



Performance Studies on Biofilter for VOC Removal Under Various Organic Loading Rates and Gas Empty Bed Residence Times

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Abstract: A biofilter which comprised of a closed compartment, a casuarina seed and a nutrient solution distributor was established and evaluated under inlet concentration ranging from 0.2 to 1.2 g m⁻³ and gas empty bed residence times (EBRTs) of 0.7 to 2.81 min. Using xylene as model VOC, the startup of the biofilter lasted approximately 10 days. The removal efficiency decreased from 99% to 82.2% when inlet concentration was increased from 0.2 to 1.2 g xylene m⁻³ at 2.81 min, but did not decline significantly when the EBRT was reduced from 1.47 to 0.7 min at 0.2 g m⁻³. Biomass concentration did not increase significantly within the casuarina seeds during the 365 day's operation as observed through the acrylic tube of the biofilter. The biofilter is suitable for treating waste gases with low VOC concentrations even at high gas flow and over long periods.

Keywords: Biofiltration, Biomass, xylene, Biofilter, casuarina seed, EBRT

I. Introduction

Volatile organic compounds such as benzene, toluene, xylene, phenol, halogenated aromatic compounds, chloroform, trichloroethylene are the major products of petroleum and fine chemical industries and the most frequently used organic solvent. Some of them are suspected as carcinogens and emit offensive odours to the air and pollute soil and groundwater. They frequently enter soil, sediments and groundwater because of leakage from underground storage tanks pipelines, accidental spills, improper practices and leaching landfills [1]. Among the many VOCs the xylene is widely used as solvent in printing, rubber, synthetic fiber, plastic, insecticide, pesticides and leather industries and as cleaner and paint thinner [2]. It causes irritation of eyes and mucous membranes at concentrations below 200 ppm and is narcotic at high concentrations. Acute exposure to high vapour concentration of xylene may cause chemical pneumonitis, hemorrhage into the air spaces, and pulmonary edema. Therefore, it is necessary to eliminate xylene from the environment, in order to prevent the significant impact on ecosystem and public health. Out of various techniques such as incineration, ozonation, combustion, adsorption, biofiltration has emerged as cost effective technology for eliminating odorous and toxic volatile organic compounds (VOCs) such as xylene from waste gas streams [3]. Biofiltration is becoming an established air pollution control technology for the control of volatile organic compound (VOC) and odor emissions from waste gas streams. The application of biofiltration technology throughout the world has increased rapidly in recent years [4]-[12]. This is largely due to, low costs for operation and maintenance and their environmental soundness over conventional methods. Biofiltration uses microorganisms fixed to a packing material to breakdown pollutants present in an airstream. 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 [13].

Biofilter systems for VOCs control are greatly affected by the selection of the packing media and microorganisms. Since the inoculation of definitely adapted microorganisms into biofilter media reduces the start-up period and may increase the biofilter performance [14], activated sludge, extracts from contaminated soils, and specially grown cultures have been used as inoculums [15]. The ideal packing media are characterized by high specific surface area, low volumetric density, minimal pressure drop, and suitable surface for the attachment of microorganisms [16]. Various kinds of materials have been used as a packing material in biofilter's. Although natural materials that are available in large quantities from agricultural operations and plants biomass may have potential to be used as low cost adsorbent, widely available and environmental friendly [17]. Such as compost, peat, and wood chip have many advantages, including high physical adsorption capacity, excellent water holding capacity, favorable biological media for microbial growth and activity for their nutrient supply [18], they may be subjected to operation difficulties such as compaction and channeling [19],

thus leading to lower process efficiency and short exchange cycle [20], [21]. To overcome such problems, some of the researchers mix some synthetic materials like ceramic, glass, polystyrene, perlite, activated carbon, sandstone, and clay to the packing media [22]. *Casuarina equisetifolia* is a genus of 17 species in the family of Casuarinaceae, native to Australia, southeastern Asia and islands of the western Pacific Ocean. Its wood can be used for various purposes such as for pole and veranda construction. The plant is known to store tannins and proline as well as being a nitrogen fixing plants. *Casuarina equisetifolia* plant is highly valued as an important species for erosion control and has ability as an excellent windbreak [23]. *Casuarina equisetifolia* plant seeds are assumed to be plant biomass since it is abandoned in nature. Therefore, the aim of this study is to investigate the potential of *Casuarina equisetifolia* plant seeds as an packing material for the removal of xylene in biofilter.

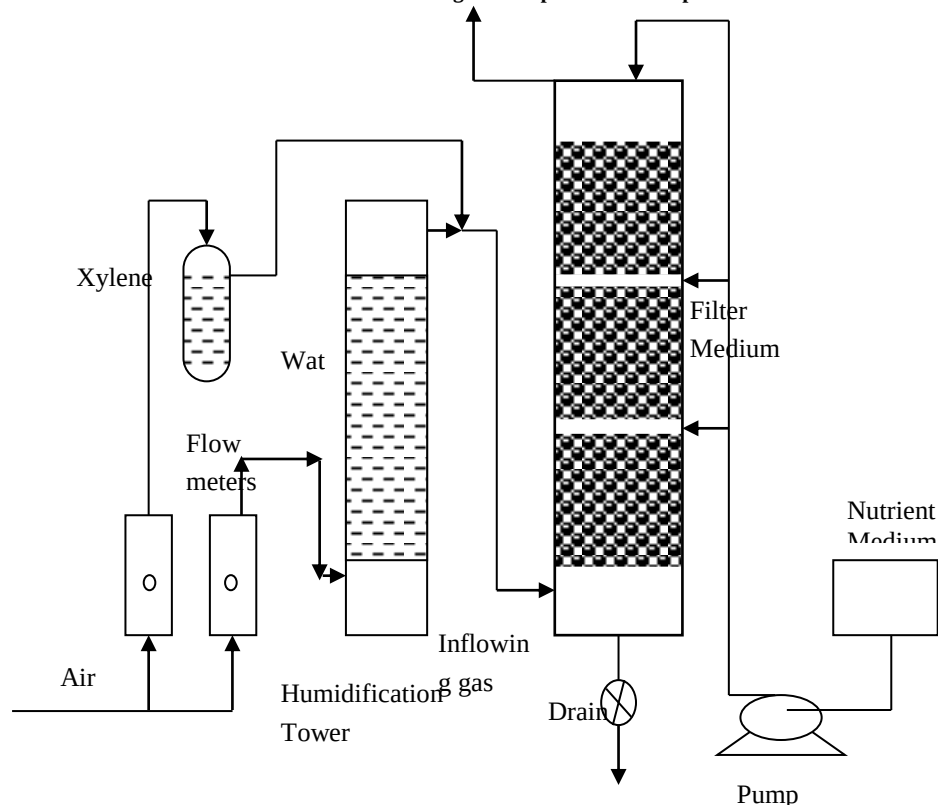
Some researchers reported the removal of xylene in compost and perlite packed xylene-degrading biofilter inoculated with mixed consortia taken from a sewage treatment unit [24], [25]). The good packing material for VOC treatment should have low inherent nitrification activity level and high slow-releasing nitrogen content rather than high soluble nitrogen content. Jorio *et al.* (2000) investigated removal of xylene in a peat biofilter mixed with conditioning agents and inoculated with a microbial consortium and obtained an elimination capacity of 67, 52, and 41 g /m³ h at gas flow rates of 0.4, 0.7, and 1 m³/ h, respectively. The understanding of the biodegradation kinetics play an important role to improve our knowledge about biodegradation reaction and thus may help in a better design for in situ application of bio-remediation. A number of models have been proposed to describe the kinetics of biodegradation of VOCs [25]. The performance of the biofiltration experiment is evaluated for the period of 391 days by changing the inlet concentration of xylene and air flow rates. The performance of biofilter is gauged in terms of removal efficiency and elimination capacity.

II. Materials and Methods

A. Biofilter system

The biofilter consisted of a cylindrical pipe had an inner diameter of 5cm and a height of 100cm; it consisted of a acrylic pipe and two perforated acrylic plates at both ends. The nutrient solution distributor at the top of the column and uniform gas distribution in the bottom of the reactor, and was impermeable for waste gases (Fig. 1). A clean air stream and a xylene vapor stream were used to synthesize the waste gas streams. The mixed gases entered in to the biofilter through the gas inlet on the bottom of the chamber and passed through the cassirine seed as packing material where biodegradation took place by microorganisms attached to the seed. The purified gas was emitted through the gas outlet located at the top of the reactor. The nutrient solution was pumped into the biofilter at a rate of 6.0 L per day and uniformly distributed throughout the packing material.

Figure 1 Experimental setup



B. Filter material

The material on which the biofilm grew in the biofilter consisted of casuarina seed obtained from the agricultural land. The porosity and pore size of the seed were 98% and approximately 12 pores per centimeter, respectively. The characteristic parameters of the casuarina seed are listed in Table 1. Seed cultures fresh activated sludge was taken from a secondary sedimentation tank at paper industry and used for seeding the biofilter.

Table 1 Characteristics and Chemical Composition of casuarina seed

Characteristics	Value	Units
Hemicellulose	60.7	%
Aplha- Cellulose	24.1	%
Lignin	45.2	%
Ash	2.2	%
Porosity	30	PPI
Radial Thickness	30	(mm)
Outer diameter	140	(mm)
Inner diameter	80	(mm)
Height	100	(mm)
Volume	0.044	m ³

C. Chemicals and mineral medium

The nutrient solution (Basal Salts Medium (BSM) about 10mlmin⁻¹ is continuously sprayed two times in a day for 30 min on the top of the packing media through the nutrient distribution system. The BSM solution used for the continuous tests has the following composition per liter of water: K₂HPO₄ - 0.91g; Na₂HPO₄·2H₂O - 2.39g; KNO₃ - 2.96g; (NH₄)₂SO₄ - 1.97g; FeSO₄·7H₂O - 0.2g; MgSO₄·7H₂O - 2.0g; MnSO₄·7H₂O - 0.88mg; Na₂MoO₄·2H₂O - 1mg; CaCl₂ - 3 mg; ZnSO₄·7H₂O - 0.04 mg; CoCl₂·6H₂O - 0.04 mg. All the chemicals used are AR grade with more than 99% purity

D. Analytical methods

Xylene concentrations in both the influent and the effluent were measured using a IR detector .

E. Operation of the biofilter

The casuarina seed was immersed into clean water for about 2 days, then placed into the activated sludge for the biofilter microbial inoculation. After about a week, the packing material was taken out from the activated sludge and filled in the reactor . Subsequently, the synthetic waste gas and nutrient solution were introduced into the biofilter. The moment at which the media was introduced into the biofilter was counted as the initial time of biofilter operation (the 1st day). The reference conditions were set at gas EBRT of 2.81 min. During operation, the biofilter was adjusted to the reference level for about week before and after a change in an operating parameter. The concentration of xylene in the synthetic waste gases can be adjusted by changing the flow rate of the clean air stream and the amount of neat xylene vapor, while the gas EBRT can be altered by changing the flow rate of the clean air stream. When the organic loading rate was increased, the inlet gas flow rate remained constant but the xylene concentration in the influent increased. The gas EBRT was decreased by increasing the flow rate of the clean air stream, so that the inlet VOC concentration decreased while the organic loading rate remained unchanged. When the effect of gas EBRT on the biofilter performance at a constant incoming xylene concentration was studied, the gas flow rate and organic loading rate were increased and the gas EBRT was decreased. While the organic loading rate, gas EBRT, or gas flow rate was changed, all other parameters including nutrient solution concentration, nutrient solution feed rates and temperature, etc., remained constant. The biofilter was operated for 391 days. Days 1–50 was considered as the initial startup stage and the biofilter was kept at the reference conditions. The operation conditions after the initial startup stage are listed in Table 2. The biomass that accumulated in the filter of the biofilter was not removed

Table 2. Operation conditions during the whole experiment.

Stages	Days of operation	Gas flow rate	Inlet xylene Concentration	Removal Efficiency
1.	1-50	0.03	0.7	87
2.	51-60	0.06	0.2	91.9
3.	61-105	0.06	0.4	79.9
4.	106-115	0.09	0.2	84.3
5.	116-165	0.09	0.6	69.6
6.	166-175	0.12	0.2	72
7.	176-225	0.12	0.8	61.5

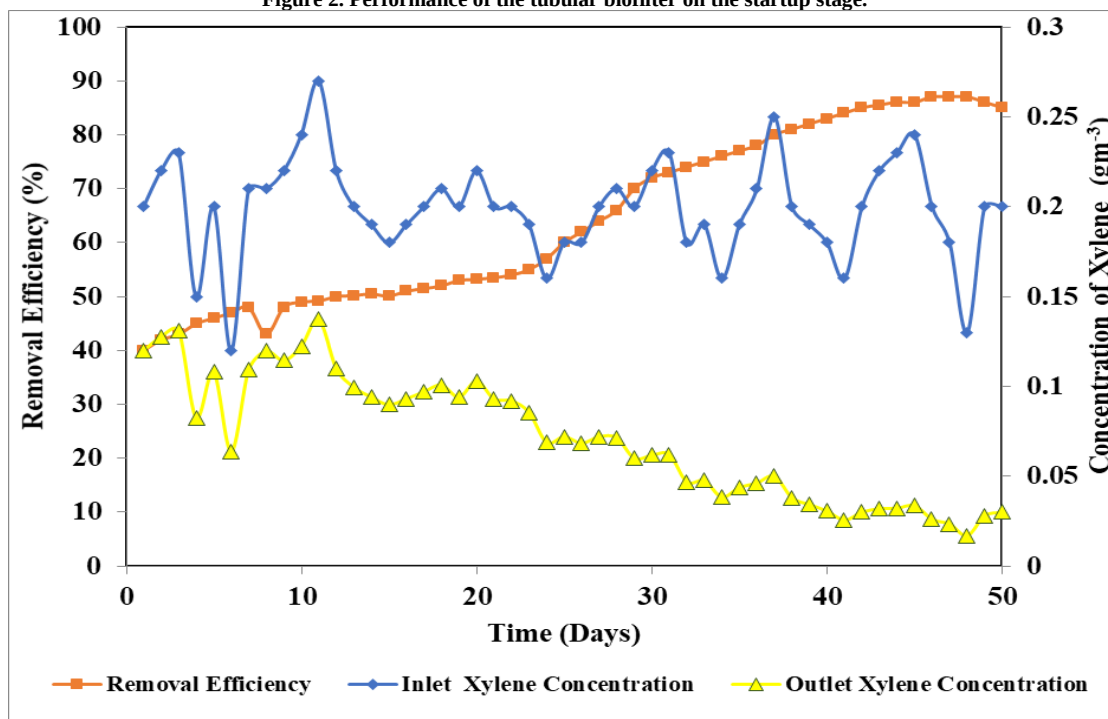
8.	226-234	0.12	0.2	64.5
9.	235-244	0.06	0.2	97
10.	245-260	0.03	0.4	98.2
11.	261-276	0.03	0.2	98.1
12.	277-300	0.03	0.6	98.8
13.	301-321	0.03	0.8	98.1
14.	322-331	0.06	0.2	96
15.	332-350	0.03	0.4	98
16.	351-360	0.03	0.2	99
17.	361-370	0.03	0.4	98
18.	371-375	0.03	0.2	99.1
19.	376-380	0.06	0.8	72
20.	381-390	0.03	0.2	98

III. Results and discussion

A. Biofilter performance on the initial startup stage

The biofilter was started and operated for 50 days at the reference operation condition. When inlet toluene concentration was maintained at 0.2 gm^{-3} in the initial startup stage, the outlet styrene concentration decreased while the RE increased with time (Fig. 2). Xylene REs were less than 50% in the first 7 days, and reached 95% at day 50.

Figure 2. Performance of the tubular biofilter on the startup stage.

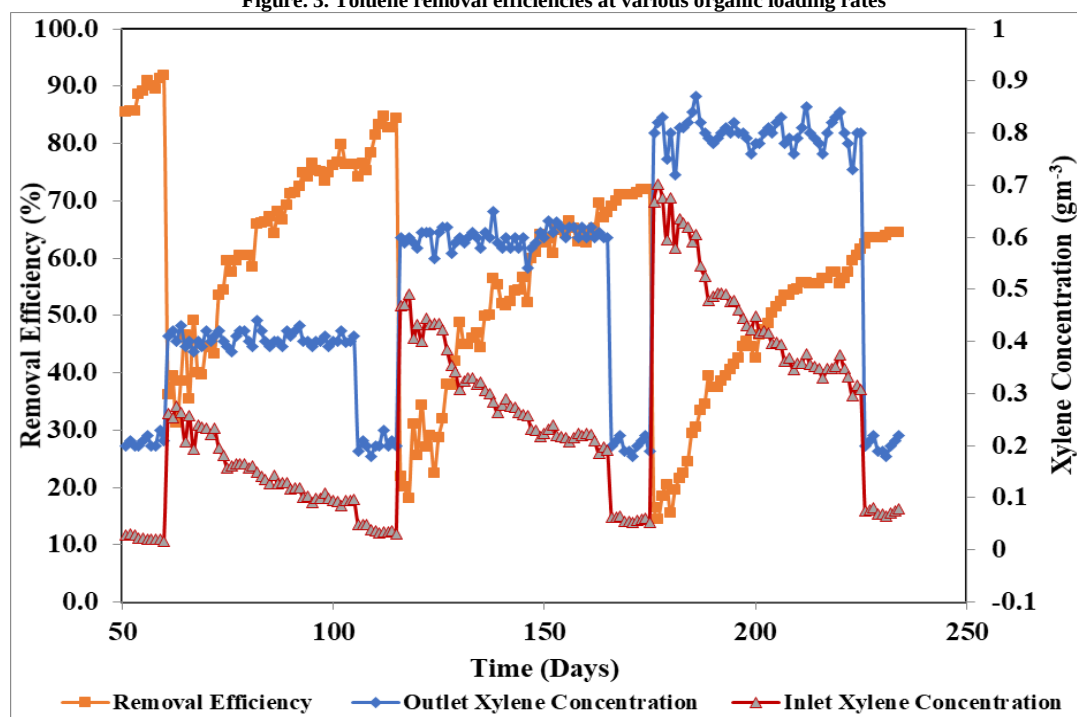


B. Effect of organic loading rate

Following the startup stage, the biofilter was operated at organic loading rates of 16.2, 30.5, 45 and $60 \text{ g m}^{-3} \text{ h}^{-1}$ from day 51 to day 202, and the corresponding average REs were 99%, 84.5%, 72.0% and 52.2%, respectively (Fig. 3). The biofilter could achieve a higher styrene RE than a peat biofilter (27) or an agricultural by-product-based biofilter (28) at a lower organic loading rate and a shorter gas EBRT. After the organic loading rate was doubled, it took the biofilter approximately 30 days to reach a relatively stable RE (Fig. 3), while it only took 2–4 days for the biofilter to recover its RE at the reference operation condition after changing from a high organic loading rate. Saravanan *et al.* 2009 also reported that the reacclimation of biofilter performance was delayed by increasing influent concentrations for biofilters. Therefore, the biofilter is recommended for treating waste gases with a stable organic loading and a moderately high or low VOC concentration.

As shown in Fig. 3, the ECs were 16.03, 25.77, 32.4, and 31.32 g m⁻³ h⁻¹ at organic loading rates of 16.2, 30.5, 45 and 60 g m⁻³ h⁻¹, respectively. Though it increased with an increase in the xylene loading rate, the EC was lower than the load at the highest organic loading rate. The EC of biofilter packed with 75 cm high pressmud achieved 34.0, 49.2 and 113.0 g toluene m³h⁻¹ at influent loading rate of 34.4, 70 and 140 gm³h⁻¹, respectively [11].

Figure. 3. Toluene removal efficiencies at various organic loading rates

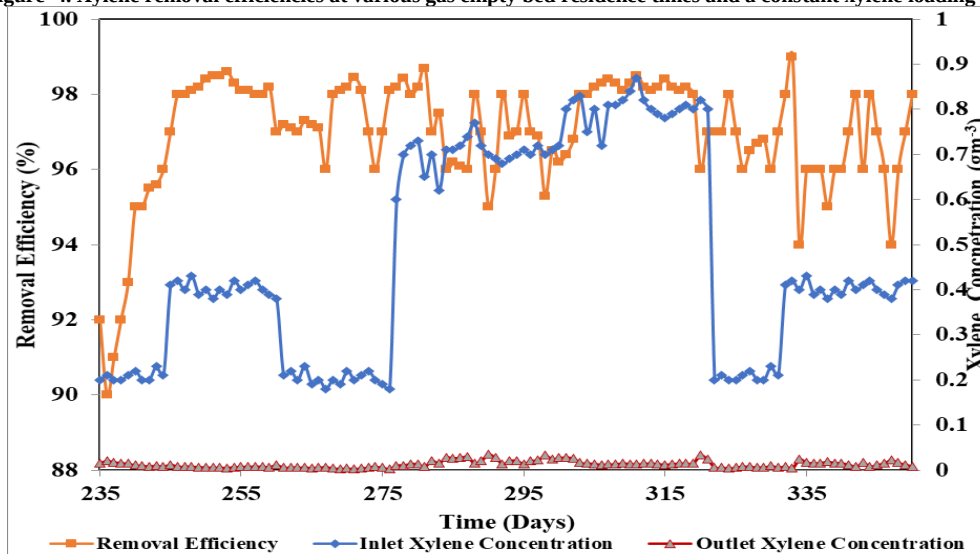


C. Effect of gas EBRT at constant xylene loading rate

The biofilter performance at gas EBRTs of 2.81, 1.47, 0.9, 0.7 and 0.3 min, was evaluated from day 226 to day 321 when the organic loading rate remained at 16.2 g m⁻³ h⁻¹ (Fig. 4). The corresponding xylene concentrations in the incoming gas streams were 0.2, 0.4, 0.8, 1.0 and 1.2 g m⁻³, respectively. The RE did not change dramatically when the gas EBRT was reduced at an organic loading rate of 16.2 g m⁻³ h⁻¹. At a gas EBRT of 0.3 min from day 300 through 309, the RE was only a little lower than that stabilized at the other gas EBRTs, but still achieved a high and stable performance even at a gas EBRT of 5 s. The outlet xylene concentration was otherwise uniformly less than 1.2 g m⁻³ in the evaluation period from day 203 to day 338. Under a constant organic loading rate of 16.2 g m⁻³ h⁻¹, the calculated EC was 15.5, 15.3, 15.4, 15.2, and 14.6 g m⁻³ h⁻¹ at gas EBRT of 2.81, 1.47, 0.9, 0.7 and 0.3 min, respectively. The EC of biofilter packed with 75 cm sugarcane bagasse achieved 15.2, 14.1, 9.5 and 8.2 g m⁻³ h⁻¹ at a toluene loading rate of 16 g m⁻³ h⁻¹ and gas EBRTs of 30, 15, 10 and 7.5 s [12]. It can be seen that the TBF can achieve a high removal performance at a short gas EBRT.

Fig. 4 also shows that the acclimation times needed for the biofilter performance recovery were only 1–3 days, such as on day 224 and day 318. In contrast during the period of days 250–272 when the biofilter was at an unsteady state, the biofilter needed a relatively long period of about 5 days to reach a high RE over 95.4%. Compared to a time of approximately 1 month needed for achieving a high and stable biofilter performance when the organic loading rate was doubled (Fig. 3), a much shorter period of time was needed when the gas EBRT was changed in this study. So, the biofilter performance is steady at various gas EBRTs from 2.81 to 0.3 mins over a long period of operation, and a short gas EBRT of less than 0.7 mins would be enough for the biofilter to achieve a high and stable performance at middle to low xylene concentrations. Although the small thickness of the filter media is helpful for decreasing the footprints of the bioreactors, problems exist such as a longer duration for the biofilter startup and deteriorating performance at high organic loading rates or short gas EBRTs or over a long period of operation. In this study, a long period of time was needed for successful start-up of the biofilter or stabilization at a high performance, and the RE dropped when the organic loading rates were increased.

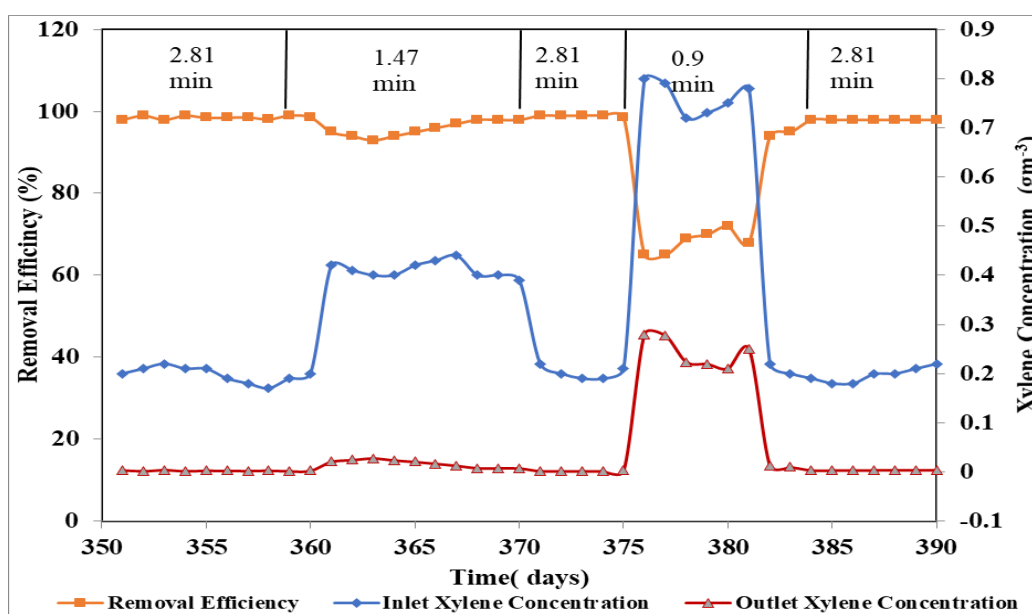
Figure 4. Xylene removal efficiencies at various gas empty bed residence times and a constant xylene loading rate



D. Effect of gas EBRT at a constant incoming pollutant concentration

The selection of a reasonable gas flow rate at fixed pollutant concentrations is important for the design of a full-scale biofilter system. Therefore, the effect of gas EBRT on biofilter performance under a constant incoming xylene concentration of 0.8 g m^{-3} was evaluated at gas flow rates of 0.03 , 0.09 and $0.12 \text{ m}^3 \text{ h}^{-1}$, and the corresponding gas EBRT were 2.81 , 0.9 and 0.7 min and the xylene loading rates were 16.2 , 37.3 and $74.6 \text{ g m}^{-3} \text{ h}^{-1}$, respectively. The biofilter achieved 99.2% , 97.4% and 83.5% xylene removal when the gas EBRT was 2.81 , 0.9 and 0.7 min, respectively, and the corresponding EC were 18.6 , 36.4 and $63.6 \text{ g m}^{-3} \text{ h}^{-1}$ (Fig. 5). The EC of $63.6 \text{ g m}^{-3} \text{ h}^{-1}$ at incoming concentration of 0.8 g m^{-3} and gas EBRT of 2.81 min was a little higher than that of $16 \text{ g m}^{-3} \text{ h}^{-1}$ at incoming concentration of 1.2 g m^{-3} and gas EBRT of 1.47 min (Fig. 3). When the reference condition was changed as shown in Fig. 5, the biofilter recovered to the reference level in a period of about 1–2 days, which is shorter than that reported in Figs. 3 and 4. That the incoming xylene concentration as illustrated in Fig. 5 was maintained constant, might contribute most to this phenomenon. The biofilters were developed to try to reduce construction and maintenance costs and to maintain the advantages of sponge materials with high porosity and high specific areas of the outermost medium bed and consequent low approaching gas velocity. [25]–[28]

Figure 5. Xylene removal efficiencies at various gas empty bed residence times



E. Characterization of Adsorbent

Scanning electron microscopy observations Figure 6a and b shows the Casuarina seed before and after biofiltration respectively. A high microbial thickness was shown at after experimentation when compared to

before. Diverse microbial structure, such as bacterial colonies, budding yeast, mycelial structures, and also some noncolonized regions on the surface of *Casuarina* seed were observed (Figures 6 b). These observations revealed a heterogeneous biofilm, in contrast to that of a smooth and homogeneous biofilm which is generally assumed in the models used to describe biofilter behaviour. In addition, the dominant microorganisms seen in the final day of the biofiltration when compared to the others. These results implied that the microorganisms indigenous to the bed medium can adjust themselves to be available in each biofiltration system. Inoculation was not necessary in our study

Figure. 6 . (a) SEM of *Casuarina equisetifolia* seeds before biofiltration

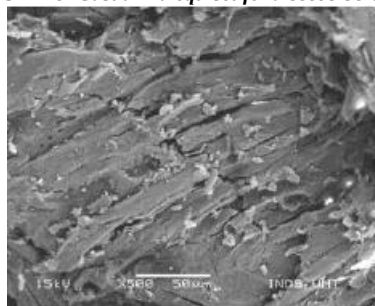
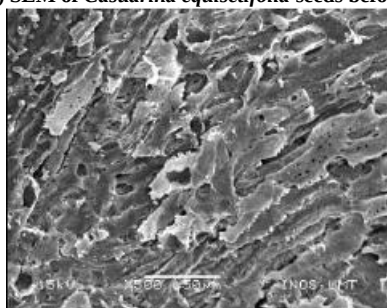


Figure. 6 . (b) SEM of *Casuarina equisetifolia* seeds before biofiltration



VI. Conclusions

A biofilter consisting of a *Casuarina* seed as packing material was successfully started in 7 weeks for xylene removal. The RE decreased when increased xylene loading rate but did not decline significantly when reduced gas EBRTs. High ECs at high organic loading rates could be obtained under low pollutant concentrations. Excessive biomass accumulation was singularly occurred on the biofilter. The biofilter is suitable for treating waste gases with low xylene concentrations even at high flow rates over long periods of operation, reduces the footprint of gas biofilters, and overcomes the deterioration in removal observed with traditional biofilters.

V. References

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