Performance of biofilter for the removal of toluene using mixed novel packing materials

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Abstract

In this experimental study, elimination of toluene vapors has been achieved in a pilot plant biofilter packed with a combination of pearl millet stack (novel) and polyurethane foam. After 10 days of start-up operation, maximum RE of 92% was attained at a loading rate of 56 g m^{-3} h^{-1} with Empty Bed Residence Time (EBRT) of 2.81 min in nonstop biofilter. The lower portion of the filter bed shows higher toluene removal due to a large number of active microbes (3.25×1010 CFU mL-1) which were significantly add-on than the other sections.

The biofilter exposed a rapid comeback to the transient ailment (shutdown, restart, and shock load) and the constancy of the biofilter was continued. Bacillus sphaericus and Pseudomonas alcaligenes bacterial segregate was predictable as the major strain involved in the toluene removal. The model predicted EC_{max} was nearly same for the experimentally observed ECmax.

Keywords: Biofilter, toluene, packing materials.

Introduction

Volatile organic compounds (VOCs) such as benzene, toluene, styrene, hexane, trichloroethylene, acetaldehyde, acetone and methanol usually sensed in the atmosphere are probable carcinogenic and lead to universal warming, ozone layer reduction and climate change [1,2]. Toluene which is highly toxic and carcinogenic is most frequently detected in the environment due to wide applications as industrial solvent and degreasing of metal parts[2].

The consumption of toluene is growing at 1.5% each year due to the widespread application. Tolerance level of toluene in the ambient air should be less than 0.003 g m^{-3} set by US Environmental Protection Agency. Toluene hazardously pollutes the water forms, soil exteriors, and atmospheric air due to its toxicity[3, 11].

Physicochemical waste gas treatment performances are functional for toluene removal for contaminated waste air [11] but not supportable due to commercial and natural limitations. Biofilters are confirmed well-organized between the other biological reactors for handling of polluted air streams[11]. Polluted air permits through the packing bed when transported from the gas phase to water phase in the biofilm and then the adsorbed waste gas spreads inside the biofilm where it is removed[5]. Biofiltration system suggests an operative, environmental friendly and energy intensive substitute which gives end product such as CO_2 and H_2O in biodegradation of VOCs with some generation of acid metabolites[6,20].

Grave limits for weakly soluble hydrophobic contaminants are gas solubility and bioavailability. These parameters typically decrease the removal of the toluene from their state[9]. Toluene is an extremely hydrophobic chlorinated compound and has low bioavailability [17]. The maximum RE and EC of toluene were achieved at polyurethane foam based biofilter 90.48 g/(m^3 h) and 88% respectively[10]. Rene et al reported that the EC of toluene ranged from 3.5 to 128 g/(m^3 h). Few researchers studied the relation between the microbial count and performance of the biofilter.

Moreover, compost comprises a huge microbial (bacterial) communal at identical time. Poultry litter compost has more nutrient content like nitrogen, potassium, and phosphorus extensively.[4,8,12] Recently several studies have successfully attempted to use tree bark[13], corn stack[14], pressmud[15], sugarcane bagasee[16] and cassurine seeds[4] as microbial source for degradation of VOCs. Packing material in biofilter must content the subsequent conditions. Mixture of pearl millet and polyurethane foam was nominated as novel packing material in this study which contents the above stated measures.

Inorganic material of polyurethane foam is often used in biofilters due to 30% of pore volume, higher surface area and spreads the wettability for microbial growth. Pearl millet stacks survive complex organic loading rate due to properties of wettability and high surface area. Experimental study needs out for the transient state conditions[4].

The main effort of the current study is to examine the toluene deletion by means of a biofilter packed with a mixture of pearl millet – polyurethane foam. Continuous performance assessment was approved out at dissimilar loading rates. The biofilter was exposed to stages of shut down and irregular loading conditions to evaluate the constancy of the biofilter bed. Lineweaver-Burk, Hanes–Woolf and Eadie–Hofstee type kinetics equation was altered to clarify the behaviour of the biofilter under continuous performance.

Material and Methods

Chemical, inoculums and media composition: Laboratory grade chemicals of toluene (>99%) were bought from Himedia Laboratories Pvt. Ltd., Mumbai, India. Mixed microbial consortium was collected in the paper industry

waste water treatment plant. The mineral salt medium composition was used in the previous literature.[16]

Biofilter packing material: The pearl millet stack waste was collected from a nearby village at Panruti, Cuddalore district, Tamil Nadu, India. Pearl millet is made in as cylinders (1.4-4.1 cm in height and 1.8-2 cm in outer diameter). Pearl millet was mixed with equal size of poly urethane foam in a 2:1 weight ratio.

Lab-scale biofilter setup and operation: The biofilter in this study is used to test gas-phase toluene removal. Figure 1 illustrates the lab-scale biofilter arrangement. The tubular shape of biofilter was finished of the polyacrylic tube (5×75 cm) having 3 sampling ports at every 25 cm distance along the biofilter height. The lowest attachment of perforated plate that is supporting the filter bed was packed to a height of 12.5 cm in biofilter.

Entire volume of biofilter consists of 1.47 L. Toluene waste gas was only sole carbon source used in the research and the biofilter is functioned in an up-flow method. Synthetic toluene waste gas flux was generated by using a mixing compartment to connect two air streams. Inlet, outlet of the packed bed and even along the height of the packed bed are gas sampling ports. U-tube manometers are distributed around the biofilter to assess the pressure drop. The biofilter worked at room temperatures of approximately 28±4 ° C and the relative humidity of the inlet gas flow was maintained at 70%. The biofilter functioning at different investigational phases is arranged in table 1.

Continuous steady-state experimentations are carried out at different toluene loading rate at different empty bed retention time (EBRT). EBRTs of the biofilter was preserved at 0.7, 0.9, 1.47 and 2.81min corresponding to gas flow rate of 0.03, 0.06, 0.09 and 0.12 m^3h^{-1} respectively. Startup experimentations are carried out for 10 days (Toluene IC range of 0.2-0.4 gm^{-1} and EBRT of 2.41 min) which help the microbial actions get adaptation to the ecological situations. Transient conditions such as shut down, restart and shock loading are also studied in biofilter operations.

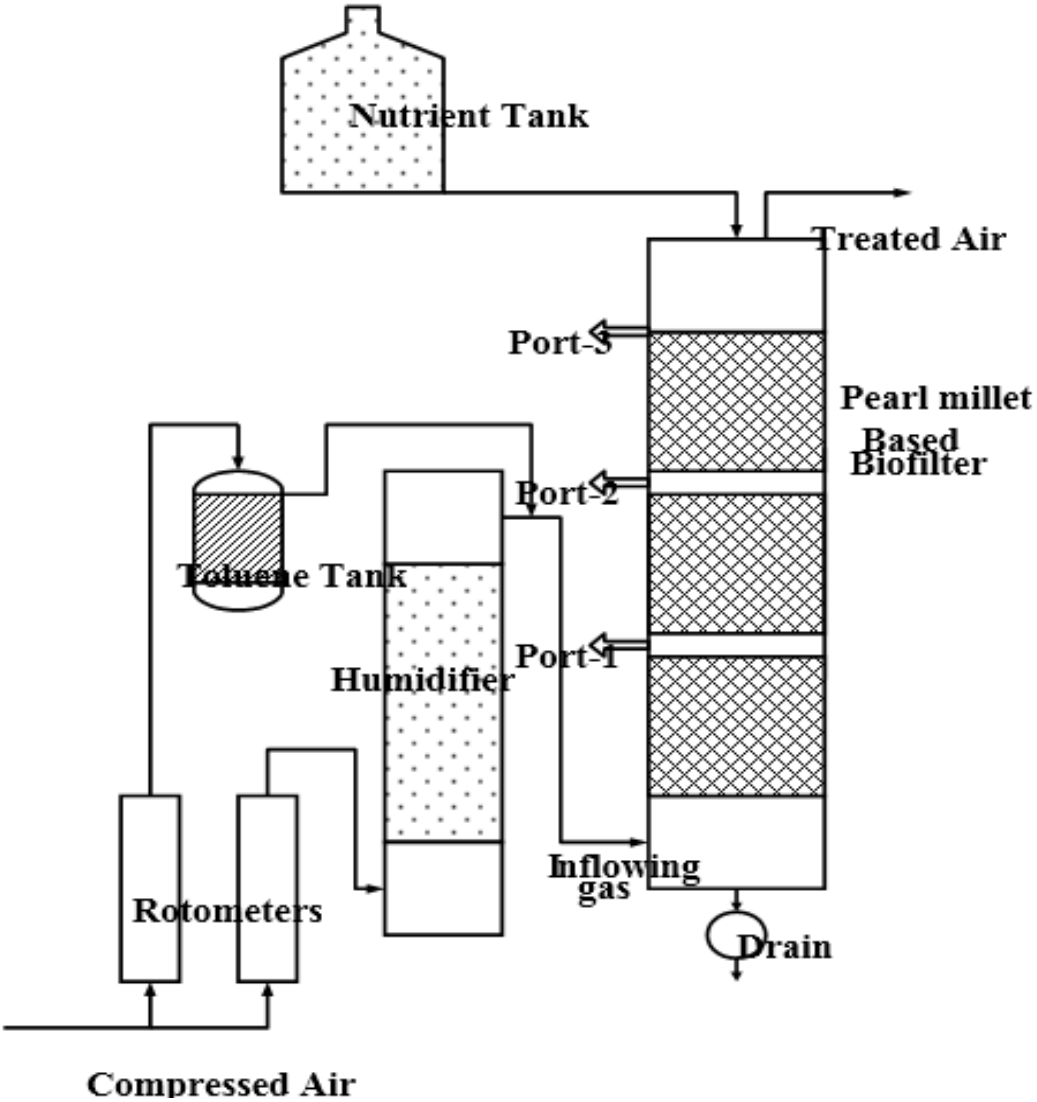

Figure 1: The schematic diagram of the integrated bioreactor

Table 1
Shows the investigational plan

Stages	Operating Days	GFR	Inlet Toluene Concentration
I	1- 10	0.03	0.2
	11-20		0.2
	21-30		04
	31-40		0.6
	41-50		0.8
	51-60		1.0
II	61-70	0.06	0.2
	71-80		04
	81-90		0.6
	91-100		0.8
	101-110		1.0
III	111- 120	0.09	0.2
	121-130		04
	131- 140		0.6
	141-150		0.8
	151-160		1.0
IV	161-170	0.12	0.2
	171-180		04
	181-190		0.6
	191-200		0.8
	201-210		1.0

Kinetic study of the toluene biofilter: For this research, we used modified Michaelis-Menten kinetics. Lineweaver-Burk, Hanes – Woolf and Eadie – Hofstee developed a kinetic model for pearl millet -poly urethane foam dependent biofilter. The biofilter activity experimental data are used in all k equation and kinetic parameters such as maximum ECmax elimination power, g m^{-3} h^{-1}) and constant of half-saturation (K, g m^{-3}) are calculated.

The equation 1 for Lineweaver-Burk kinetic model is:

$$\frac{1}{EC} = \frac{K_L}{EC_{max} C_{ln}} + \frac{1}{EC_{max}} \quad (1)$$

where *EC* is the EC (the reaction rate), K_L is the Michaelis–Menten constant, EC_{max} is the maximum EC, and [C_{ln}] is the substrate concentration.

A plot of 1/Cln against 1/ EC will yield K_L as the y-intercept, EC_{Max}/K_L as the x-intercept.

The equation 2 for Eadie - Hofstee kinetic model:

$$EC = \frac{ECK_E}{C_{ln}} + EC_{max} \quad (2)$$

where K_E is the Michaelis–Menten constant.

A plot of EC/Cln against EC will yield K_E as the y-intercept, EC_{Max} as the x-intercept.

The equation 3 for Hanes–Woolfe kinetic model is:

$$\frac{C_{ln}}{EC} = \frac{C_{ln}}{EC_{max}} + \frac{K_H}{EC_{max}} \quad (3)$$

where K_H is the Michaelis–Menten constant.

A plot of C_{ln} against C_{ln} /EC will yield K_H/EC_{max} as the y-intercept, $1/EC_{Max}$ is the x-intercept and

$$C_{ln} = \frac{C_{in} - C_{Out}}{\ln\left(\frac{C_{in}}{C_{out}}\right)} \quad (4)$$

where C_{in} and C_{out} are the inlet and outlet toluene concentration (g m^{-3}) respectively.

Sampling and analytical methods: The inlet and outlet concentration of toluene was determined by PI detector in Gas Alert Micro 5 PID - model (Honeywell Analytics, Inc., IL USA). Isolation of the microbes was determined by standard technique. Fawole and Oso reported the serial dilution for the isolation of bacteria (10^6 cfu/mL) and fungi (10^4 sfu/mL).

Biochemical studies using the KB001 Biochemical Test Kit (KB001 HiIMViCTM, Himedia, India) have identified the predominant toluene degrading strains in the biofilter. SEM research (Scanning Electron Microscopy-System JEOL-JSW-5610LV, Japan) performed morphological observation of the biofilter media.

Results and Discussion

Startup performance of the compost biofilter: Biofilter functioned for 10 days as start-up period to rise the cell holdup to the packing media. Figure 2 shows the variations of the IC, outlet concentration and RE of the pearl millet stack based biofilter during the start up period at a low gas flow rate of 0.03 $m^{-3}h^{-1}$ corresponding to EBRT of 2.81 min with a inlet toluene concentration range from 0.2 to 1.2 g m^{-3}. Initially, the efficiency of removal reached 18% on the first day and then increased dramatically to stabilize between 78-83%. After 10 days of start up, stable operation was achieved with high RE of 80 percent and the packed bed surface is found filled with microorganisms that are reported from the light colored biomass observed on the packaging. This makes the growth of microbials porous.

The result of start up indicated that the biofilter filled with pearl millet stack – polyurethane foam could be quickly acclimatized by inoculating mixed culture and it could well utilize the toluene as its sole carbon and energy source Due to the low gas flow rate, the pressure drop was marginal during the start-up cycle and the volume occupied by biomass was lower compared to the effective porous space.

Performance estimation of the toluene biofilter in constant operation: The biofilter was continuously run at 4 phases with different loading rate for 200 days after the startup experiment. Each phase was divided into 5 stages. Figure 2 shows the performance evaluation of toluene biofilter in terms of RE across different phases and stages of operation. The biofilter was run for 50 days in phase I service with a concentration range of 0.2 to 1.2 g m^{-3} (EBRT = 2.81 min).

In phase 1, stage 1, the biofilter RE increased steadily and reached a new steady-state value of 92 percent at 0.2 g m^{-3} IC. In stage 2, toluene RE decreased to 88 per cent at toluene IC to 0.2 to 0.6 g m^{-3}. In the stage 3, the RE of toluene decreased to 84 % at an inlet toluene concentration of 0.8 g m^{-3}. In the stage 4, the RE of toluene decreased to 80 % at an inlet toluene concentration of 1.0 g m^{-3}. In the stage 5, the RE of toluene decreased to 76 % at an inlet toluene concentration of 1.2 g m^{-3}.

In phase II operations, the IC range is of 0.2 to 1.2 g m^{-3} with EBRT 1.47 min. In phase II operation, the biofilter was operated for 30 days at a concentration range of 0.2 to 1.2 g m^{-3} (EBRT = 1.47 min). In phase 1I, stage 1 the RE of biofilter was steadily increasing and reached a new steady-state value of 90% at a IC of 0.2 g m^{-3}. In stage 2, the RE of toluene decreased to 90 % as the IC of toluene increased to 0.2 to 0.6 g m^{-3}. In the stage 3, the RE of toluene decreased to 88 % at an inlet toluene concentration of 1.2 g m^{-3}. In the stage 3, the RE of toluene decreased to 84 % at an inlet toluene concentration of 0.8 g m^{-3}. In the stage 4, the RE of toluene decreased to 80 % at an inlet toluene concentration of 1.0 g m^{-3}. In the stage 5, the RE of toluene decreased to 76 % at an inlet toluene concentration of 1.2 g m^{-3}.

In phase III operations, the IC range is of 0.2 to 1.2 g m^{-3} with EBRT 0.9 min. The biofilter was run for 30 days in phase III service with a concentration range of 0.2 to 1.2 g m^{-3} (EBRT = 0.9 min). In step I1I, stage 1, the biofilter RE increased steadily and achieved a new steady-state value of 87 % at an IC of 0.2 g m^{-3}.

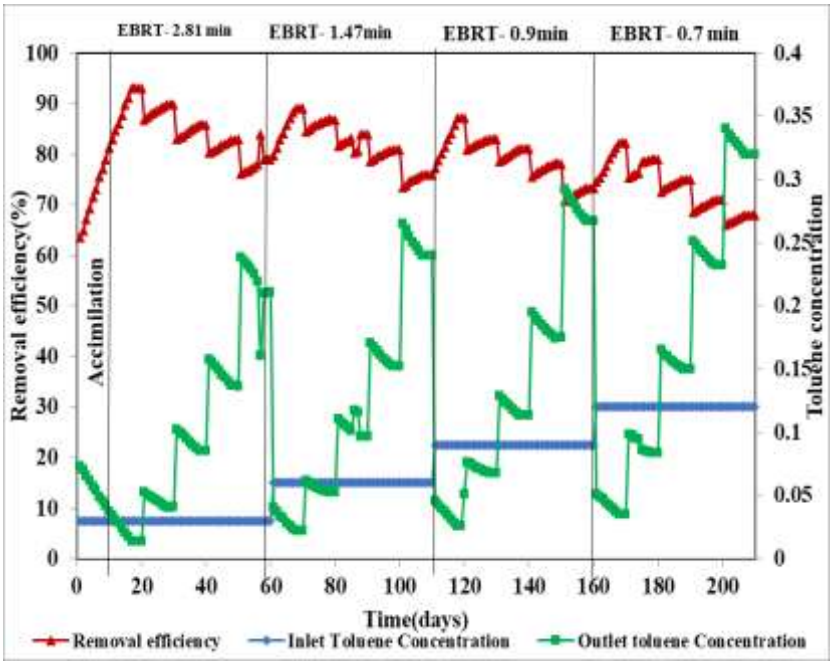

Figure 2: Effect of RE toluene by varying different IC and different EBRTs.

In stage 2, the toluene RE increased to 0.2 to 0.6 g m^{-3} at the toluene IC to 86 %. In stage 3, toluene RE decreases. In phase IV operations, the IC range is of 0.2 to 1.2 g m^{-3} with EBRT 0.9 min. In phase IV operation, the biofilter was operated for 30 days at a concentration range of 0.2 to 1.2 g m^{-3} (EBRT = 0.7 min). In phase I1I, stage 1 the RE of biofilter was steadily increasing and reached a new steady-state value of 85% at a IC of 0.2 g m^{-3}.

In stage 2, the RE of toluene decreased to 82 % at the IC of toluene increased to 0.2 to 0.6 g m^{-3}. In the stage 3, the RE of toluene decreased to 78 % at an inlet toluene concentration of 1.2 g $m^{-3.}$ In the stage 3, the RE of toluene decreased to 84 % at an inlet toluene concentration of 0.8 g $m^{-3.}$ In the stage 4, the RE of toluene decreased to 80 % at an inlet toluene concentration of 1.0 g m^{-3}. In the stage 5, the RE of toluene decreased to 76 % at an inlet toluene concentration of 1.2 g m^{-3}.

High toluene RE of approximately 92 per cent was obtained in stage 1 operations during phase I. In stage 2, the rise in toluene concentration from 0.2 g m^{-3} to 0.4 g m^{-3}, the RE not only decreased dramatically but also displayed noticeable variations. Different findings for stage 2 and stage 4 were obtained in phase 1.

Phase II raised the rate of gas flow to 0.03 to 0.06 m^3h^{-1}. The RE fell to 81 percent and then slowly increased over time, hitting a steady-state value by day 37. Similar results for rising the gas flow rate have been obtained. Possibly, the dramatic shift was due to part of the gas channeling induced by massive gas flow. The result indicated that the optimum concentration of EBRT and inlet toluene should be 2.81 min and 0.2 g m^{-3} respectively in the actual application of the biofilter based on pearl millet-polyurethane foam.

Figure 3 presents the effect of inlet loading rate on toluene biofilter removal efficiency and RE. The removal potential was increased linearly with toluene inlet loads below 59 gm^{-3h-1} and the RE remained nearly 90%, suggesting that the reaction was a first-order process regulated by gaseous diffusion[17]. When inlet toluene loading rate exceeded 59 gm^{-3h-1}, the EC and RE were changed to non-linearly and presented a slow decrease with the increase of inlet loading rate up to 67 $gm^{-3}h^{-1}$. The maximum EC and RE for toluene in the whole operation were 43 $gm^{-3}h^{-1}$ and 90 % observed at an inlet load of 59 $gm^{-3}h.^{-1}$

The increased concentration of contaminants did not immediately affect the efficiency of the reactor. The pearl millet stack-polyurethane foam used as a biosupport material for bacteria immobilization in this study allowed the sorption and degradation of waste gass. Since loading increased, the packaging material's sorption potential was depleted and no toluene was used by the microbes in biofilter due to substrate inhibition. The pearl millet stack's maximum EC – a biofilter based on polyurethane foam was much lower than the typical biofilter that treats other waste gass.[15] Toluene is highly volatile and less insoluble in water.

Microbial Aspects: For the creation of the microbial growth within the biofilter, microbial counts in the filter material were also determined. Figures 4 and 5 display the toluene degradation, bacteria and fungi intensity in the packaging material prior to biofilter operation and in the samples collected from the filter bed at various phases of biofilter operation. The raw sludge contained 108 CFUs per gram of dry microbe weight while the one gram of dry raw pearl millet- polyurethane foam contained no microbes. This is taken as an indication of the inoculum's suitable development which likely resulted in good biofilter performance during the initial days of service.

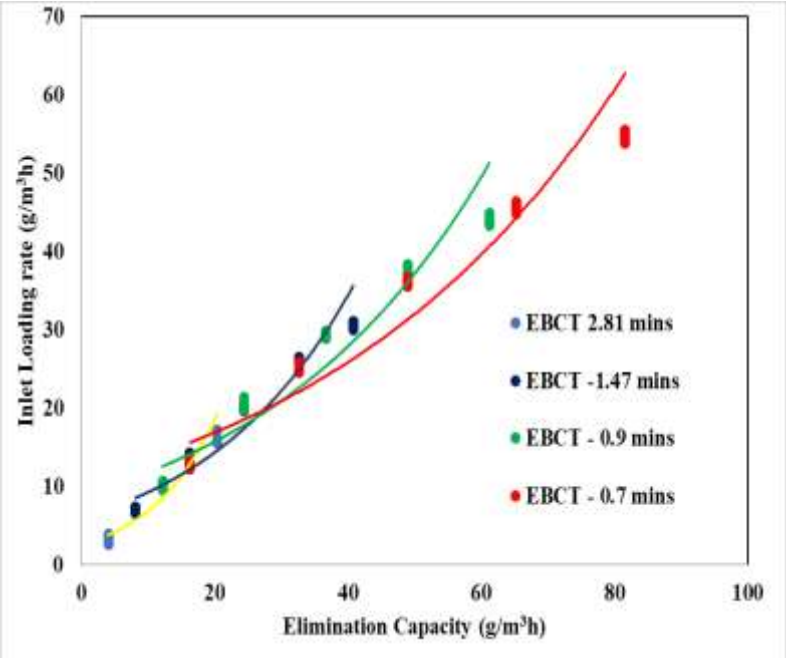

Figure 3: EC vs. Toluene inlet load for various gas flow rates

At day 20, the counts of fungi present in the bottom are about 5.0×10^8 CFU g^{-1}. The value decreased in the lowest to the biofilter, which can be connected to the broad removal of toluene at the first two sections of the biofilter. The toluene degrading microbes increased by to an average count of 6×10^9CFU g^{-1} at day 60 of operation and then further increased to 8×10^8 CFU g^{-1} at day 110. Similar observation was seen in the bacteria count as shown in figure 5. The maximum count was 9.1×10^8 CFU g^{-1} and 9.8×10^8 CFU g^{-1} at day 210 for fungal and bacteria respectively. These values are higher than those recorded for xylene degradation in biofilters packed with pressmud [16] and biofilter based on Sugarcane bagasse [17].

Microbial isolation and identification: Numerous colonies were obtained after sub culturing; from these colonies we selected two major colonies for isolation of strain. These two colonies were further isolated and stored at 4^OC for this study. Several tests for this tow-isolated strain were reviewed such as biochemical tests like gram staining. The table revealed the results. From these findings, the various characteristic phenotypic properties of the species Pseudomonas and Bacillus are clear. Several biochemical tests including gram staining were conducted to further classify the isolated organism.

Results showed that the isolated organism is a rod-shaped, gram-negative bacteria (data not shown). In addition, the organism was found to be oxidase, catalase and lipase positive but negative in indole and hydrogen sulphide production and urease test (Table 2). All these findings are found in several biochemical tests including gram staining, to further identify the isolated organism. Results showed that the isolated organism is a rod-, gram- bacteria (data not shown). In addition, the organism was found to be positive but negative in the synthesis of indole and hydrogen sulphide and urease oxidase, catalase and lipase.

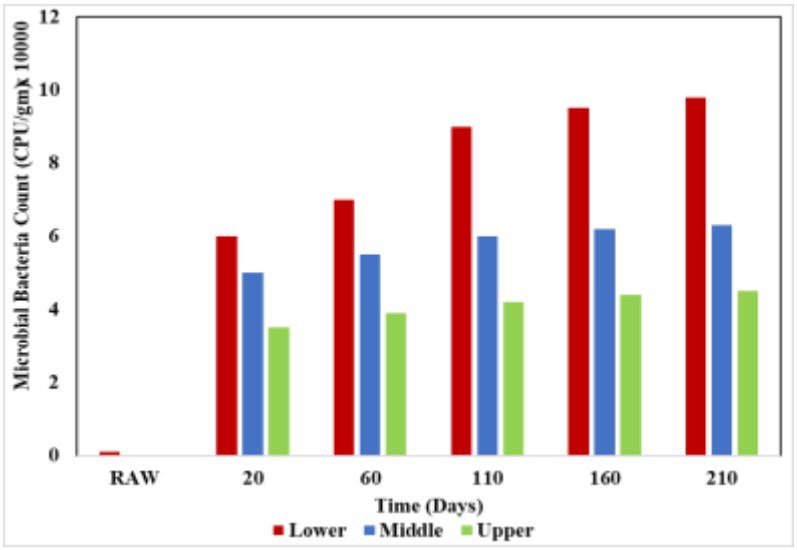

Figure 4: Total bacteria counts of toluene degraders in the biofilter bed at different days of operation (RW: Raw Compost).

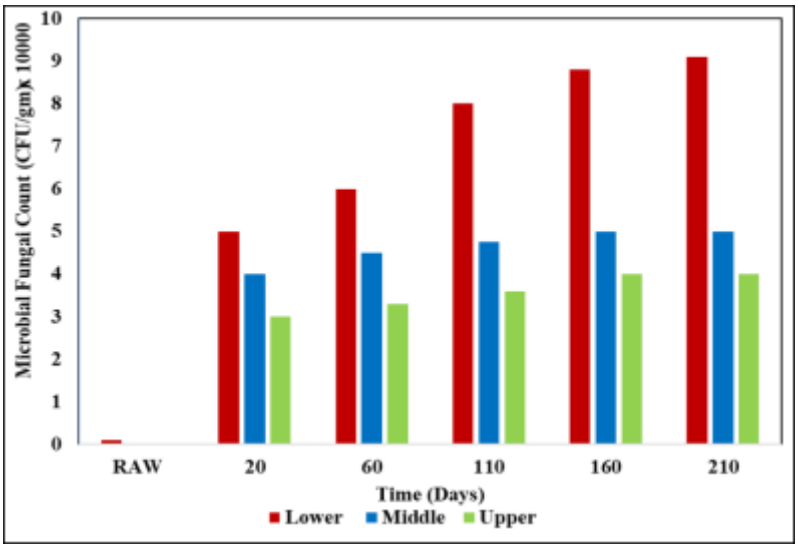

Figure 5: Total fungal counts of toluene degraders in the biofilter bed at different days of operation (RW: Raw Compost).

Microscopic View: Figure 6(a) shows the SEM image of pearl millet stack which exposed that very porous and raw exterior with big openings, which allowed the microbial attachment. In addition, at the end of the operating cycle, a sample was taken and analysed using SEM. A thick biofilm protected the perl millet stack surface was applied to the pores (Figure 6b). Although the morphology is hard to observe, some coccoid and rod shaped bacteria are embedded in a matrix. Because such dense biofilm restricts waste gas diffusion towards the inner particles, it may improve microorganism resistance against high toluene concentrations. Similar observation was made when xylene was biofiltrated using scoria as packing material[10].

Performance under transient conditions: Transient toluene loading experiment was conducted to realize the dynamics of waste gas removal performance in the biofilter. Results of experiments for shutdown and fluctuating charging conditions are shown in figure 7. The pearl millet stack-a biofilter was based on polyurethane foam subjected to a shutdown time of 30 days by stopping the toluene inlet flow to the filter pad. Intermittent sprinkling of the nutrient medium preserved clear air flow of 0.5 L mim^{-1}, with no toluene supplied to the biofilter and moisture content from the top section. The reactor was found to recover well and recover from the starvation process during restart within seventh day. The RE was restored to 72% at the inlet loading of 43 g m^{-3}h-1 and the corresponding EC is 36 g $m^{-3}h^{-1}$.

Saravanan et al[11] analyzed the effect of xylene shock loading in biofilter showing that when the toluene concentration increased, there was no inhibition zone around the reactor, which is usually found in industry. Biomass in biofilter may sustain saprophytic activity with dead cell products, toluene degradation co-products and biofilm exopolysaccharides.

Table 2
Biological Characteristics of isolated Strains

Characteristics	**Isolated -I**	**Isolated -II**
Morphology		
Cell type (Shape)	Rod	Slender, rod shape
Colour	Yellowish White	**Greyish white**
Size	0.4 -0.5 X 1.55 -2.7 µm	1.5–3 mm × 0.5 mm
Arrangement	Isolated	singly or in pairs
Surface	Smooth	**Smooth**
Density	Opaque	**Translucent – Opaque**
Motility	Positive	**Positive**
Biochemical Test		
Gram,s reaction	+	-
Catalase Test	+	+
Spore	**+ central**	-
Indole Production	-	-
Starch hydrolysis	+	+
Citrate utilization	+	-
Methyl red	+	+
Vogas – Proskauer	+	-
Citrate	-	-
H2S Production	-	+
Urease	+	+
Sugar Fermentation		
Glucose	+	+
Fructose	+	+
Maltose	-	+
Lactose	-	-
Sucrose	-	+
Mannitol	+	+
Xylose	-	-
Probable Strain	B. sphaericus	**Pseudomonas** *alcaligenes*

Therefore, the rapid re-adaptation of the biofilter compost may lead to the retention of its biomass activity. This may result in various physiological changes in the microbial cell body and allow microorganisms to recover activity[1]. By rising the inlet toluene concentration to 1.2 g m-3 after recovery of steady state RE pattern, biofilter was subjected to minor shock loads on 52 day of operation. This shock loading did not significantly affect the removal profile and will remain stable to about 68-70%. The pearl millet stack-a biofilter based on polyurethane foam has a good ability to withstand conditions of shutdown and shock load.

Biokinetic constants of the compost biofilter: At the steady state conditions, the plot of all three kinetic equations was studied. The relationship is assuming that oxygen and toluene are not restricted. Figures 8 to 10 demonstrate the linear form of the kinetic expression. Table 3 provides a tabulated comparison of ECmax and K values. ECmax corresponds to the maximum achievable elimination ability in the biofilter over the entire operating conditions period. while Ks contributes the gas-phase toluene sensitivity to the microbial community present in the filter bed. Experimentally obtained ECmax (80 g/m^3h) lower than the model ECmax expected during a steady state operation. If the acid metabolites produced in biofilter are periodically removed by washing with a mineral medium, then the biofilter can achieve higher elimination efficiency.

Conclusion

In the present study, the performance of biofilter in toluene removal was improved by using pearl millet stack – poly urethane foam mixture as packing material. The biofilter showed a quick response to shock loadings and has a good capacity to tolerate 30 days of non-use periods, and it restored shortly. The dynamics of the toluene removal was understood by profile of normalized concentration along the filter bed height.

Bacillus sphaericus and *Pseudomonas alcaligenes* isolates identified were found to be active and potent microorganism for biodegradation of toluene. Lineweaver-Burk, Eadie – Hofstee and Hanes–Woolf equation was able to explain the behaviour of the biofilter at steady state operation.

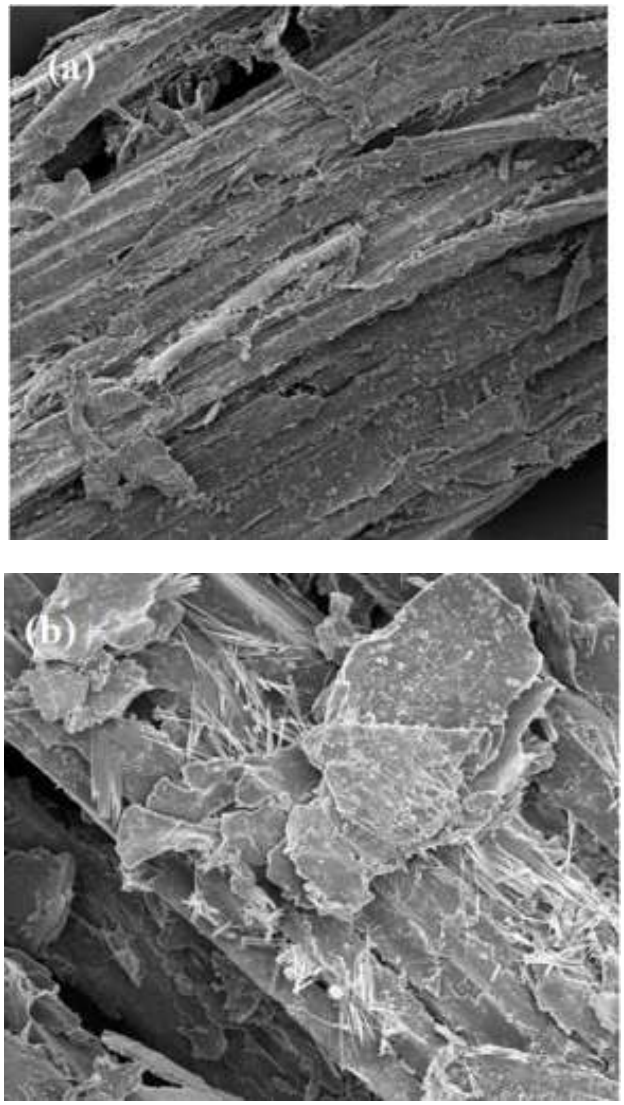

Figure 6: Scanning electron micrograph of the filter media (a) before and (b) after experimentation.

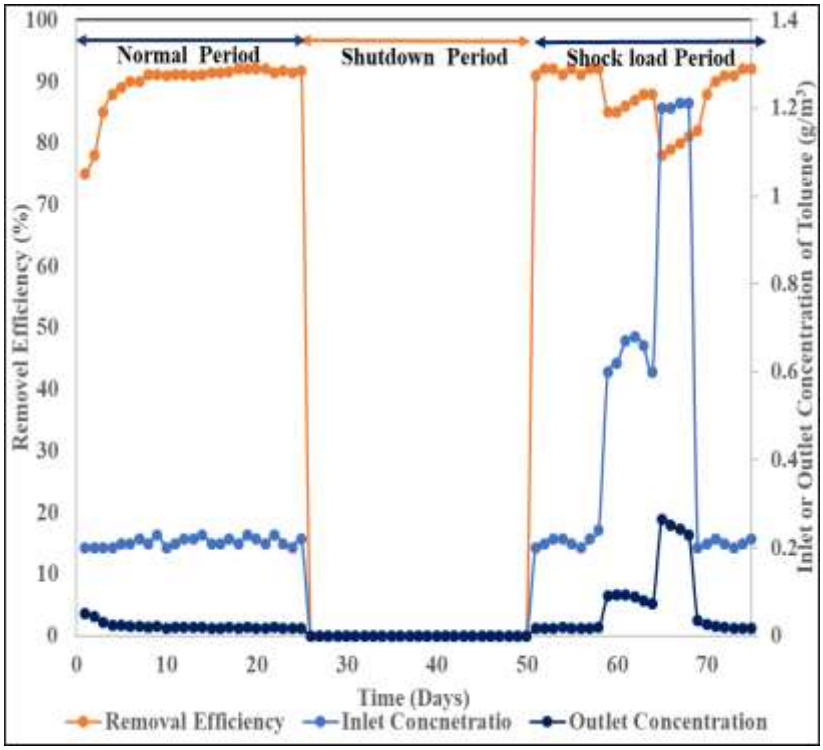

Figure 7: RE of the biofilter during transient operating conditions

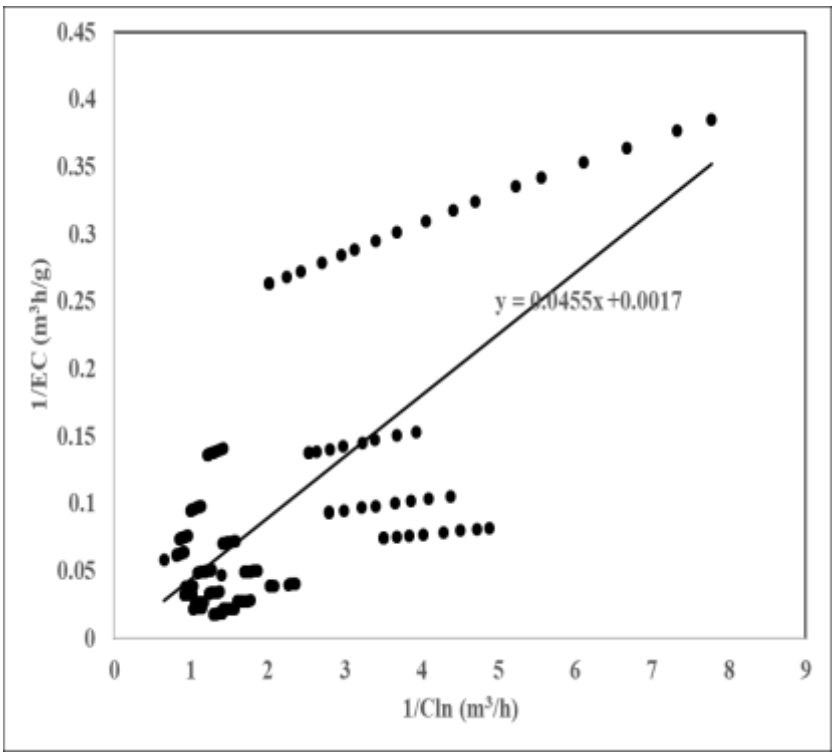

Figure 8: Lineweaver-Burk plot for the removal of toluene biofilter

Table 3
Kinetic constant for all three model for the removal of toluene

S.N.	Model	Kinetic Constant		Experimental EC $_{max}$ (g/m³h)
		K (g.m³)	EC_{max} (g/m³h)	
1.	Lineweaver-Burk	1.22	70	85
2.	Eadie - Hofstee	1.09	81	
3.	Hanes–Woolf e	1.31	85	

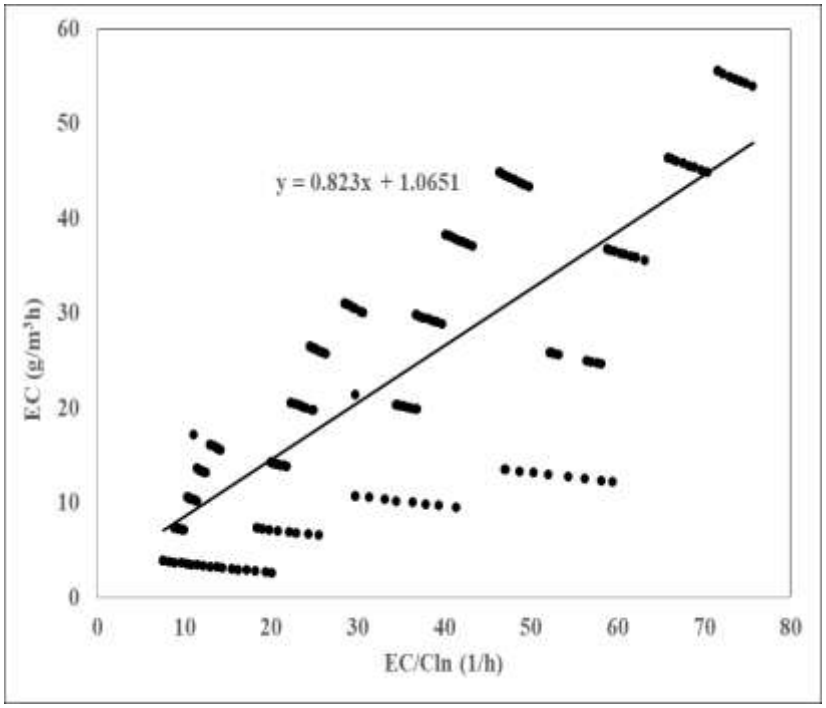

Figure 9: Eadie - Hofstee plot for the removal of toluene biofilter

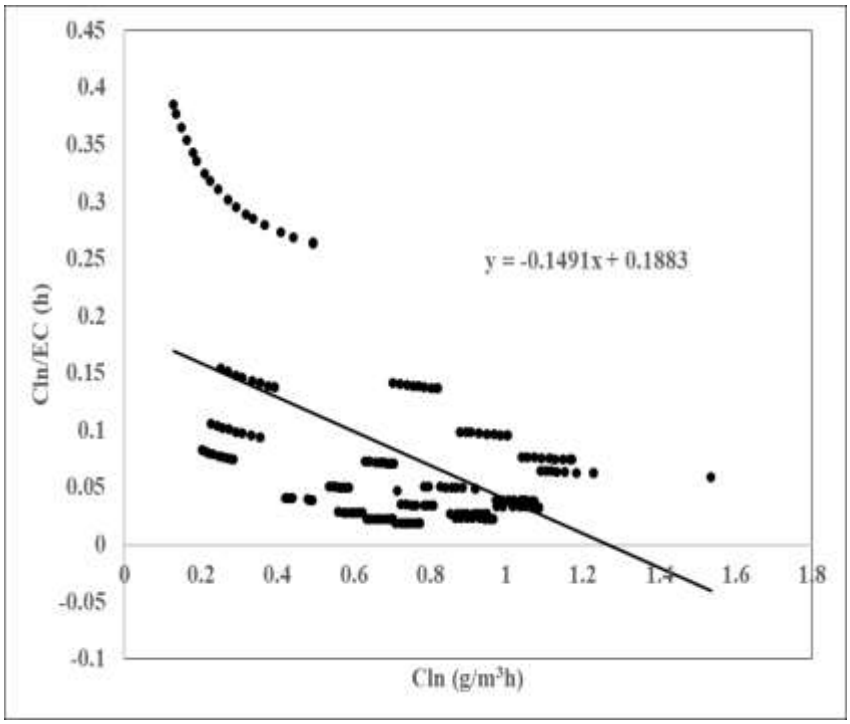

Figure 10: Hanes–Woolf e plot for the removal of toluene biofilter

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