

## Performance of a hollow fiber membrane bioreactor for the treatment of gaseous toluene

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**ABSTRACT.** A hollow fiber membrane bioreactor (HFMB) has emerged as a treatment technology for the air emission of biodegradable volatile organic compounds (VOCs). The hollow fiber membrane serves as a support for VOC-degrading microorganisms and provides a large surface area for the VOC mass transfer. In this study, a bioreactor system using a submerged hollow fiber membrane module was applied to investigate feasibility and biodegradation capacity of the system for the treatment of gaseous toluene. The HFMB was operated at different inlet toluene loading rates of 50, 100, 500 g/m<sup>3</sup>.h, and overall removal efficiencies were maintained in the range of 70 – 80%. In addition, elimination capacities (EC) were increased up to 800 g/m<sup>3</sup>.h, which was substantially higher than maximum ECs for toluene reported in the biofiltration literature. These findings imply that the HFMB be a feasible alternate over conventional packed-bed type biofilters. However, an increase in pressure drop across the hollow fiber membrane module was observed with the increasing operation time. As a result, more integrated research is required to improve the HFMB performance as well as to minimize the pressure drop across the membrane.

### 1 INTRODUCTION

The removal of a wide variety of odorous and volatile organic compounds (VOCs) from contaminated waste gas streams is a major environmental concern. Packed-bed type biofiltration technologies such as biofilters have successfully treated waste streams with low to moderate concentrations and are recognized as a cost-effective alternative to traditional treatment methods (van Groenestijn and Hesselink, 1993). However, biofilter performance over extended periods of operation can be unreliable due to excessive biomass accumulation, which leads to clogging in biofilter columns, as well as inactivation of the pollutant-degrading microorganisms (Smith *et al.*, 1996; Song and Kinney, 2000). These problems become severe when biofilters are subjected to high organic loading rates.

To overcome the problems commonly encountered in the biofiltration technologies, a novel treatment method using hollow fiber membranes has been developed. Compared

to packed-bed biofilters, hollow fiber membrane bioreactors (HFMBs) offer the following advantages (Kennes and Veiga, 2001; He *et al.*, 2004):

- HFMBs provide a relatively large interfacial surface area between the gas and the liquid phases, yielding high mass transfer rates of the less soluble organic compounds to the liquid phase. The specific surface area of the membranes can be as high as  $6000 \text{ m}^2/\text{m}^3$ . Furthermore, the surface area remains constant because the two fluids are separated by a membrane with fixed geometry.
- HFMBs are less likely exhibit flow channelling, liquid flooding and foaming than packed-bed biofilters.
- It is easy to continuously remove biomass from the liquid phase; therefore HFMBs are not prey to clogging at high VOC loading rates. Also, it is possible to maintain a high quantity of active biomass in the system, resulting in high pollutant removal efficiencies.
- There is no need to humidify the inlet air streams to externally supply moisture. In addition, it is easy to control the concentrations of essential nutrients in optimal ranges to maintain high microbial activity.

Several pioneering studies have showed that HFMBs can effectively degrade a wide variety of VOCs including TCE (Pressman *et al.*, 2000), toluene (Ergas *et al.*, 1999) and odorous compounds such as dimethyl sulfide (Langenhove *et al.*, 2004). The common operating mode of the HFMBs reported in the literature was a membrane contactor type, where waste gas streams are blown through the lumen (inside) of the hollow fibers and pollutants from the gas phase diffuse through the membranes to the liquid phase on the shell side (outside) of the hollow fiber modules (Kennes and Veiga, 2001).

It is also possible to operate HFMBs in a membrane diffuser type, where waste gases are actually diffused into the liquid phase through hollow fiber membranes as fine bubbles. In this operating mode, VOCs supplied through the membranes can be degraded by both fixed biofilms attached in the outside of the membranes and suspended microorganisms in the liquid phase. This type of HFMB operation is believed to provide high bioreactor performance as well as high flexibility in system operation and modification. The study presented herein was designed to evaluate a diffuser type HFMB subjected to high toluene loading rates. A series of bioreactor experiments were conducted over a range of toluene loading rates, and bioreactor performance and elimination capacities (ECs) were determined.

## 2 MATERIALS AND METHODS

### 2.1 Reactor configuration and operation

A lab-scale bioreactor used in this study consisted of an acrylic container and a hollow fiber membrane module at the bottom of the container, as shown in Figure 1. The bioreactor was filled with a liquid (a total volume of 3 L) made with a suspended microbial culture capable of degrading toluene and a nutrient medium consisting of 1.36 g/L  $\text{KH}_2\text{PO}_4$ , 1.42 g/L  $\text{Na}_2\text{HPO}_4$ , 0.5 g/L  $(\text{NH}_4)_2\text{SO}_4$ , 3.03 g/L  $\text{KNO}_3$  and trace elements used by Song and Kinney (2000). The liquid was constantly recirculated using a water pump at a flowrate of 6 L/min. To provide nutrients and waste possible byproducts, a 0.3 L of the liquid medium was withdrawn and replaced with a fresh one on a daily basis.

The compressed air supply to the bioreactor was contaminated with toluene vapor by slowly injecting pure toluene into the air stream using a syringe pump (KD Scientific, USA). The contaminated air stream was supplied to the bioreactor through inlet ports

connected to the membrane module (Figure 1), and then diffused through the membrane micro-pores into the liquid phase where toluene biodegradation took place.

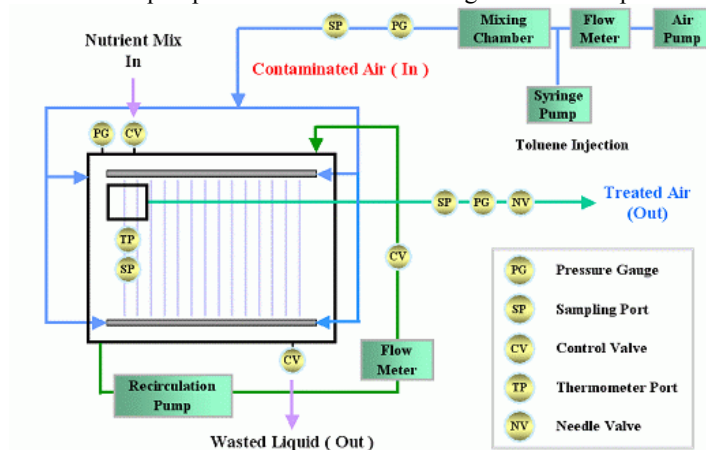


Figure 1. Schematic of the hollow fiber membrane bioreactor system.

Three bioreactor experiments were sequentially performed at the inlet toluene loading rates of 50, 100, and 500 g/m<sup>3</sup>.h, respectively, for 5 days each. During the experiments, the gas contact time was remained constant at 30 seconds corresponding to a gas flowrate of 6 L/min. To ensure that there was no carry over effect between experiments, the bioreactor and the membrane module were cleaned with a NaOCl solution prior to bioreactor start-up.

### 2.2 Hollow fiber membrane

The membrane module (KMS, Korea) used in this study was originally manufactured for an application of biological wastewater treatments. For this application to treat vapor-phase toluene, the module was slightly modified to fit into the bioreactor. Table 1 summarizes the characteristics of the membrane used in this study.

Table 1. Characteristics of the hollow fiber membrane used in this study.

Items	Specification
Material	polyethylene
Nominal pore size	0.4 μm
O.D. / I.D. / Wall thickness	650 / 410 / 120 μm
Fiber length	0.3 m
Number of fibers in a module	7 × 75

### 2.3 Analytical methods

To determine overall toluene removal efficiencies and elimination capacities (ECs), gas samples were collected from each sampling port with 0.5-mL gas-tight syringes (Hamilton, USA) and immediately analyzed using a gas chromatograph (GC) (Agilent 6890, USA) equipped with a flame ionization detector. The EC curves were determined by increasing the inlet concentration stepwise for two hours, similar to the procedure used by Deshusses and Johnson (2000). Five to six different loadings were applied and

the EC values were calculated for each loading using the overall toluene removal efficiency observed.

### 3 RESULTS AND DISCUSSION

#### 3.1 Removal efficiency

Three HFMB experiments were performed for 5 days each at the inlet toluene loading rates of  $50 \text{ g/m}^3\cdot\text{h}$  (referred to as L-50),  $100 \text{ g/m}^3\cdot\text{h}$  (L-100), and  $500 \text{ g/m}^3\cdot\text{h}$  (L-500), respectively. The loading rates were corresponding to the inlet toluene concentrations of approximately 100, 200 and 1000 ppm, respectively. In experiment L-50, the toluene removal efficiency was approximately 80%, and the removal efficiency remained almost constant throughout the experiment (see Figure 2). At the loading rates of 100 and  $500 \text{ g/m}^3\cdot\text{h}$ , the toluene removal efficiencies slowly increased after bioreactor start-up and were maintained at greater than 75%. With increasing loading rates, the concentration of biomass in the liquid phase increased (data not shown). These experimental results indicate that the HFMB operated in the diffuser mode is a promising method for the treatment of gaseous VOCs, and it allows biological VOC abatement technologies to expand their applications to higher organic loading rates. However, it is not clear that how the changes in active biomass quantity in the system affects the HFMB performance. Further investigation of changes in active biomass quantity and HFMB performance at various organic loading rates is required to better assess and predict the long-term efficacy of HFMBs operated in the diffuser mode.

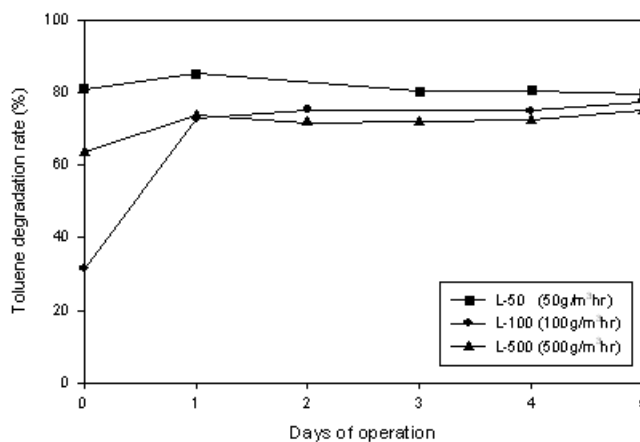


Figure 2. Overall toluene removal efficiencies at different loading rates.

#### 3.2 Elimination capacity

After each experiment was terminated (on day 5), the elimination capacity (EC) test was performed by increasing the inlet concentration stepwise. As shown in Figure 3, in experiments L-50 and L-100, EC values in a range of  $300 - 400 \text{ g/m}^3\cdot\text{h}$  were observed at an inlet toluene loading of  $600 \text{ g/m}^3\cdot\text{h}$ . In experiment L-500 where the HFMB was subjected to the highest loading during the 5-day operational period, the EC value increased up to  $800 \text{ g/m}^3\cdot\text{h}$ , which was substantially higher than maximum ECs for toluene reported in the biofiltration literature (Deshusses and Johnson, 2000). These

findings imply that the HFMB be a feasible alternate over conventional packed-bed type biofilters.

The maximum EC can be generally defined as the point where the EC curve with a curvature achieves its highest value. In this study using the HFMB, each EC curve increased with increasing inlet loading and did not clearly show a maximum point. Especially, at L-500 (the inlet toluene loading rate of  $500 \text{ g/m}^3\cdot\text{h}$ ), the EC value increased almost linearly up to the inlet loading rate of  $1100 \text{ g/m}^3\cdot\text{h}$  applied in this test. A higher EC value would have been achieved if a higher inlet loading had applied in this test. However, a higher toluene loading greater than  $1100 \text{ g/m}^3\cdot\text{h}$  was not applied to protect the system from any possible damaging and to prevent from receiving a toxicity effect on toluene-degrading microorganisms.

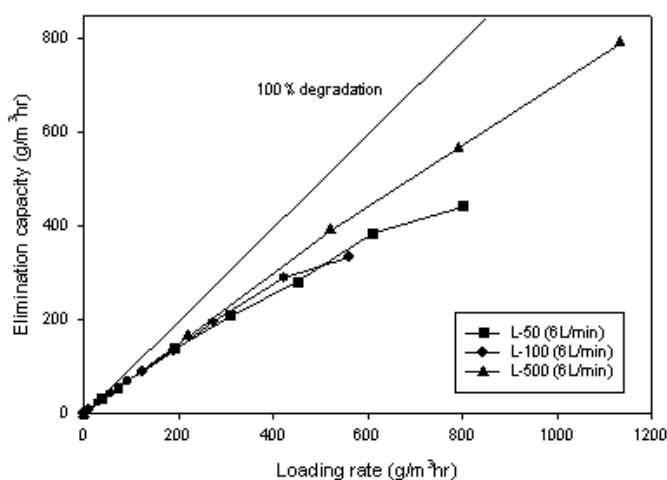


Figure 3. Elimination capacity curves determined at the end of each loading experiment.

In spite of its outstanding performance, the HFMB showed an increase in pressure drop across the hollow fiber membrane module with the increasing operation time (data not shown), indicating that biofouling on the membrane surface by microorganisms played a major role on the pressure drop. As a result, more integrated research is required to balance sustaining high HFMB performance with minimizing the pressure drop across the membrane.

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