1 and BF2 respectively. Nearly 100% removal is achieved at a gas flow rate of 0.03 m^{3}h^{-1}, for both the biofilters. When the gas flow rate is increased, the EC at constant IL and RE at constant MEK inlet concentration is found to decrease. This is because of decreased contact time between the pollutant and the microbial population at higher gas flow rate. EC is found to increase with IL up to a certain value and decreases on further increase in inlet concentration. The increase in EC with the increase of the MEK inlet concentration is due to enhanced transfer rate of MEK from the gas phase to the biofilm, so that more microorganisms participate to the biodegradation activity. This behavior can be described as a diffusion limitation regime. As IL is increased above the upper limit of the diffusion limitation regime, EC decreases.

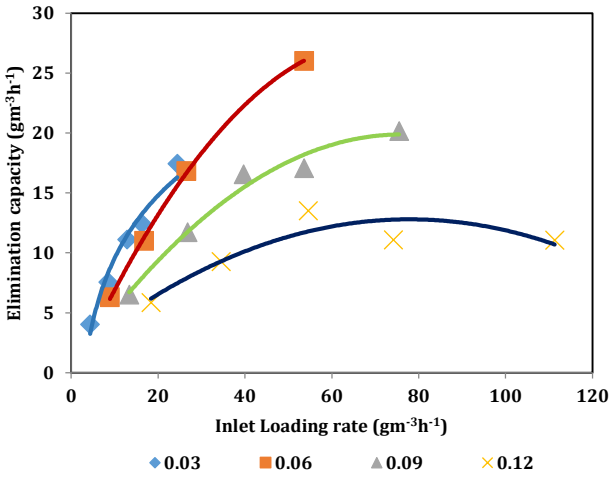

Fig. 5 Inlet Load Vs Elimination capacity for the removal of MEK using press mud based biofilter

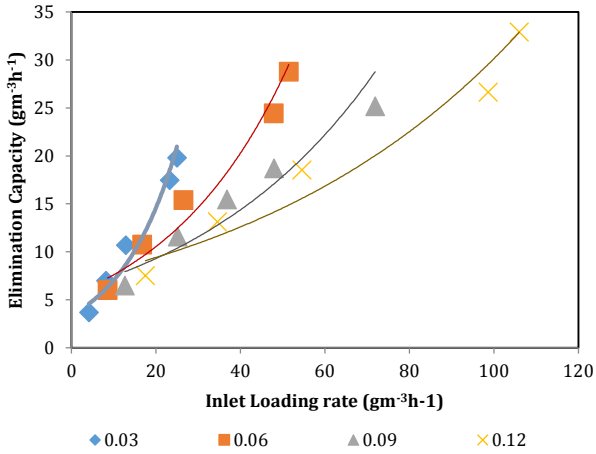

Fig. 6 Inlet Load Vs Elimination capacity for the removal of MEK using Press mud based biofilter

3.3 Application of the Theoretical Model

According to equation (11), the outlet concentration of MEK, in the situation of diffusion limitation can be described by the following equation:

$$\sqrt{C_i} = \sqrt{C_o} - k_1 \frac{V}{Q} \qquad (20)$$

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_o. According to equation (20), the theoretical diffusion model can be judged to be appropriate if the experimental points are on a line with a slope equal to 0.6. By knowing the gas flow rate and the filter bed volume, the constant of the line equation enables to estimate the parameter k_1. 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 MEK can be estimated from the following relationship

$$EC = \frac{Q}{V} C_{o,Crit} L \left(1 - \left((1 - k_1 \frac{V}{Q} \frac{1}{\sqrt{C_{o,crit}}}\right)^2\right) = k_o \qquad (21)$$

Hence,

$$C_{o,Crit} = \frac{1}{4}\left(\frac{k_o}{k_1} + \frac{k_1 V}{Q}\right)^2 \qquad (22)$$

The model is tested for the biofiltration of MEK using press mud and cornstack based biofilter. For each packing material, the plot has a portion displaying increasing elimination capacity with pollutant load which can be identified by the diffusion limitation behavior, and a portion displaying constant elimination capacity, which is attributed to the reaction limitation behavior. Thus, diffusion limitation is valid for low concentrations and theoretical reaction limitation model seems to be valid for high concentrations of MEK in both the biofilters. The values of model parameters, kinetic constants and maximum EC for at different operating conditions were tabulated in Table 1.

The biofilm thickness was also calculated for different phases using Eq. (11) by taking the values of effective diffusivity of biofilm (D) and Henry's constant (m) for MEKas 1.026 X 10^{-6} m^2 h^{-1} and 0.00235 respectively. The values of biofilm thickness were reported in Table 1. An increasing trend was observed for the biofilm thickness for different phases.

3.4 Modified Ottengraf Model

In the Ottengraf model, two different equations are proposed. 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 model gives a mathematical continuity to the two Ottengraf's equations. In this way, the contribution of both phenomena can be taken into consideration simultaneously. The modified model was tested with experimental data obtained in this study. In this study, the modified Ottengraf model was also used. This new model considers both diffusion and reaction limitations as a single equation.

3.5 Fundamentals of the New Model

Ottengraf's 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:

$$EC_{dL} = L\left(1 - \left(1 - A_s \sqrt{\frac{k_0 D}{2m}} \sqrt{\frac{V}{QL}}\right)^2\right) \qquad (23)$$

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

$$EC_{rl} = EC_{\max} = A_s k_0 \delta \qquad (24)$$

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

$$EC = EC_{\max} + \frac{(EC_{dl} - EC_{\max})}{1 + \left(\frac{L}{L^*}\right)^p} \qquad (25)$$

where L* is the load at which the transition between reaction and diffusion limitation occurs: for L < L*, conditions of diffusion limiting area are verified, while for L > L* the bioreaction is the rate determining step. For L << L*, the denominator of the second term on the right side becomes equal to 1 and in such conditions, EC ≡ EC_{dl}. Similarly, for L>>L*, all the second term on the right side becomes zero and therefore EC ≡ EC_{rl}. Parameter p was calculated by fitting of the experimental data. Its value specifies the rate at which the passage between the two different limiting conditions occurs. Having a sole equation has many advantages, including the possibility to correlate directly the removal efficiency to the load and to the inlet concentration. Indeed:

$$\eta = \frac{C_i - C_o}{C_i} = \frac{EC}{L} = \left(EC_{\max} + \frac{(EC_{dl} - EC_{\max})}{1 + \left(\frac{L}{L^*}\right)^p} \right) / L \quad (26)$$

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

$$\eta = \frac{\left(A_s k_o \delta + \frac{\frac{C_i Q}{V}\left(1 - \left(1 - A_s \frac{V}{Q}\sqrt{\frac{k_o D}{2mC_i}}\right)^2\right)}{1 + \left(\frac{C_{g,in}}{C_g^*}\right)^p} \right)}{\frac{C_i Q}{V}} \quad (27)$$

$$C_o = C_i - \left[\frac{Q.A_s k_o \delta}{V} + \frac{C_i\left(1 - \left(1 - A_s \frac{V}{Q}\sqrt{\frac{k_o D}{2mC_i}}\right)^2\right) - A_s k_o \delta}{1 + \left(\frac{C_i}{C^*}\right)^p} \right] \quad (28)$$

where C* is the inlet concentration at which load is equal to the L*, at constant flow rate and volume. This simple modification of Ottengraf's model is not merely an algebraically expedient to give mathematical continuity to equations (27) and (28). Indeed, it is expected that, inside a biofilter, diffusion and reaction limitation conditions simultaneously occur. This may be due to the progressive reduction of pollutant concentration along the reactor, to the presence of some areas with different superficial velocities and to changes in the thickness of the (biomass/solid) film. However, in 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. 7 and8. It was noticed that for this parameters set, the modified model individuates an area with efficiency higher than 100% at very low load values. The arbitrary choice of the parameter p could also cause this anomaly.

3.6 Modified Ottengraf Model - Advantages and Limitations

Since it is based on Ottengraf studies, the model has the same limitations. First of all, it is restricted to stationary conditions. The response of the system to external variations is thus not considered. However, it can be used for a first attempt or to evaluate how parameters vary during the operation. In addition, the degradation rate follows a zero-order kinetic. This assumption may be valid for high inlet concentrations and for very soluble pollutants. Indeed, it has been demonstrated that for certain types of contaminants, first-order kinetic prevails [16]. Oxygen limitations are also not considered in the kinetic model. Stratification of the biofilm along the reactor and the contribution of the moisture level are also not included in the model. In addition, Ottengraf model dealt with conventional biofilters; hence it does not consider the effects of the biofilter on the removal efficiency. Anyway, the Ottengraf-modified model furnishes one equation for the entire range of mass loading rate and, thereby, many equations can be written to relate loads, concentration, elimination capacity and efficiency.

3.7 Data Fitting

Experimental data are fitted by using the modified Ottengraf's model. This model relates the elimination capacity and the mass loading rate by the following equation.

$$EC = A_s k_0 \delta + \left[\frac{L\left(1 - \left(1 - A_s\sqrt{\frac{k_0 D}{2m}}\sqrt{\frac{V}{QL}}\right)^2\right) - A_s k_0 \delta}{1 + \left(\frac{L}{L^*}\right)^p} \right] \quad (29)$$

and the calculation of the removal efficiency can be easily obtained by using the definitions of EC and L:

$$RE = \frac{C_i - C_o}{C_i} X100 = \frac{EC}{L} X100 \quad (30)$$

EC and L data used for data fitting are obtained during the test to assess EC_{max}. Fixed and calculated parameters were reported in Table 1 for both the biofilters. The value of L* for the initial set was determined by using the definition of the critical Thiele module as referred by Ottengraf.

$$\phi_{cr} = \delta\sqrt{\frac{k_o m}{DC^*}} = \sqrt{2} \quad (31)$$

Indeed, as previously described, the transition between the reaction and the diffusion limitation area occurs at $\Phi_{cr} = \Phi$ or at $C_i = C^*$.

Using the definition of mass loading rate, L* can be thus expressed as follows:

$$L^* = \frac{C^* Q}{V} = \frac{\delta^2 k_o Q}{2DV} \quad (32)$$

Fitting was carried out for IL vs EC for BF1 and BF2 and were shown in Fig. 7 and Fig. 8 respectively. The final parameter set was successively used to calculate the dependence of the removal efficiency on the inlet loading rate. Fig. 9, report the model fitting for RE vs inlet loading rate for 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 1. In spite of all the limits encountered and discussed, the new model has a good agreement with the experimental data.

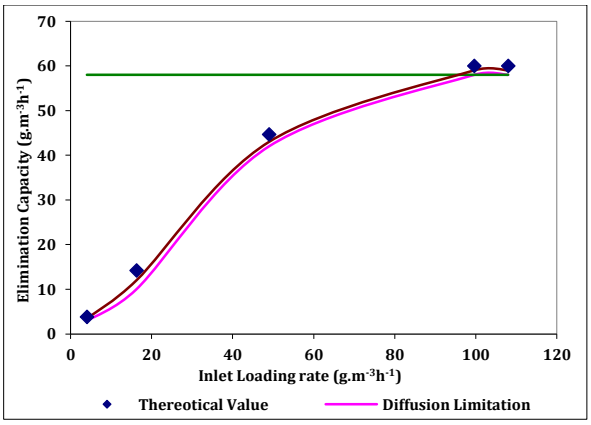

Fig. 7 Comparison of ottengraf model and modified ottengraf model with experimental values for MEK removal in a Press mud based biofilter.

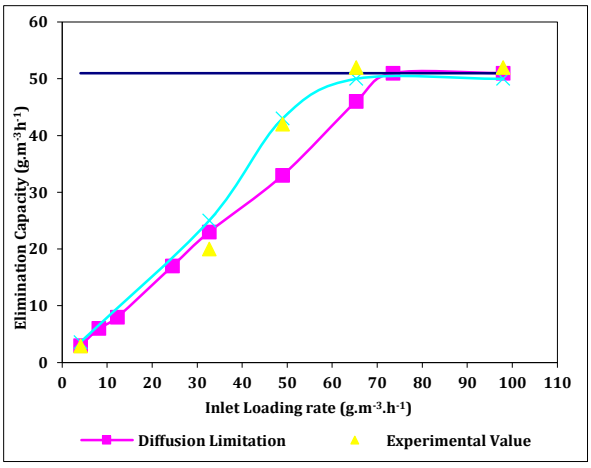

Fig. 8 Comparison of ottengraf model and modified ottengraf model with experimental values for MEK removal in a cornstack based biofilter.

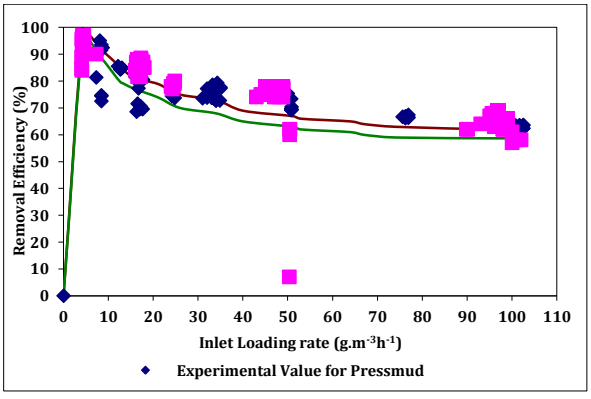

Fig. 9 Comparison of experimental and model predicted values for RE of MEK using Press mud and cornstack based biofilter.

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Table 1 Model parameters and kinetic constants at various operating conditions

Packing material	C_i (gm^{-3})	GF (m^3h^{-1})	IL ($gm^{-3}h^{-1}$)	k_1 ($gm^{-3}h^{-1}$)	k_d ($gm^{-3}h^{-1}$)	k_0 $gm^{-3}h^{-1}$	$C_{Critical}$ (gm^{-3})	$IL_{Critical}$ ($gm^{-3}h^{-1}$)	δ (μm)
Pressmud	0.2 – 1.2	0.03	4.16 - 25.02	0.711	1.235	13.2	0.81	16.5	254
		0.06	12.48- 50.04	0.701	1.29	32.45	0.801	40.4	269
		0.09	18.72- 75.06	0.773	1.56	49	0.833	51	320
		0.12	24.96-100.08	0.778	2.061	64	0.85	70	356
Corn Stack	0.2 – 1.2	0.03	4.16 - 25.02	0.761	0.126	12.1	0.931	19	241
		0.06	12.48- 50.04	0.740	0.133	28.05	0.784	32	255
		0.09	18.72- 75.06	0.702	0.152	42	0.784	48	302
		0.12	24.96-100.08	0.671	0.18	57.7	0.918	75	345

4. Conclusion

Two filter materials were evaluated for the biofiltration of MEK vapours. The press mud material has demonstrated as an optimal biofilter material, with low-pressure drop, and adequate removal efficiency along the bed height with respect to cornstack material. High MEK concentration values up to 1.2 g m^{-3} have been adequately treated, with a maximum elimination capacity of 95 $gm^{-3}h^{-1}$ and 90 $gm^{-3}h^{-1}$ for press mud and cornstack material respectively. The EBRT of 0.4 min has been established as the minimum operational EBRT to avoid high pressure drop and loss of biodegradation efficiency. In any case, press mud 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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