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Experimental studies and kinetic modeling for removal of methyl ethyl ketone using biofiltration

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ABSTRACT

The removal of toxic methyl ethyl ketone (MEK) is studied in a lab scale biofilter packed with mixture of coal and matured compost. The biofiltration operation is divided into 5 phases for a period of 60 days followed by shock loading conditions for three weeks. The maximum removal efficiency of 95% is achieved during phase II for an inlet concentration of 0.59 g m $^{-3}$, and 82–91% for the inlet concentration in the range of 0.45–1.23 g m $^{-3}$ of MEK during shock loads. The Michaelis–Menten kinetic constants obtained are 0.086 g m $^{-3}$ h $^{-1}$ and 0.577 g m $^{-3}$. The obtained experimental results are validated using Ottengrafvan den Oever model for zero-order diffusion-controlled region to understand the mechanism of biofiltration. The critical inlet concentration of MEK, critical inlet load of MEK and biofilm thickness are estimated using the results obtained from model predictions.

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1. Introduction

Methyl ethyl ketone (MEK) is one such highly toxic ketone compound released into the atmosphere by chemical, petrochemical, food processing, pulp and paper mills, color printing, paint and coating, electronic industries, etc leading to endanger the air quality and public health (Mitchell, 1992). Therefore, it is necessary to eliminate MEK from the environment, in order to prevent the significant impact on ecosystem and public health. The stricter environmental regulations these days have made it compulsory to look for the cost effective technology for the elimination of MEK from the waste air streams of industries.

Out of various techniques such as incineration, ozonation, combustion, adsorption, biofiltration has emerged to be cost effective technology for eliminating odorous and toxic volatile organic compounds (VOCs) such as MEK from waste gas streams in recent years. (Devinny et al., 1999).

Biofiltration uses microorganisms fixed to a packing material to breakdown pollutants present in an air stream. Biofilters are not filtration units but a kind of bioreactor which is an example of bioremediation technique. Biofiltration takes place by combination of basic processes such as absorption, adsorption and degradation (Devinny et al., 1999). In this process, microorganisms degrade the contaminants by consuming it as carbon source for their

growth and thus releasing end products such as carbon dioxide, water and biomass. The biomass can be reused by sending it back to the environment.

Biofiltration was studied for the removal of MEK and MIBK by Deshusses et al. (1996) using the mixed culture and compost as packing material and focused on the operational aspects of the process. Mathur and Majumder (2008) carried out the biofiltration in a coal based biotrickling filter for the removal of toluene, *n*-butyl acetate and o-xylene (MTBX) emitted from the paint industry. Chan and Peng (2008) studied the performance of biofilter using composite bead made up of (PVA)/peat/KNO₃/GAC as filter material for the removal of MEK, methyl isopropyl ketone (MIPK) and acetone. It was found that the microbial growth rate k_g and biochemical reaction rate k_d would be inhibited at higher average inlet concentration of ketone compounds. In another study, Chan and Su (2008) studied the biofiltration of ethyl acetate and amyl acetate using the composite bead biofilter. Alvarez-Hornos et al. (2008) investigated the use of suitable packing material by carrying out experiments on soil amendment biofilter and fibrous peat biofilter for the removal of ethylbenzene vapors. They concluded that the fibrous peat biofilter performed better and showed the effective removal of high concentration of ethylbenzene (4.8 g m^{-3}).

The performance of biofiltration system mainly depends on the selection of microbial culture and packing material. The variety of packing materials used for biofiltration process are peat, coal, compost, soil, etc., as they provide basic nutrients and support needed for biofiltration process (Delhomenie and Heitz, 2005). Earlier studies were carried out using single packing materials which have

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Nomenclature C_{gi} inlet MEK concentration (g m⁻³) critical inlet concentration (g m⁻³) $C_{critical}$ exit MEK concentration (g m⁻³) critical inlet load (g m⁻³ h⁻¹ $C_{\rm g0}$ $IL_{critical}$ **EBRT** empty bed residence time (s) biofilm thickness (µm) elimination capacity (g m $^{-3}$ \acute{h}^{-1}) EC zero-order kinetic constant (g $m^{-3} h^{-1}$) K_0 inlet load (g $m^{-3} h^{-1}$) interfacial area per unit filter volume (m⁻¹) IL а saturation constant in the gas phase (g m⁻³) effective diffusion coefficient in biofilm (m² h⁻¹) K_{ς} De flow rate ($m^3 h^{-1}$) 0 m Henry coefficient of MEK in water (–) maximum degradation rate per unit filter volume $r_{\rm max}$ $(g \, m^{-3} \, h^{-1})$ V volume of filter bed (m³)

certain disadvantages. The peat and coal as a packing material do not provide enough nutrients or indigenous ecosystem for the microbial growth. The drawbacks of using only compost as a packing material include the development of back-pressure due to gradual compaction with time, and aging effects due to microbial mineralization. Hence, there is a need to use a combination of packing materials which has the advantages over the above mentioned shortcomings. One of the suitable combinations of packing materials could be coal and compost (hybrid bed system) which is used in the present study.

In the previous studies, the removal of MEK was carried out using biofiltration but the studies were not focused on the effective use of cheaper supporting medium in the biofilter. Very few studies have focused on the shock loading study which gives a better idea about the performance and the stability of the biofilter operation. Most of the studies are limited to judge the performance of biofilter column using the experimental data. The behavior of biofilter column can be better understood by studying the kinetic aspects of biofiltration process. The kinetic model has been neglected in most of the previous studies which are helpful in the design of a biofilter column as various parameters needed can be calculated using the kinetic model.

Hence, the present work deals with the removal of MEK from waste air streams using biofiltration. A mixture of matured compost and coal which is cheap and is readily available is used as a packing material for the biofilter column. The seeding of the column is carried out with the acclimated mixed culture for MEK obtained from the shake flask studies. The performance of the biofiltration experiment is evaluated for the period of 60 days by changing the inlet concentration of MEK and air flow rates. The performance of biofilter is gauged in terms of removal efficiency and elimination capacity. The stability of the biofilter performance is assessed in terms of subjecting the biofilter's column to shock loads for a period of three weeks after sufficient microbial culture was obtained from 60 days operation without giving any starvation period which is not studied in detail in the literature. The work also deals with the kinetic aspects of biodegradation in biofilters by fitting the experimental data generated in this study with the Michaelis-Menten kinetic model and the corresponding kinetic parameters are evaluated. The obtained experimental results are also validated with the Ottengraf-van-den Oever model for various phases.

2. Methods

2.1. Media preparation

Stock glucose solution was prepared by dissolving 10 g of glucose in 100 ml of distilled water. The Minimal Salt Medium (MSM), was prepared which has the following composition (in g L⁻¹): $K_2HPO_4-0.8$, $KH_2PO_4-0.2$, $CaSO_4.2H_2O_7$

0.05, MgSO₄ \cdot 7H₂O - 0.5, (NH₄)2SO₄ - 1.0, FeSO₄ - 0.01 in distilled water. The value of pH of MSM obtained was 6.7 after the addition of all the salts.

2.2. Development of MEK utilizing acclimated culture

The development of MEK enriched culture was carried out in MSM with MEK as a sole carbon source. The source for the activated sludge was the secondary clarifier of Sewage Treatment Plant of Birla Institute of Technology & Science (BITS) Pilani, India. The activated sludge obtained was allowed to settle for 4 h away from sunlight. 10 gm of settled sludge was taken and thoroughly mixed with 100 ml of distilled water in a beaker. The shaking was carried out gently and then sludge was allowed to settle in order to subsequently screen out the solid particles. Fifty milliliters of supernatant was then taken in a 50 ml centrifuge tube. The centrifugation was carried out for 2 min at 10,000 rpm at 40 °C in a Centrifuge (Remi Cooling Centrifuge, India). The portion of the upper liquid was removed carefully from the top of the centrifuge tube without disturbing the pellet. The shake flask study was carried out by taking out the pellet with the help of a loop and transferred into a 250 ml flask containing 100 ml MSM in an aseptic environment along with MEK and glucose. Thus the acclimated culture was obtained from 15 days cycle by increasing the amount of MEK from 160 to 480 mg L^{-1} and decreasing the amount of glucose from 1000 to 0 mg L⁻¹. The final acclimated culture was obtained containing only MEK as a sole carbon source which was used as a seeding culture for the biofiltration study. It is observed from several studies that the acclimated mixed culture is much better choice over pure culture as in both the cases better or almost comparable removal efficiencies are obtained (Arnold et al., 1997 and Dehghanzadeh et al., 2005).

2.3. Packing material

The column was packed with matured mixture of compost and coal in the ratio of 2:1 (V/V). The matured compost was obtained from The JRD Tata Foundation for Research in Yoga, Naturopathy & Ayurvedic Sciences, Chitrakut, UP (India). The matured compost was derived from cow dung and subjected to anaerobic digestion. The coal was obtained from the local market. It was sieved through 8-10 mm mesh screen and final coal size was obtained as 2.36 mm. The coal was then washed with normal water and then with distilled water. The coal was then kept for removing the moisture by heating in the oven which was maintained at a temperature of 1000 °C for 1 day (Mathur and Majumder, 2008). The coal and compost was then thoroughly mixed with 200 mL of acclimated mixed culture obtained from the shake flask study prior to the packing of the column. The moisture content of the packing material was obtained as 60.2% on wet weight basis.

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