

## **5. Biodegradation of Methyl ethyl ketone, Toluene and Xylene (MTX) from Air Using Modified Wood charcoal Beads as Biofilter Media**

In this experiment wood charcoal has been used as base material for modified biofilter media. It is an excellent material having very high surface area, good water adsorption capacity, excellent strength and also possesses adsorption properties which may be helpful in biofiltration. The only major limitation is unavailability of inherent nutrients and this limitation will be overcome by adding required nutrients during modification process.

### **5.1 Preparation of PVA/Wood Charcoal/KNO<sub>3</sub> Composite beads**

The method of preparation of composite beads is similar to the method adopted by Chan and Lin (2006). Wood Charcoal (200 g) was added to an aqueous solution (800 ml water mixed with 128 g KNO<sub>3</sub>) in a 2000 ml bucket. The mixture was sealed and kept for approximately 24 h for wood charcoal to adsorb KNO<sub>3</sub> and water. Now, an aqueous solution using 2000 ml water mixed with 128 g KNO<sub>3</sub> was prepared and 200g of PVA powder is added to it and the final mixture was heated to 90°C for dissolution. The wood charcoal /KNO<sub>3</sub> mixture was slowly added to the PVA/KNO<sub>3</sub> mixture at 90°C and stirred for 1.5 h maintaining the same temperature and then cooled to 40°C. The final cooled mixture was slowly dripped into a 6% boric acid aqueous solution (1500 ml) for 60 min leading to the formation of beads. The beads were transferred into the phosphate aqueous solution (150 g NaH<sub>2</sub>PO<sub>4</sub>·2H<sub>2</sub>O and 335 g Na<sub>2</sub>HPO<sub>4</sub>·12H<sub>2</sub>O in 450 ml water) and stirred for 30 min, and then immersed in the 0.384M KNO<sub>3</sub> solution because some amount of water soluble nitrogen was dissolved out in boric acid solution. Finally, beads were dried and stored in a desiccator at room temperature for use. Bed porosity, dry weight, moisture retention capacity and pH of the composite beads were estimated using the conventional method of analysis. CHN content of composite bead was measured using CHN analyser (Perkin Elmer). Bed porosity, dry weight, moisture retention capacity and pH of

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## 5.2 Result and Discussions

### 5.2.1 Physico-chemical characterization results

The Table 5.1 shows that there is enhancement in the physical properties like moisture retention capacity, bed porosity after modification of the wood charcoal which is favorable condition for successful biofilter operation.

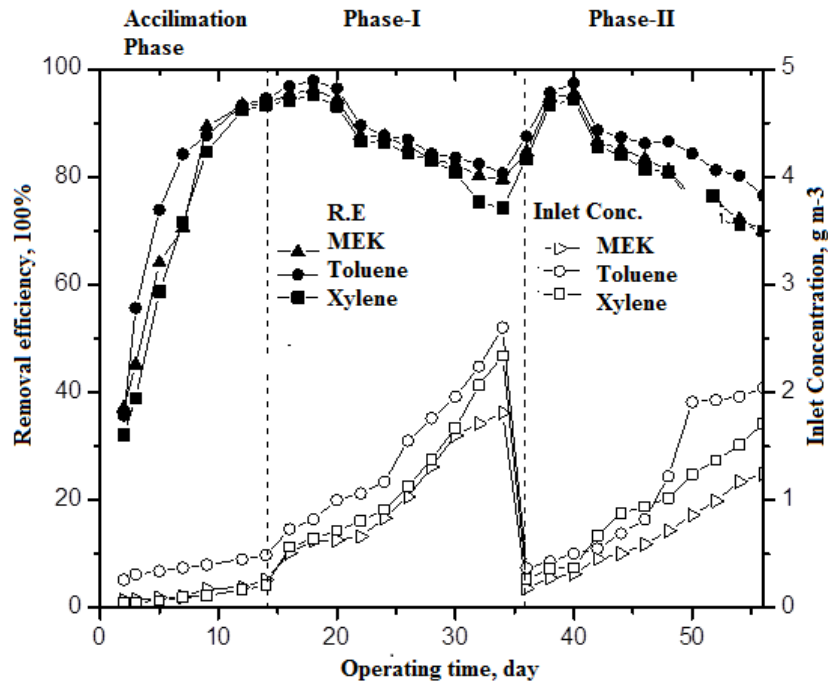
**Table 5.1:** Initial characteristics of PVA/Wood charcoal/ $\text{KNO}_3$  composite beads.

Sl. No.	Parameters	Wood charcoal Composite bead	Wood charcoal cubes
1.	Particle size(mm)	8-10	6-8
2.	Dry weight (g)	0.47	0.65
3.	Moisture retention capacity (%)	82.8	64.4
4.	Bed porosity (%)	77	68
5.	C H N Content (%)		
	C	68.63	43.25
	H	3.65	1.635
	N	2.29	1.415

### 5.2.2 Performance of biofilter under individual loading rates of MEK, Toluene and xylene

Continuous biodegradation of MEK, Toluene and xylene vapours were carried out for a period of 56 days in three distinct phases as shown in Tables 5.2. During the startup period, the biofilter was operated with an initial flow rate of  $0.06 \text{ m}^3 \text{ h}^{-1}$  corresponding to an EBRT of 57.9

sec with an average inlet concentration of MEK, toluene and xylene ranging from 0.073 to 0.26, 0.26 to 0.49  $\text{gm}^{-3}$  0.047 to 0.21 respectively for 14 days. The results are presented in Figures 5.1.



**Fig.5.1:** Variation of removal efficiency with achange in inlet concentration of MEK, Toluene and Xylene at different gas flow rates.

In the acclimation phase, the removal efficiency gradually increased and attained the values of 93.65, 94.55 and 93.35% for MEK, toluene and xylene respectively (on the 14<sup>th</sup> day). The conditions in the biofilter was stabilized which is indicated by almost constant RE during last phase of acclimation period. During phase I, the flow rate was increased to  $0.12 \text{ m}^3\text{h}^{-1}$  with corresponding EBRT of 28.4 sec. The inlet concentrations of MEK, toluene and xylene vapour were varied in the range of 0.51 to 1.81, 0.73 to 2.60 and 0.562 to 2.34  $\text{g m}^{-3}$  respectively. The removal efficiencies were observed in the range of 80 to 96%, 80 to 98% and 74 to 95% for MEK, toluene and xylene vapour respectively. The maximum removal efficiencies of 96.35, 97.87 and 95.2% were achieved for MEK, toluene and xylene vapour respectively on the 18<sup>th</sup> day

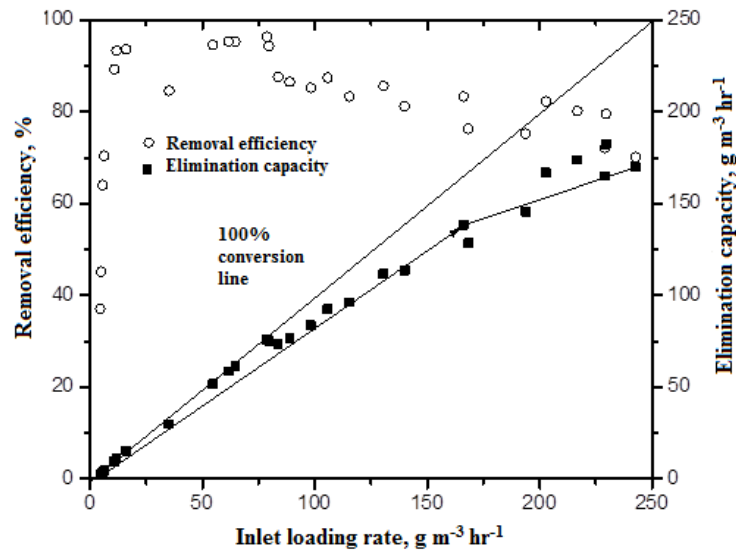
of operation. Further increase in inlet concentrations, a reduction in RE was observed and at the end of this phase removal efficiencies reduced to 80, 80, and 74% for MEK, toluene and xylene. During phase II, the gas flow rate was again increased to  $0.19 \text{ m}^3 \text{ h}^{-1}$  corresponding to EBRT of 18.5 sec and inlet concentrations were decreased to 0.18, 0.37, 0.26  $\text{g m}^{-3}$  from 1.81, 2.6, 2.34  $\text{g m}^{-3}$  for MEK, toluene and xylene respectively. After changing the flow rate and inlet concentration, first reading in this phase was recorded after two days i.e. on the 36<sup>th</sup> day of operation. First reading in this phase showed an increase in RE for MEK, toluene, xylene from 79 to 84 %, 80 to 87% and 74 to 83%. When inlet concentration increased continuously from 0.18 to 1.24, 0.37 to 2.04, and 0.266 to 1.74  $\text{g m}^{-3}$  for MEK, toluene and xylene, respectively, RE first increased and reached 95.35%, 97.36 % and 94.32% on 40<sup>th</sup> day and then decreased to 70.12 %, 76.53% and 70% at the end of this phase. Results show that biofilter responded very quickly to the change in flow rate and inlet concentration of all components of MTX.

**Table 5.2:** Experimental scheme for continuous MEK, Toluene and xylene biodegradation

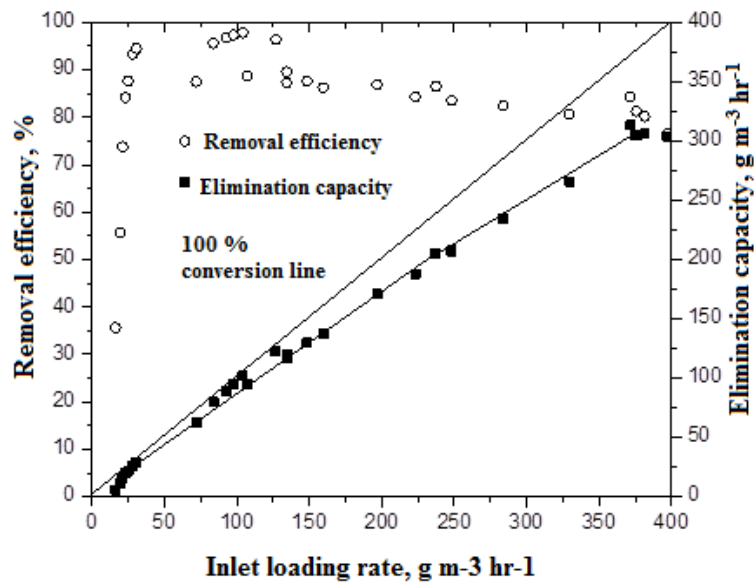
<b>Nature of biofilter operation</b>	<b>Inlet concentration range of MEK (<math>\text{g m}^{-3}</math>)</b>	<b>Inlet concentration range of Toluene (<math>\text{g m}^{-3}</math>)</b>	<b>Inlet concentration range of Xylene (<math>\text{g m}^{-3}</math>)</b>	<b>Operating Time (Days)</b>
Acclimation phase	0.073- 0.26	0.26- 0.49	0.047- 0.21	14
Phase I	0.51-1.81	0.73- 2.60	0.562 to 2.34	19
Phase II	0.18-1.24	0.37-2.04	0.266-1.71	23

### 5.2.3 Effect of Inlet Loading Rate on Removal Efficiency and Elimination Capacity

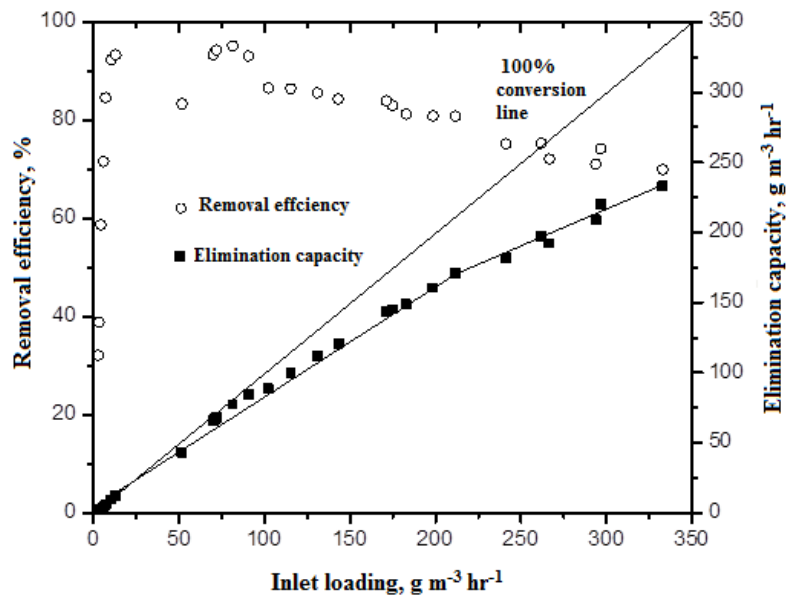
Figures 5.2, 5.3 and 5.4 shows the effect of inlet loading rates on removal efficiencies and elimination capacities for MEK, toluene, and xylene. A nearly linear relation between the inlet loading rates and elimination capacities were observed till loading rates of 166.05, 223.09 and 211.69  $\text{g m}^{-3} \text{h}^{-1}$  for MEK, toluene and xylene respectively. Further increase in loading rates, elimination capacities of all three components of MTX mixture were increased gradually and reached their maximum values of 182.53, 313.39 and 232.96  $\text{g m}^{-3} \text{h}^{-1}$  at inlet loading rates of 229.43, 371.67, and 332.75  $\text{g m}^{-3} \text{h}^{-1}$  respectively. Beyond loading rates of 78.7, 103.94 and 81.38  $\text{g m}^{-3} \text{h}^{-1}$ , RE started decreasing and EC increased with aslow rate. The reason for decrease in RE and slow increase in EC has already been discussed in previous section.



**Fig.5.2:** Influence of MEK loading rate on elimination capacity and removal efficiency of the biofilter.



**Fig.5.3:** Influence of Toluene loading rate on elimination capacity and removal efficiency of the biofilter.



**Fig.5.4:** Influence of Xylene loading rate on elimination capacity and removal efficiency of the biofilter.

#### 5.2.4 Performance of biofilter under combined loading rates of MTX

The continuous biodegradation of MTX vapour was carried out for a period of 56 days in three distinct phases as shown in Table 5.3.

**Table 5.3:**Experimental scheme for continuous MTX biodegradation

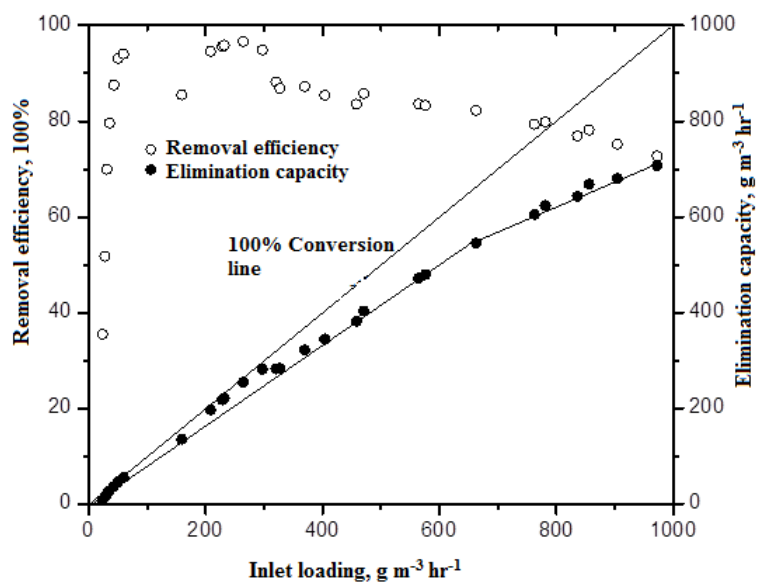
Nature of biofilter operation	Flow Rate ( $\text{m}^3 \text{h}^{-1}$ )	EBR T (sec)	Inlet Concentration range of MTX ( $\text{g m}^{-3}$ )	Removal Efficiency (%)	Operating Time (Days)
Acclimation phase	0.06	57.8	0.38 – 0.96	35.5-94.04	0-14
Phase I	0.12	28.4	1.8 – 6.75	95.58-78.13	15-34
Phase II	0.19	18.5	0.82 – 4.99	85.52-72.7	35-56

During the startup, the biofilter was operated with an initial flow rate of  $0.06 \text{ m}^3 \text{ h}^{-1}$  corresponding to an EBRT of 57.9 sec with inlet concentration ranging from 0.38 to  $0.96 \text{ gm}^{-3}$  for 14 days. In this phase, low flow rate and concentration of the pollutants were kept to provide sufficient microbial growth in the biofilter. The results are presented in Fig. 5.1. With the change in inlet concentration, overall removal efficiency gradually increased during this phase and an efficiency of 94.04 % was achieved at the end of this period (on the 14<sup>th</sup> day). During phase I, Flow rate was increased to  $0.12 \text{ m}^3 \text{ h}^{-1}$  corresponding to EBRT of 28.4 sec. The inlet concentration of MTX vapour was varied in the range of 1.8 to  $6.75 \text{ g m}^{-3}$  and the removal efficiency of MTX was observed in the range of 95.58-78.13%. The maximum removal

efficiency of 96.59 % was achieved at the concentration of  $2.08 \text{ g m}^{-3}$  on the 18<sup>th</sup> day of operation. Further increase in inlet concentration, a reduction in RE was observed. By the end of this phase, removal efficiency reduced to 78.13 %. During phase II, the gas flow rate was again increased to  $0.18 \text{ m}^3 \text{ h}^{-1}$  corresponding to EBRT of 18.5 sec and inlet concentration was decreased to 0.81 from  $6.75 \text{ g m}^{-3}$ . After changing the flow rate and inlet concentration, first reading in this phase was recorded after two days i.e. on the 36<sup>th</sup> day of operation. The reading showed an increase in overall RE from 78% to 85 %. When inlet concentration increased continuously from 0.81 to  $4.99 \text{ g m}^{-3}$ , RE first increased and reached to 95.88 % on the 40<sup>th</sup> day and then decreased to 72.7 % at the end of this phase. Results show that biofilter responded very quickly to the change in flow rate and inlet concentration, which is an indication of active biofilm present in it.

Figure 5.6 shows the variation of RE and EC with respect to inlet loading rate. Up to the loading rate of approximately  $662.70 \text{ g m}^{-3} \text{ h}^{-1}$ , the RE was almost constant and then started decreasing. The mass transfer and reaction controlled zones are also clearly visible. In the acclimation period, the maximum total elimination capacity of  $56.13 \text{ g m}^{-3} \text{ h}^{-1}$  was achieved at a total inlet loading rate of  $59.68 \text{ g m}^{-3} \text{ h}^{-1}$  and in phase I, the maximum elimination capacity of  $668.52 \text{ g m}^{-3} \text{ h}^{-1}$  was achieved at total inlet load of  $855.63 \text{ g m}^{-3} \text{ h}^{-1}$ . In phase II, the maximum elimination capacity of  $706.88 \text{ g m}^{-3} \text{ h}^{-1}$  was observed at total inlet load of  $972.33 \text{ g m}^{-3} \text{ h}^{-1}$ . The results clearly indicate that the trend of results in this study is similar to the result discussed in the previous study.

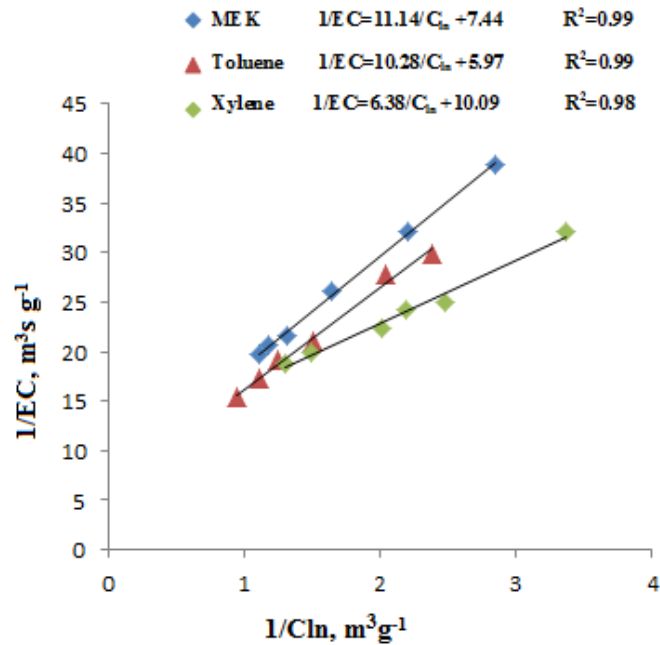




**Fig 5.6:** Variation of Elimination Capacity and Removal Efficiency with total MTX loading rates

### 5.3 Kinetic Analysis

The Michaelis-Menten constants were evaluated for methyl ethyl ketone (MEK), toluene and xylene respectively based on steady state values obtained during the experiments. Figure 5.7 represents the plot between  $1/EC$  versus  $1/C_{In}$  and based on the plot the Michaelis-Menten constants were evaluated.



**Fig.5.7** Determination of Michaelis-Menten constant for MEK, Toluene and Xylene degradation.

The value of ECmax was found to be 0.13, 0.16, 0.099 m<sup>-3</sup>gs<sup>-1</sup> for methyl ethyl ketone, toluene and xylene respectively. Similarly, the value of Ks was 1.49, 1.71, 0.63 gm<sup>-3</sup> respectively (Fig. 5.7). In all calculations correlation coefficient (R<sup>2</sup>) was more than 0.95. ECmaxvalue predicted by the model equation was higher than the experimentally observed ECmax. So, it could be predicted that higher elimination capacity could be achieved under well-controlled conditions in the biofilter. The ECmax and Ks values reported for MEK, toluene and xylene in a coal based biofilter degrading MTBX were 0.085, 0.033, and 0.024 g m<sup>-3</sup>sec<sup>-1</sup> and, 1.785, 0.736, 0.679 g m<sup>-3</sup> respectively (Mathur and Majumder, 2008) which is well comparable with the values obtained in this work. For the biofiltration of mono-chlorobenzene, the Cmax and Ks were evaluated 0.121 g m<sup>-3</sup> s<sup>-1</sup> and 7.45 g m<sup>-3</sup> respectively (Mathure *et al.*, 2006).

At last, summarily we can state that without external supply of nutrient solution the biofilter demonstrated very good performance throughout the operation. Physicochemical characterization results indicated improvement in some of the important properties (bed porosity, density etc.) of composite beads as compared to the base material (wood charcoal) using which it was prepared. No significant variation in the pressure drop across the bed was observed during whole operation. During whole operation inlet stream and bed, temperature variations were found in the range of 24.5 - 33.2 and 25.9 - 34.3°C respectively. Bed temperature was always found more than the temperature of inlet stream which might be due to exothermic bioreaction in the biofilter. The pH of the leachate was found almost constant (6.93 - 7.02) and this may be due to the buffer capacity of phosphate solution in which the composite beads were immersed during the preparation. Relative humidity of inlet stream was found in the range of 80-95%.

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