International Journal of ChemTech Research
CODEN (USA): IJCRGG ISSN: 0974-4290
Vol.8, No.6, pp 358-367, 2015

Biotreatment of waste gas containing MEK in a using agro based biofilter

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Abstract: In the research described herein, operating strategies was evaluated over a period of 200 days in biofilters treating a model waste gas stream containing a methyl ethyl ketone (MEK). The biofilter is packed with pressmud and cornstack with mixed culture obtained from activated sludge of waste water treamtment plant that containing MEK used as a solvent, as an intermediate for synthesis and as a catalyst for a variety of applications. Experiments were carried out by subjecting the biofilter to different flow rates (0.03–0.12m^3h^{-1}) and concentrations (0.2–1.2 gm^{-3}), corresponding to inlet loading rates varying from as low as 3 $gm^{-3}h^{-1}$ to as high as 1390 g $m^{-3}h^{-1}$. The biofilter proved to be highly efficient in the removal of MEK at a gas flow rate of 0.2 m^3h^{-1} corresponding to a gas residence time of 2.8min. For all the tested inlet concentrations, the removal efficiency decreased for high gas flow rates. For all the tested gas flow rates, a decrease in the removal efficiency was noticed for high MEK inlet concentration. The follow-up of carbon dioxide concentration profile through the biofilter revealed that the mass ratio of carbon dioxide produced to the MEK removed was approximately 2.52, which confirms complete degradation of MEK if one considers the fraction of the consumed organic carbon used for the microbial growth. Pseudomonas species enrichment of MEK- degrading microorganisms, was operated on a continuous feed basis for a period of 200 days.
Keywords: MEK; Cornstack; Pressmud; Identification; Isolation; Elimination Capacity.

Introduction

Biofiltration is an air pollution control technology that holds great promise for effectively and economically removing biodegradable organic and inorganic compounds from gas-phase waste streams. Biofiltration technology offers environmental advantages: it does not generate undesirable byproducts by converting many organic and inorganic compounds into harmless oxidation products. Biofilters work by absorbing noxious gases into a biofilm where microorganisms break down the gases into carbon dioxide, water, and salts and use the energy and nutrients to grow and reproduce. The quantities of carbon dioxide, water, heat and biomass produced depend on the metabolism and the process conditions. Biofilter contain packing materials such as compost, soil, peat, granular activated carbon or other porous media capable of adsorbing gaseous compounds and support biological growth.

Methyl ethyl ketone (MEK), methyl isobutyl ketone (MIBK) and methyl isopropyl ketone (MIPK) are widely used industrial chemicals. These ketone compounds were designed high-priority toxic chemicals. Large volumes of these ketone compounds are released into the atmosphere during manufacturing processes every year, leading to endanger the air quality and public health. The removal of volatile organic compounds (VOCs) from a polluted air stream using a biological process is highly efficient and has low installation and maintenance costs.

Ottengraph reported that the greater the complexity of a waste stream in terms of number of constituents present, the lower the biodegradation of compounds achieved [1]. The inhibitory effect on the degradation rate of MEK Removal was studied[2]. The performance equilibrium and kinetic studies on MEK adsorption in compost and granular activated carbon. 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 [3]. To study the treatment of MEK contaminated air streams. Two types of packing materials, polypropylene spheres and wood bars, were tested in reactors with a treatment volume of 0.141 m3. Influent MEK concentrations ranging from 0.9 to 5 g/m3 were tested. Removal efficiencies ranging from 40 to greater than 97% were reported[4]. The treatment of a MEK contaminated gas stream using three separate reactor columns connected in series[5]. The columns were packed with 13 mm ceramic Berl saddles as an inert support for the biofilm, and the system had a total treatment volume of 0.003 m3. Influent MEK concentrations ranged between approximately 60 and 70 g/m3, and removal efficiencies ranged between 56 and 96%[5]. Reports on the biodegradation of MEK and MIBK in a biofilter are limited[6,7].

In the work described herein, the behavior of a laboratory scale biofilter inoculated with a mixed culture from a waste water sludge was investigated at different initial loading rates of MEK polluted gas. The effects of inlet gas concentration, flow rate, EBRT, carbondioxode production, pressure drop and temperature on the removal efficiency (RE) of MEK were studied. In addition, this research to identify the predominant isolates used for degradation of MEK.

Materials and Methods

Microorganism and Culture Media Used

The microbial mixed culture obtained form a pharmaceuticals industry wastewater treatment plant was acclimatized with MEK as the carbon source in a mineral salt medium.The inoculum was obtained from a two-month acclimatized culture seeded with activated sludge from the secondary clarifier of pharmaceutical industry. 200mL of the concentrated sludge were placed in an aerated batch reactor and diluted with 1 L of nutrient solution containing N and P (3.84 g l^{-1} K_2HPO_4, 1.94 g l^{-1} KH_2PO_4, 3.00 g l^{-1} NH_4Cl, pH 6.97). Vitamins and trace minerals were added by diluting 3 g of Supradyn. The pH of the mineral salt media was adjusted to 6.5 and the cultures were grown under ambient conditions in a rotary shaker.

Biofilter system

In this study, two different packing medium was used PressMud and Cornstack. The packing media was sterilized by autoclave before packing. 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 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 upflow mode of operation. The flow rates were controlled by valves and metered by previously calibrated flowmeters 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.

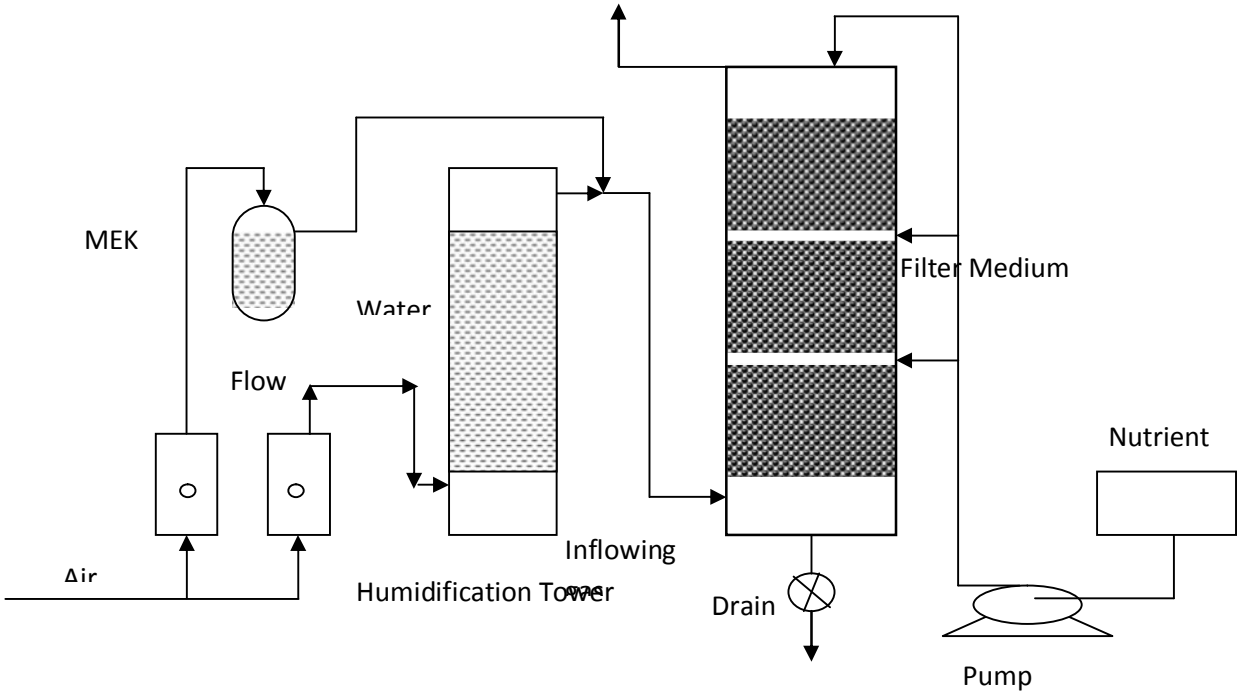

Fig. 1. Schematic diagram of up flow biofilter system

Procedure for Microbial count, isolation and identification

Counting of microorganisms

The microorganism concentration was measured in the units of colony forming units CFU g^{-1} of packing material. After end of the experiment (200 days of biofilter operation), one gram of moist packing material (Corn stack and pressmud) was taken from the middle of the column. 0.8% NaCl solution was prepared by adding nine milliliter of sterilized water and was added in the collected sample of packing material. This solution was shaken in a shaker for 15 min vigorously and serially diluted with sterilized water. Then the plating was carried out in the nutrient agar media in the petri dishes in the aseptic conditions. The petri dishes were incubated in an incubator for 24 hours at 30^{o}C. Then the colonies were counted in a colony counter to study the microbial concentration for the acclimation period and end of the experiment.

Isolation of micro organisms

The microbial isolation was carried out using the serial dilution technique. After isolation, individual colonies were carefully picked up based on their morphological characteristics. Each colony was inoculated on separate nutrient agar plates. The microbes were routinely cultured in nutrient agar to obtain a pure culture and were maintained in agar slants. The identification of microorganisms is done by conducting a series of standard biochemical tests, staining and motility tests.

Biofilter Operation

Experiments were performed for a period of 200 days. The experimental operation was divided into periods (I, II, III and IV) according to Empty Bed Residence Time (EBRT). The operating conditions of each period were summarized in Table 1. The inlet concentration of pollutant was varied from 0.2 to 1.2 gm^{-3}. The EBRT was varied from 2.81 to 0.7 min.

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).

Performance evaluation

The performance of the biofilter was evaluated by the following performance parameters, xylene Inlet Load (IL), $g.m^{-3}h^{-1}$, Removal Efficiency (RE), %, Elimination Capacity (EC), $gm^{-3}\ h^{-1}$. The definitions for these parameters are set out below:

$$EBRT = \frac{V}{F}(s) \text{ ----------(1)}$$

$$RE = \frac{C_i - C_o}{C_i} X100 \text{ (\%)-- ----------(2)}$$

$$EC = \frac{F(C_i - C_o)}{V} \text{ } (g.m^{-3}h^{-1}) \text{----------(3)}$$

$$\Delta CO_2 = C_{CO_2,out} - C_{CO_2,in} \text{ } (g.m^{-3}h^{-1}) \text{----------(4)}$$

$$P_{CO_2} = \frac{Q(C_{CO_2,out} - C_{CO_2,in})}{V} \text{ } (g.m^{-3}h^{-1}) \text{------(5)}$$

Where V is the volume of the packed bed section (m^3); F is the gas flow rate (m^3h^{-1}); and C_i and C_o are the inlet and outlet concentration ($g.m^{-3}$) of the pollutant, respectively. The carbon dioxide concentrations measured at the inlet and exit of the biofilter are denoted by $C_{CO_2,in}$ and $C_{CO_2,out}$ ($g.m^{-3}$), respectively. Operational parameters such as the pollutant inlet concentration and the EBRT are in general not constant but fluctuate within certain ranges

Result and Discussion

Overall system performance under variable loading condition

The biofiltration of gas stream containing MEK was carried out for 200 days at various operating conditions in an up flow mode biofilter. The biofilter was operated in four stages. Each stage is divided in to five phases as shown in Table 1. Various gas flow rate and concentration were maintained so that the corresponding loading rate could be maintained and regulated in the reactor to study the performance of the reactor (Figure 2)

Table 1 Summary of Experimental Plan

Days of Operation	Gas Flow rate (m^3h^{-1})	Inlet Concentration (gm^{-3})
0-10	0.03	0.2
11-20		0.4
21-30		0.8
31-40		1.2
0-10	0.06	0.2
11-20		0.4
21-30		0.8
31-40		1.2
0-10	0.09	0.2
11-20		0.4
21-30		0.8
31-40		1.2
0-10	0.12	0.2
11-20		0.4
21-30		0.8
31-40		1.2

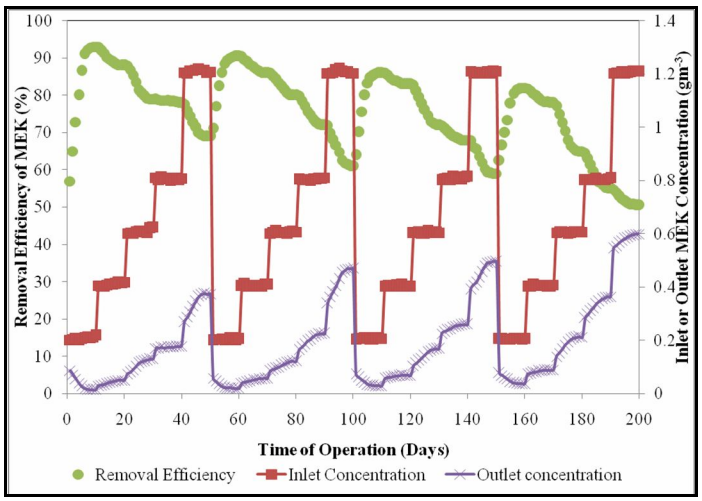

Fig. 2. Start-up of the biofilter for the removal of MEK using pressmud and corn stack based biofilter

Phase - I Inlet Concentration of 0.2 ± 10 % (1 to 40days)

The first phase of the experiment lasted for 40 days. In the I phase of the first run, the flow rate and inlet concentration of MEK were maintained at 0.03m^3h^{-1} and at 0.2 ± 10 % gm^{-3}, respectively, so that the average loading rate of 4.08 gm^{-3} h^{-1} can be applied to the reactor. The corresponding EBRT was maintained at 2.8 min. Gradual increase in removal efficiency was observed. It was evident from the figure. 3 the maximum removal efficiency of 93 % was obtained after 8 days of operation.

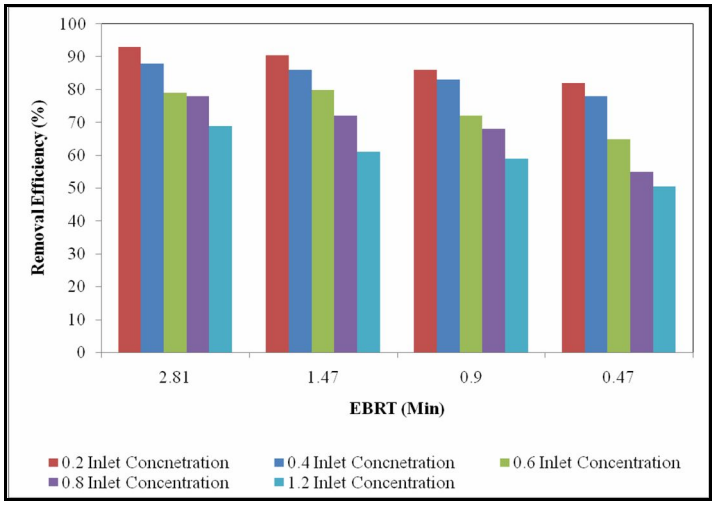

Fig 3 Shows the effect of EBRT on the removal efficieny

In run II, average loading rate has been increased by nearly twice from 4.08 to 8.16 gm^{-3} h^{-1}. The flow rate and EBRT was maintained at 0.06 m^3h^{-1} and 1.47 min respectively. In this run, the average inlet MEK concentration was kept constant at 0.2 ± 10 % gm^{-3}. Due to the shock load there was a fall in the removal efficiency of MEK in earlier stage from 93% to 80% and took 3 days to recover. Later it recovered gradually from this shock loading to 91%, at steady state.

In run III, average loading rate was increased from 8.16 to 12.14 gm^{-3} h^{-1} and flow rate of main stream and EBRT was 0.09m^3h^{-1} and 0.7 min. The inlet concentration was kept constant at maintained at 0.6 gm^{-3}. With the sudden increase in the loading rate to the reactor removal efficiency of 91% to 65%, but later on it recovered gradually from this shock loading to 88% at steady state. Run III lasted for 10 days.

In run IV, average loading rate was increased from 12.14 gm^{-3} h^{-1} to 16.33 and flow rate of main stream and EBRT was 0.09m^3h^{-1} and 0.7 min. The inlet concentration was kept constant at maintained at 0.8 gm^{-3}. With the sudden increase in the loading rate to the reactor removal efficiency of 55%, but later on it recovered gradually from this shock loading to 85% at steady state. Run IV lasted for 10 days.

Phase- II: Inlet Concentration of 0.4 ± 10 % (41 to 80 days)

The second phase of the experiment lasted for 40 days. In the I run of the second stage, the flow rate and inlet concentration of MEK were maintained at 0.03m^3h^{-1} and at 0.4 gm^{-3}, respectively, so that the loading rate 8.16 gm^{-3} h^{-1} can be applied to the reactor. The corresponding EBRT was 2.8 min. Gradual increase in removal efficiency was observed (Figure. 2 and more than 95 % removal was obtained after 8 days of operation. In run II, loading rate has been increased by nearly twice from 8.16 to 16.33 gm^{-3} h^{-1}. The MEK concentration was kept constant at 0.4 gm^{-3}. In this phase, the flow rate of main stream and EBRT was maintained at 0.06 m^3h^{-1} and 1.47 min. Due to the shock load there was a fall in the removal efficiency of MEK 95% to 80% and took 3 days to recover. Later it recovered gradually from this shock loading to 91%, at steady state. Phase II lasted for 10 days.

In run III, loading rate has been increased by 32.58 gm^{-3} h^{-1}. The MEK concentration was kept constant at 0.4 gm^{-3}. In this phase, the flow rate of main stream and EBRT was maintained at 0.09 m^3h^{-1} and 0.7 min. Due to the shock load there was a fall in the removal efficiency of MEK from 95% to 80%, and took 3 days to recover. Later it recovered gradually from this shock loading to 91%, at steady state. Run III lasted for 10 days.

In run IV, loading rate has been increased by 48.98 gm^{-3} h^{-1}. The MEK concentration was kept constant at 0.4 gm^{-3}. In this phase, the flow rate of main stream and EBRT was maintained at 0.12 m^3h^{-1} and 0.45 min. Due to the shock load there was a fall in the removal efficiency of MEK from 95% to 80%, and took 3 days to recover. Later it recovered gradually from this shock loading to 91%, at steady state. Phase III lasted for 10 days.

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} as shown in figure 2. Similar trend is observed in the gas flow rate of 0.12 m^3 h^{-1}.

Effect of empty bed residence time on biofiltration operation

The influence of EBRT on the RE has been studied and it is shown in Figure 3. The maximum RE is observed at the EBRT of 2.81 min, the RE is 97.3%. A decrease in the EBRT from 2.81 to 1.47 min caused a decrease in the removal efficiency from 97.3% to 91%. Further decrease in EBRT leads to decrease in RE. This is due to less contact time between the pollutant and packing material. These observations are in consistent with literature[8,9,10,11].

Carbon dioxide production

In the biofiltration process, the organic pollutants are aerobically degraded to water and carbon dioxide and used as the essential carbon source for the microbial growth. Hence, the profile of carbon dioxide concentration in the gas phase at the inlet and the outlet of the biofilter provide valuable information on the biofilter performance.

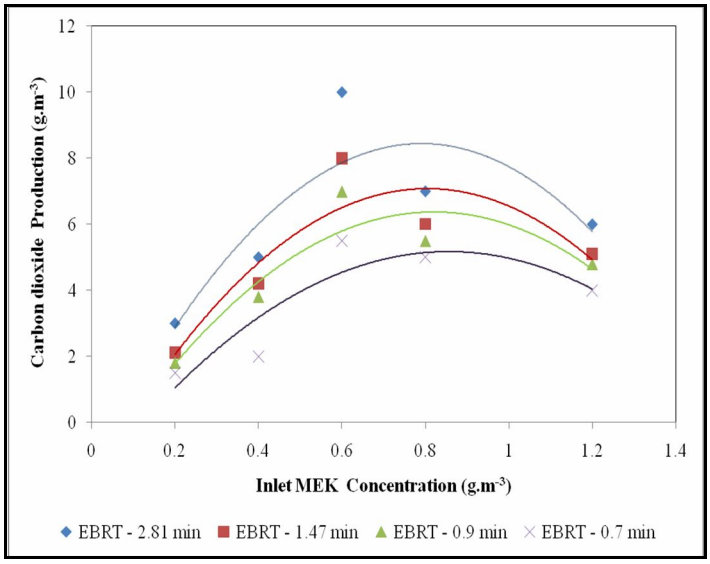

Fig 4 Effect of inlet MEK concentration on carbon dioxide production at various gas flow rates.

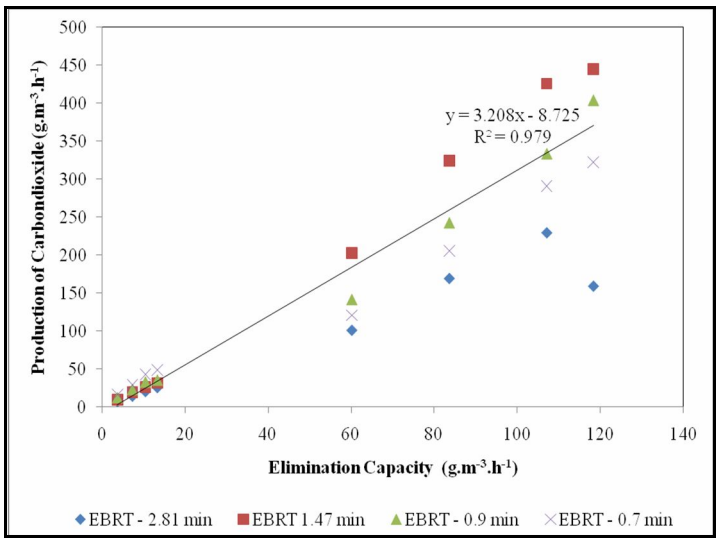

Fig 5 Profile of carbon dioxide production rate at different inlet concentration during the removal of MEK

In biofiltration process, the production of CO_2 is an important parameter to evaluate the degree of pollutant degradability, because pollutants are finally biodegraded to water and carbon dioxide and utilized as carbon source to form biomass for microbial growth. Figures 4 shows the effect of MEK inlet concentration and EBRT on CO_2 production. In all operating conditions, the outlet CO_2 concentration is always higher than the inlet CO_2 concentration because of CO_2 present in the atmosphere (0.72 gm^{-3})[9]. At an EBRT of 2.8 min and the inlet MEK concentration of 0.6 gm^{-3}, the maximum outlet CO_2 concentration is 10 gm^{-3}. At higher MEK inlet concentration, the CO_2 decreases. This is shown in Figure 5. The concentration of CO_2 at the exit of the biofilter decreases as the EBRT decreases. This result confirms the higher performance of the biofilter at higher EBRT. The same remark is also valid for the results obtained with the EBRT of 1.4, 0.7 and 0.4 min.

The variation of PCO_2 as a function of EC for various operating conditions is presented in Figures. From the figures it is observed that the PCO_2 is increased with increase in EC for different EBRT. The maximum PCO_2 of 444 $gm^{-3}.h^{-1}$ is obtained at the maximum EC. In this figure, the mean experimental data lie reasonably around the line y =3.028x. This indicates that the ratio between PCO_2 and EC, i.e. the mass of CO_2 produced per mass of MEK removed, is on average equal to 3.028 for all tested conditions. In fact, this ratio should be 3.3 in the case of complete oxidation of MEK to water and carbon dioxide according to the following stoichiometric reaction:

$$C_8H_{10} + xO_2 = yCO_2 + zH_2O + w\text{biomass}$$

However, in case of biodegradation of organic pollutants, a fraction of consumed organic carbon is used for the microbial growth according to the following metabolism:

$$\text{Organic Pollutant} + \text{Oxygen} \xrightarrow{\text{Microorganisms}} CO_2 + H_2O + \text{Heat} + \text{Biomass}$$

This explains the observed deficit in CO_2 production in comparison with the case of complete chemical oxidation of MEK. Further, in the biofiltration process, the biodegradation of pollutants occurs in the liquid phase (the wet biofilm), and the CO_2 produced may partly accumulate in biofilm as one of its solute species, HCO^{3-}, H_2CO_3 or CO_3^{2-} which can cause a deficit in CO_2 in the gas phase. This may also partly explain the fluctuations of the experimental ratio. Interestingly however, the small difference between the experimental ratio and the ratio evaluated from the stoichiometric reaction of complete oxidation is evidence of the removal of MEK exclusively by aerobic degradation and eliminates any option like adsorption or incomplete oxidation of MEK inn explaining the decrease of MEK concentration through the biofilter. Also, this analysis reveals that the follow up of the CO_2 concentration profile through the biofilter can be efficiently used for describing the biofilter performance.

Effect of temperature on elimination capacity of biofilter

The temperature of the filter material increases along the filter bed from the bottom to the top. This phenomenon is predictable since the metabolic biodegradation of the organic pollutants is an exothermic process. The energy released by these reactions causes the progressive rise of the gas temperature in the bioreactor, which in turn causes a positive temperature gradient in the filter bed from the gas inlet location to the outlet. Also, the lowest level of the bed is subject to heating effect of the inlet gas, which is at a temperature of 30°C. During all the operation period, the temperature increases with increase in elimination capacity.

Temperature variations at the mid level are very representative of the variation trends noticed in the other two levels. The daily follow-up of the filter bed temperature variations clearly revealed a sensitive dependence between the temperature of the filter bed and its biofiltration performance. During the first 10 days following the start-up of the biofilter at a gas flow rate of 0.03 m^3h^{-1}, the temperature increased from 23°C to 25°C as shown in figure 6. The increase in EC from 3.56 gm^{-3} h^{-1} to 15.97 gm^{-3} h^{-1} leads to progressive increase in the filter bed temperature of 23°C -25°C at the midlevel. At a gas flow rate of 0.03 m^3h^{-1}, the daily fluctuations of the temperature of the filter bed generally are subsequent to the change in EC. During the second experimental phase with a gas flow rate of 0.06 m^3h^{-1}, the filter bed temperature is generally higher than the temperature measured in the first experimental period (with a gas flow rate of 0.03 m^3h^{-1}).

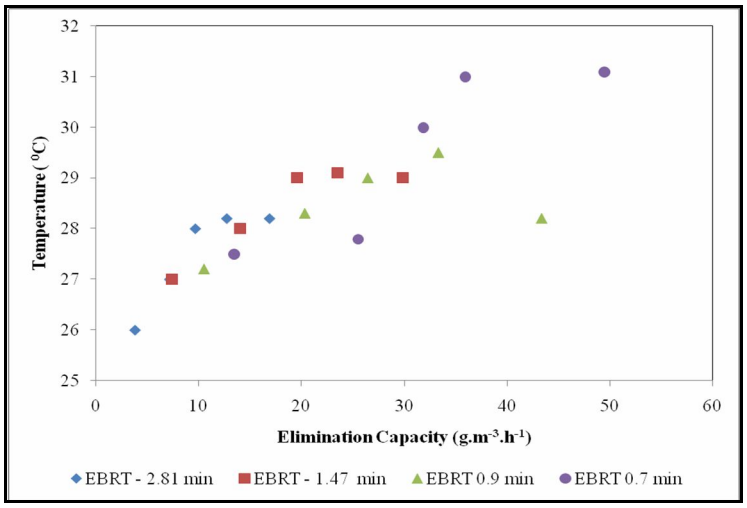

Fig6 Profile of temperature at different inlet concentration during the removal of MEK

In the third and fourth experimental period (with a gas flow rate of 0.09 and 0.12 m^3h^{-1}) similar trends are observed. The maximum temperature attained is 30.8°C with corresponding elimination capacity of 64 gm^{-3} h^{-1}. This behavior suggests a higher intensity of the metabolic microbial activity at that gas flow rate. Also, the cooling effect of the entering gas is less important at a smaller gas flow rate, which may partly explain higher bed temperature for smaller gas flow rates. Similar observations are reported in the MEK removal[12,13].

Pressure drop and airflow rate

Biofiltration technology offers the most effective method for aerobic biological treatment of waste air when dealing with high removal of biodegradable organics. The total pressure drop in both biofilters began to increase significantly after 10 days of operation. On day 10, with flow rate at 0.03m^3h^{-1}, an increase in pressure drop could have also been due to the increase in the airflow through the biofilters. Since the gas flow rate is increased the pressure drop increases more rapidly. The pressure drops in the biofilter is higher than in the pressmud based biofilter [13]. This is due to higher moisture retention capacity of the Cornstack (0.76 g water/g bed) than that of the pressmud based biofilter (0.46 g water/g bed)[15].

The packing material composing the bed serves as biofilm carrier and possibly as filter medium for the suspended solid retention. Pressure drop in biofilter media is a function of the moisture content of the packing material and gas flow rate [14]. Airflow rates are changed in order to change the MEK inlet concentration to the filter during the course of operation of the biofilter. Variation of pressure drop across the biofilter is measured with the increase in each gas flow rate and it is shown in figure 7. The pressure drop is in the range of 21 Pam^{-1} to 200 Pam^{-1}

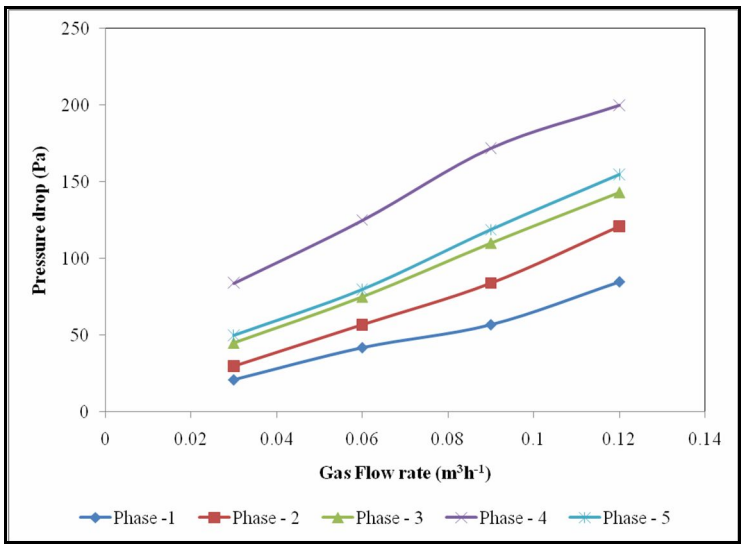

Fig 7 Effect of pressure drop on the different gas flow rate for cornstack and pressmud based biofilter for the removal of MEK

The pressure drop increases with increase in operating days due to biomass accumulation in the bed. It is maximum at the last phase for each flow rate in both the biofilter. The pressure drop is higher in our study when compared to others. The pressure drop values in the range of 15 Pam^{-1} to 460 Pam^{-1} was obtained using pig manure and saw dust as a packing material[15]. The pressure drop values in the range of 50 Pa m^{-1} to 1000 Pa m^{-1} for compost and they also suggested replacement of filter material when the pressure drop exceeds 250 Pam^{-1}. The results obtained in this study are in the range of 21 Pam^{-1} to 200 Pam^{-1} and 25 Pam^{-1} to 220 Pam^{-1} for BF1 and BF2 respectively. This value is below the critical values prescribed by earlier authors and comparable to the values obtained for saw dust, pig manure and compost. This indicates that agriculture reside used in the biofilter, as inert support material, is good in terms of maintaining the less compaction characteristics[16].

Biochemical characterization of microorganisms present in the biofilter

The biochemical characterization of the isolates were carried out following standard biochemical tests. From the test, we could identify isolate 1 as *Bacillus subtilis*. Isolate 2 belongs to *Bacillus* species and Isolate 3 belonged to *Pseudomonas* species. Further studies should be carried out in the molecular level in order to find out the particular strain of the microorganisms.

Conclusion

The biofiltration of Methyl ethyl ketone had been carried out in five phases that lasted for 200 days, using Press mud and Cornstalk as the packing materials. A pharamceticuals sludge provides easily adaptable microbial strains capable of degrading VOCs. From the study, it was found that the biofilters showed removal efficiency greater than 95% at a higher EBRT and low inlet MEK concentration. A sensitive dependence between the temperature of the filter bed and the biofiltration performance is noticed. Increase in temperature from 23 to 30.8^{o}C for increases elimination capacity from 3.8 to 64 gm^{-3} h^{-1}. The amount of CO_2 production increased from 1.40 to 8 gm^{-3} .The increase in CO_2 production is due to increase in elimination capacity. The pressure drop increases with increase in height. The pressure drop increases during the period of operation and reaches maximum at the top of the biofilter on 200th day of operation. The predominant bacteria present in the biofilter have been identified for the first time, as *Bacillus subtilis* and *Pseudomonas* species. Further studies at the genetic and molecular level are being carried out to identify and confirm the specific strain of these bacterial species. Studies on the MEK degrading fungal community present in the biofilter also have to be carried out.

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