ORIGINAL ARTICLE

Macrokinetic determination of isopropanol removal using a downward flow biofilter

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Abstract

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Biofiltration is a process for eliminating contaminants in air using microorganisms immobilized on a surface of solid support media. This technique has been used successfully to control a number of air contaminants such as odors, Volatile Organic Compounds (VOCs), and Hazardous Air Pollutants (HAPs) due to its economic attraction.

Microorganisms obtained from local activated sludge (Huay-Kwang wastewater treatment plant (Bangkok, Thailand)) were selectively enriched and inoculated to the biofilter. The downward flow biofilter was chosen, due to the ease of water compensation at the dry zone, to operate continuously for more than 3 months under various concentrations of isopropanol alcohol (IPA) input at a constant filtered air flow rate of 3 L/min. The maximum IPA elimination capacity of 276 g/m^3-h was achieved at the IPA inlet of 342 g/m^3-h with acetone production rate of 56 g/m^3-h as the intermediate. It was also found that the acetone vapour was partly degraded by the acetone-utilizing microorganisms before leaving the bed.

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In order to understand the transport phenomena of biofiltration, it is necessary to consider the kinetic behavior of the bioreaction. Therefore, this paper introduces Wani's method of macrokinetic determination based on the simple Monod kinetic (Wani, Lau and Branion, 1999). In this study, the maximum reaction rate per unit volume (R_m) and the Monod constant (K_M) were found to be 0.12 g/m^3 -s and 2.72 g/m^3 respectively.

Key words : biofilter, isopropanol removal, macrokinetic determination, waste gas treatment

บทคัดย่อ

สถิดา ไกรลาศ ศุภลักษณ์ ทองทา และ วิษณุ มีอยู่

การหาค่าจลนพลศาสตร์แบบมหภาคของการกำจัดไอโซโพรพานอล ด้วยระบบกรองชีวภาพแบบไหลลง

ว.สงขลานครินทร์ วทท. 2547 26(ฉบับพิเศษ 1) : 55-64

ระบบกรองชีวภาพ เป็นกระบวนการหนึ่งในการกำจัดมลสารในอากาศ โดยใช้จุลินทรีย์ที่ถูกตรึงบนพื้นผิวของของแข็งตัวกลางในเบด เทคนิคนี้ได้รับการประยุกต์ใช้อย่างกว้างขวางในการบำบัดกลิ่น สารอินทรีย์ระเหยง่าย (VOCs) และมลพิษในอากาศ (HAPs) อันเนื่องมาจากเหตุผลทางเศรษฐศาสตร์ นั่นคือ มีต้นทุนในการติดตั้งและการปฏิบัติงานที่ต่ำเมื่อเทียบกับระบบแบบอื่น

จุลินทรีย์ที่ใช้ในระบบได้มาจากโรงงานบำบัดน้ำเสียห้วยขวาง กรุงเทพฯ แล้วนำมาคัดแยกด้วยวิธีทางธรรมชาติ และนำไปตรึงในระบบ การไหลของอากาศสำหรับระบบกรองชีวภาพนี้เป็นแบบไหลลงเพราะสามารถชดเชยน้ำที่ระเหยไปจากระบบอันเนื่องมาจากการปฏิบัติงานได้ง่าย การปฏิบัติงานกระทำที่ความเข้มข้นไอโซโพรพานอล (IPA) ต่าง ๆ กัน ที่อัตราการไหลคงที่ 3 ลิตร/นาที เป็นเวลาต่อเนื่องกัน 3 เดือน ค่าประสิทธิภาพการกำจัด IPA สูงสุดเป็น 276 กรัม/ลบ.เมตร ชั่วโมง เมื่อภาระบรรทุกมลสาร (IPA) ขาเข้าเป็น 342 กรัม/ลบ.เมตร ชั่วโมง และพบอัตราการเกิดอะซิโตนซึ่งเป็นสารตัวกลางในการย่อยสลาย IPA ของจุลินทรีย์มีค่าเป็น 56 กรัม/ลบ.เมตร ชั่วโมง และยังพบอีกด้วยว่า อะซิโตนถูกย่อยสลายไปบางส่วนโดยจุลินทรีย์ในเบดก่อนที่จะถูกปล่อยออกจากระบบ

เพื่อความเข้าใจในพฤติกรรมของระบบกรองชีวภาพ จึงมีความจำเป็นที่จะต้องทราบเกี่ยวกับจลนพลศาสตร์ของจุลินทรีย์ในระบบ ดังนั้น บทความนี้ มีวัตถุประสงค์จะนำวิธีการหาค่าจลนพลศาสตร์ของ Monod อย่างง่ายแบบมหภาคโดยวิธีของ Wani มาประยุกต์ใช้กับระบบการบำบัด IPA (Wani, Lau and Branion, 1999) ในการศึกษานี้ ค่าอัตราการเกิดปฏิกิริยาสูงสุดต่อหน่วยปริมาตร (R_m) พบว่ามีค่าเป็น 0.12 กรัม/ลบ.เมตร วินาที และค่าคงที่ของ Monod (K_M) เป็น 2.72 กรัม/ลบ.เมตร วินาที

ศูนย์วิจัยวัสดุประยุกต์และสิ่งแวดล้อม มหาวิทยาลัยเทคโนโลยีมหานคร ถนนเชื่อมสัมพันธ์ หนองจอก กรุงเทพฯ 10530

Biofiltration is a low-cost and highly effective air pollution control technology that can significantly decrease capital cost and long-term operating costs compared to conventional technologies such as carbon filtration or oxidation. Biofiltration has been used successfully to control a number of air contaminants such as odors, Volatile Organic Compounds (VOCs), and Hazardous Air Pollutants (HAPs) stemming from a wide range of industrial and public sector sources. Biofiltration removes air pollutants by passing the contaminated air through a damp, porous medium which supports microorganisms inoculated from activated sludge. The contaminants are transferred

from the air phase to the biofilm, then biodegraded within the biofilm. Microorganisms in the biofilter convert the air pollutants into harmless by-products that are primarily carbon dioxide and water. Michaelis-Menten kinetics (Monod kinetics) can be generally used to correlate the reaction rate.

Isopropyl alcohol (IPA) is a volatile organic compound with offensive odour and can be well degraded by microorganisms at low concentrations. If inhaled directly, it causes headache, cough and hard breathing. It also results in a rash on the skin when contacted directly. IPA threshold limit in vapour phase taken into the body, is 400 ppm or 983 mg/m^3 under 8 hours per day or 40 hours per week (Material Safety Data Sheet, 1991). Recently, highly solvent-tolerant bacterial consortia have been successfully used in biofiltration at high concentration of IPA vapour up to 10,500 ppm (Bustard, *et al*. 2001). In addition, Bustard's work (Bustard, *et al*. 2001) on IPA degradation reported that acetone was generated as an intermediate only at the start-up and acclimation periods, and none was observed at the steady-state period.

In order to understand the transport phenomena of biofiltration, the kinetic behavior of the bioreaction has to be obtained. The kinetic constants of the bioreaction can be determined either microkinetically or macrokinetically. For microkinetic determination, the microorganisms are isolated from the biofilter media and inoculated into shake flasks containing a nutrient solution with the contaminant solution as the sole carbon source. The concentration of contaminant is continuously measured as a function of time to obtain the kinetic constants (Ottengraf, *et al*. 1983). However, the microkinetic determination in biofiltration system is still controversial because the kinetic behaviors of biofiltration systems and suspended cell systems may be different (Wani, *et al*. 1999). Wani's method of macrokinetic determination based on the simple Monod kinetic has then become an alternative approach to obtain the kinetic parameters (Wani, *et al*. 1999). This method was successfully applied in the methanol removal system using biofiltration (Krailas, *et al*. 2002). Therefore, the macrokinetic approach using Wani's method was adopted in this paper in order to obtain the kinetic parameters of IPA biodegradation.

Material and Methods

Microorganisms and culture medium

The microorganisms obtained from Huay-Kwang wastewater treatment plant in Bangkok Thailand were continuously enriched in a 100-mL batch system with IPA being a main carbon source. Nutrient solution consisted of (g/L): Na_2HPO_3 4.0; KH_2PO_4 1.5; NH_4Cl 1.0; $MgSO_4$ 0.2; $CaCl_2$ 0.1; and $FeNH_4$-citrate 0.005; in water (Oh, 1993). 500-mL of nutrient solution was added into the batch reactors once a week. After 1 month with IPA as the sole carbon source, only mixed culture of IPA utilizing organisms were expected to survive. Visual observation was carried out by microscope every week.

Packing materials

The packing materials consisted of a mixture (1:1 v/v) of compost and pyrolised charcoal of 2-cm diameter and 450 mg iodine no.400 /g charcoal which was obtained from Pyrolysis Pilot Plant at Mahanakorn University of Technology. The charcoal was used as the additional support medium for microorganisms and to provide the bed porosity. 4.7×10^6 CFU/g dry solid of IPA utilizing microorganisms were immobilized onto the surface of charcoal before blending with the compost. The mixture of filter materials was then loosely packed into the biofilter column.

Experimental setup

Figure 1 shows the schematic diagram of a downward flow biofilter used in this study. The biofilter column was made of acrylic with 10 cm inside diameter and packed with prepared filter materials to a height of 60 cm on a bed support made of plastic screen. The solid media were taken from top and middle of solid sampling ports.

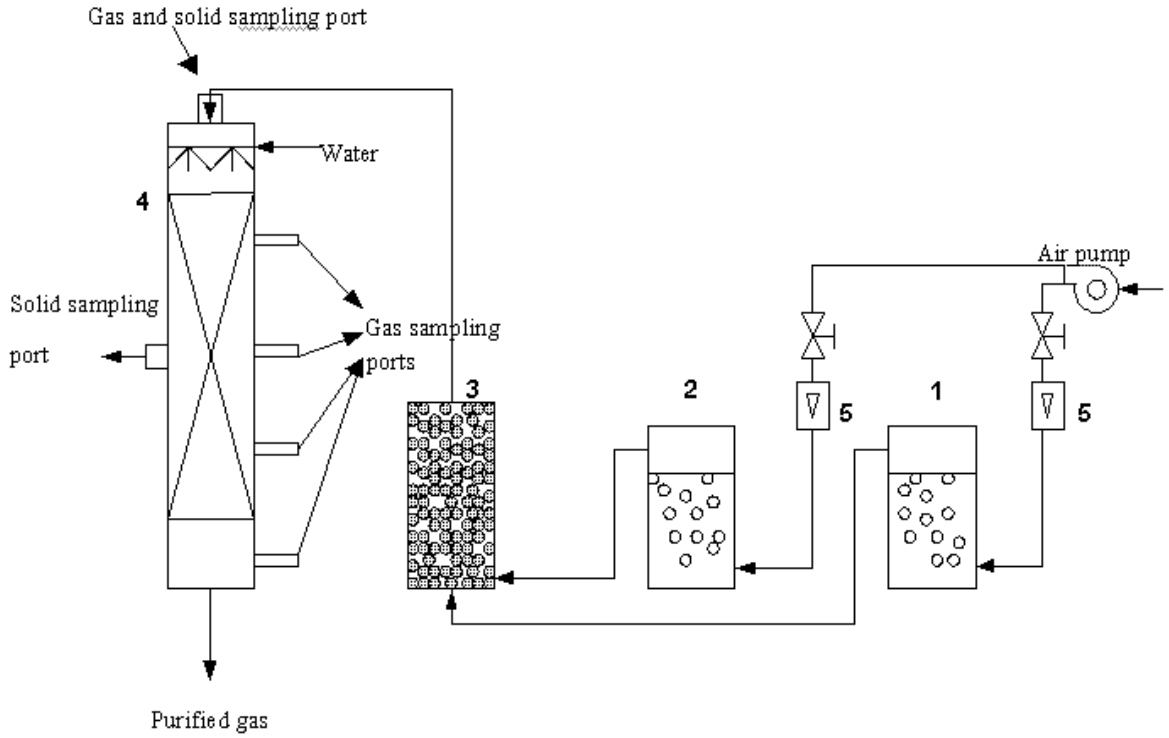

Figure 1. Schematic diagram of the downward flow lab scale biofilter in a temperature controlled room (30±2 °C)
(1) Water reservoir (2) Solvent (IPA) reservoir (3) Mixing chamber (4) Biofilter column (5) Rotameters

There were five gas sampling ports along the columns. Humidified air and IPA vapour were mixed in a mixing chamber and then passed through the biofilter bed. IPA concentration in the feed stream was varied by adjusting the flow rate of air stream passing through the water and IPA solution while the total flow rate was kept constant. The biofilter was operated continuously for three months at the total air flow rate of 3 L/min with 6 step changes of IPA concentration, i.e., 1.10, 2.30, 4.52, 5.35, 6.23, and 7.98 g/m^3 with A, B, C, D, E, and F representing the operating conditions, respectively.

Analytical methods

IPA and acetone concentrations were determined by a Shimadzu GC-8A Gas Chromatograph equipped with a 10% AT 1000 on chromosorb WAW 80-100 (a length of 2 m). Flow rate of N_2 carrier gas was 30 mL/min, the temperature of the Flame Ionization Detection (FID) system and oven were 120°C and 80°C, respectively. One mL of gas sample was withdrawn from the biofilter with a syringe (1 mL gas tight SGE syringe) at specified time intervals (3-5 minutes, approximately), and injected into the gas chromatograph in triplicate for analysis.

Total bacterial count (TBC) was performed when the system reached equilibrium, which can be noticed by monitoring the almost unchanged IPA concentration at output. Typically, 1 g of filter material was taken from the entrance and middle of the biofilter. Then, 10 cm^3 distilled water was added to each filter sample and centrifuged at 500 rpm for 2 min to gently separate the cultures from the sample. The cultures were inoculated into agar medium in triplicate and allowed to grow for 2-3 days in the incubator (30°C) before

counting the colonies. These results have also been reported elsewhere (Krailas, *et al*. 2002).

Results and Discussion

IPA biodegradation

The experiments were carried out at a constant flow rate with a 6 step increment. Each operating condition was maintained until the system reached its equilibrium. Figure 2 shows IPA degradation at different concentration. It was found that the biofilter was able to degrade IPA efficiently even when a high concentration of IPA was applied (7.98 g/m^3, Run F). A small amount of acetone was detected as an intermediate and found to increase proportionally with increasing concentration of IPA. The experiments were also carried out to investigate the fate of IPA and acetone along the bed at steady state conditions (Figures 3A-3F). In the case of low IPA concentration applied (Run A, Figure 3A), most of IPA was eliminated at 30 cm bed height while acetone rapidly increased to its maximum at 15 cm bed height before dropping to 0.01 g/m^3 at the outlet. This can be explained by the bacteria being able to utilize both IPA and acetone, resulting in innocuous final products like CO_2 and new cells as typically found in biological reactions (Ottengraf, *et al*. 1983). When IPA at high inlet concentrations was introduced to the bed from Runs B to F (Figures 3B-3F), acetone was found to increase in direct proportion to IPA input while IPA was removed continuously along the biofilter bed. It can be noted that there was no inhibitory effect due to the presence of acetone in the system since an acceptable IPA elimination capacity of the biofilter was still achieved, which can be seen in Figure 4 where the IPA elimination capacity did not exhibit a drop. In addition, the increases of acetone production rate observed in Runs B to F

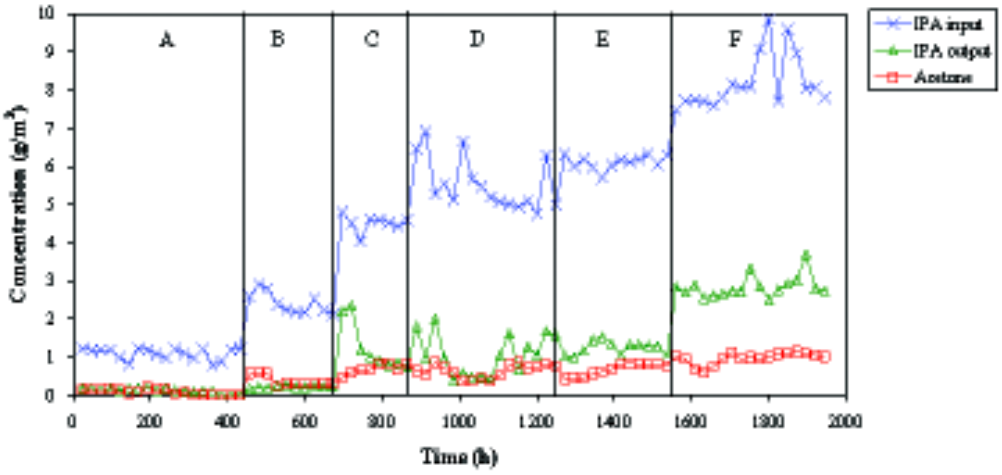

Figure 2. IPA degradation in the downward flow biofilter versus time.

Run	Flow rate (L/min)	Initial IPA Concentration (g/m^3)
A	3	1.10
B	3	2.30
C	3	4.52
D	3	5.35
E	3	6.23
F	3	7.98

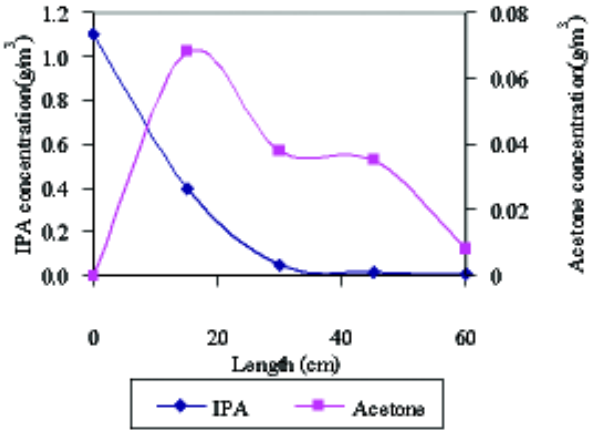

Figure 3A. IPA biodegradation and acetone generation at steady state for IPA input concentration of 1.10 g/m³ (Run A) at the air flow rate of 3 L/min.

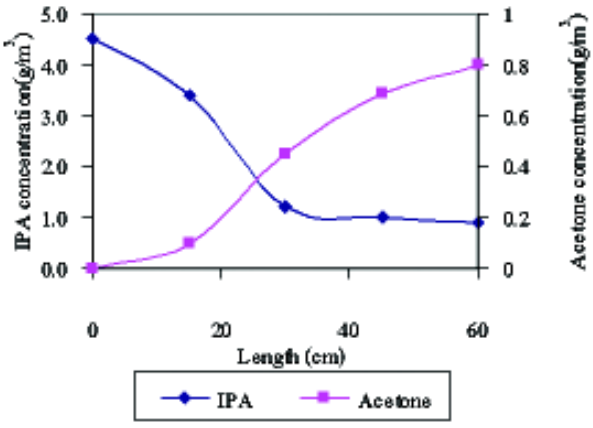

Figure 3C. IPA biodegradation and acetone generation at steady state for IPA input concentration of 4.52 g/m³ at the air flow rate of 3 L/min.

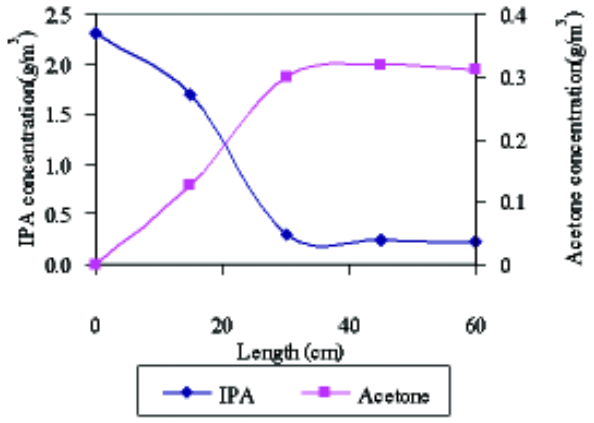

Figure 3B. IPA biodegradation and acetone generation at steady state for IPA input concentration of 2.30 g/m³ at the air flow rate of 3 L/min.

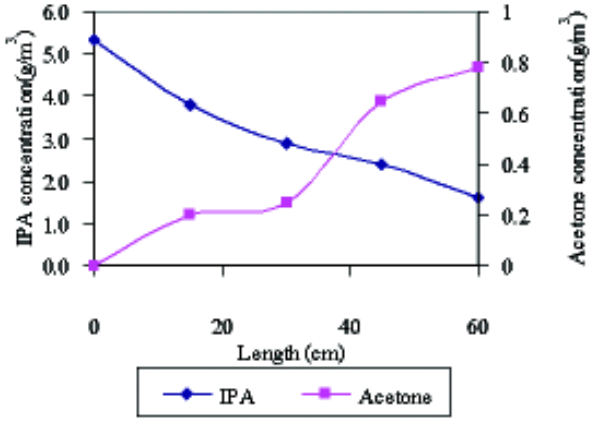

Figure 3D. IPA biodegradation and acetone generation at steady state for IPA input concentration of 5.35 g/m³ at the air flow rate of 3 L/min.

are due to the fact that a high inlet mass loading rate of IPA resulted in a better conversion rate to produce acetone. A certain amount of acetone may be still biodegraded by the bacteria simultaneously, but not distinctively seen compared to its generation rate. The concentrations of acetone generated of 0.31 g/m^3 (Run B), 0.78 g/m^3 (Run C), 0.78 g/m^3 (Run D), 0.80 g/m^3 (Run E), and 1.06 g/m^3 (Run F), respectively, were determined at the vent gas in this study.

Macrokinetic determination

Figure 4 illustrates the IPA elimination capacity (EC) of biofilter at various inlet IPA mass loadings (IL). It can be seen that there was no inhibitory effect due to the presence of acetone in the system. The maximum IPA elimination capacity of 276 g/m^3 h was achieved at the inlet

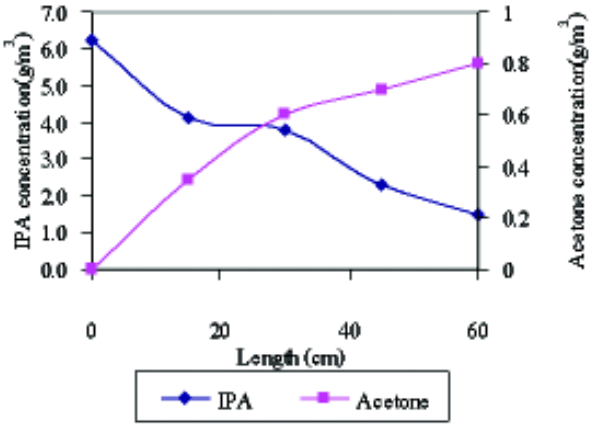

Figure 3E. IPA biodegradation and acetone generation at steady state for IPA input concentration of 6.23 g/m^3 at the air flow rate of 3 L/min.

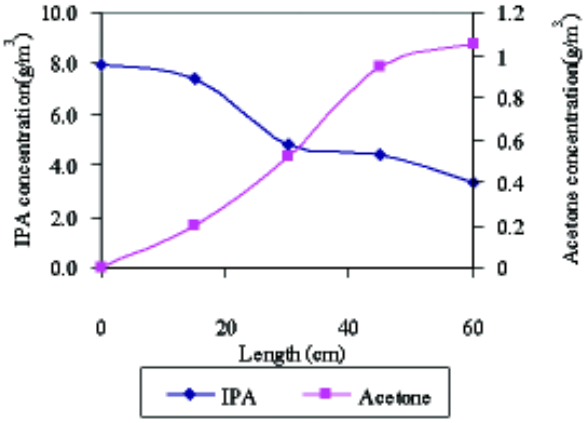

Figure 3F. IPA biodegradation and acetone generation at steady state for IPA input concentration of 7.98 g/m^3 at the air flow rate of 3 L/min.

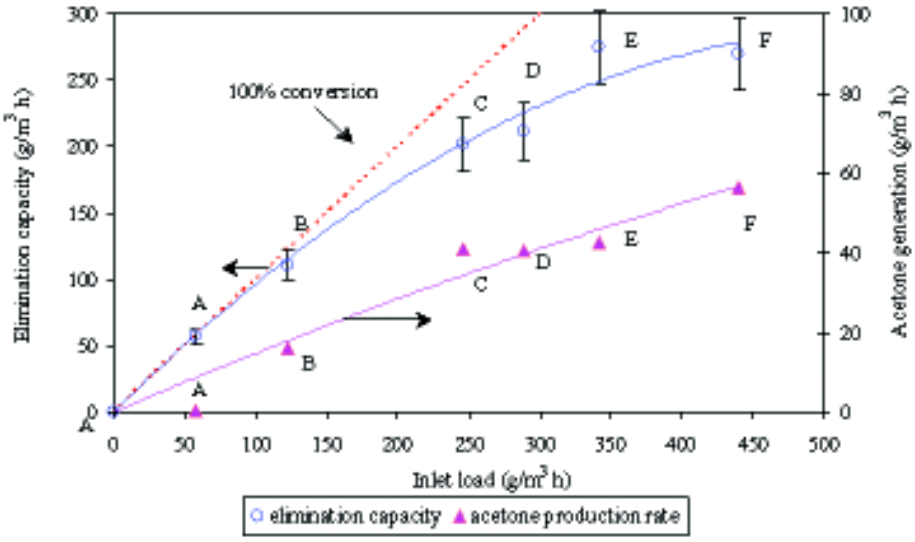

Figure 4. Elimination capacity of IPA versus inlet mass loading at a constant volumetric flow rate of 3 L/min (with ±10% error).

IPA elimination capacity $= \frac{Q}{V}(C_i - C_o)$

Acetone production rate $= \frac{QC_{po}}{V}$ **where** C_{po} **= acetone concentration at output**

IPA inlet mass loading rate = $= \frac{QC_i}{V}$

loading of 342 g/m^3 h. It is also observed that the elimination capacity was an increasing function of inlet load and tended to stabilize asymptotically for an inlet load beyond 246 g/ m^3 h.

At inlet mass loading rate below 122 g/ m^3 h (Runs A-B), the system was in a diffusion-controlled regime (Bibeau *et al*., 1997). Therefore, the bioreaction took place rapidly and the carbon source from IPA was completely utilized at outer surface of biofilm, resulting in insufficient supply of carbon source at the internal layer of biofilm (adjacent to packing media). The biofilm at this stage was then not fully active since the bioreaction occurred only at the outer layer of biofilm.

In contrast, at higher inlet mass loadings (Runs C-F), the process was found to be independent of the inlet mass loading because the system was controlled by bioreaction rate. In this phenomenon, IPA was slowly utilized and able to reach the inner layer of biofilm. The carbon nutrient was then enough to assist bacteria growth thoroughly. Consequently, the biofilm appeared to be fully active at this stage which means the bioreaction took place at the entire biofilm thickness. Acetone production rate was found to increase with a raise of inlet mass loading in Runs A-C, and then the plateau could be expected as it followed the IPA elimination trend. In addition, the acetone production rate was found to reach maximum at 56 g/ m^3 h in Run F.

In this study, the macrokinetics of the system can be expressed by a Michaelis-Menten type relationship by assuming that oxygen limitation is not present in the system since the system is aerobic, and the system is in the reaction-controlled regime (the bioreaction took place thoroughly in the entire biofilm). At steady state, the growth rate of microorganisms is balanced by their own decay rate, resulting in the biological equilibrium of the system. Hence kinetic constants remain constant over the period of time considered. Runs C to F were used in estimating to assure the reaction-controlled regime, in accordance with the assumptions made by Wani, *et al*. (1999).

The kinetic constants can be determined using the plug flow model without dispersion at steady state (eqn (1)).

$$\frac{\partial C}{\partial t} = -u\frac{\partial C}{\partial h} + R_x \qquad (1)$$

where C is IPA concentration (g/m^3), u is the superficial velocity (m/s), t is the time interval (s), h is the distance in the bed (m), and R_x is the overall reaction rate defined as follows.

$$R_x = \frac{R_m C}{K_m + C} \qquad (2)$$

where R_m is the maximum bioreaction rate per unit biofilter volume (g/m^{3}s) and K_M is the saturation (Michaelis-Menten) constant (g/m^3) in the gas phase.

At steady state, the accumulation term ($\partial C / \partial t$) equals zero. Integrating under the given conditions C=C_i at h=0 and C=C_o at h=L, where L is the biofilter length (m), the following is obtained.

$$\frac{V/Q}{C_i - C_o} = \frac{K_M}{R_m}\frac{1}{C_{Ln}} + \frac{1}{R_m} \qquad (3)$$

where C_{Ln} is the log mean concentration [=(C_i-C_o)/ln(C_i/C_o)], V is the biofilter volume (m^3), and Q is the volumetric flow rate (m^3/s). R_m and K_M for the gas phase can be obtained by plotting [(V/Q)/(C_i-C_o)] against (1/C_{Ln}). This macrokinetic determination has also been applied in other works (Chung,*et al*.1997, Cho *et al*. 1991, Hirai, *et al*.1990, Chung, *et al*.1997).

It was found that one simple rate expression which can be suitable for explaining the IPA degradation was the simple Monod kinetics with a correlation coefficient of 0.9691 (see Figure 5). R_m and K_M for the gas phase obtained from Figure 5 and eqn (3) are 0.12 g/m^{3}s and 2.72 g/m^3, respectively.

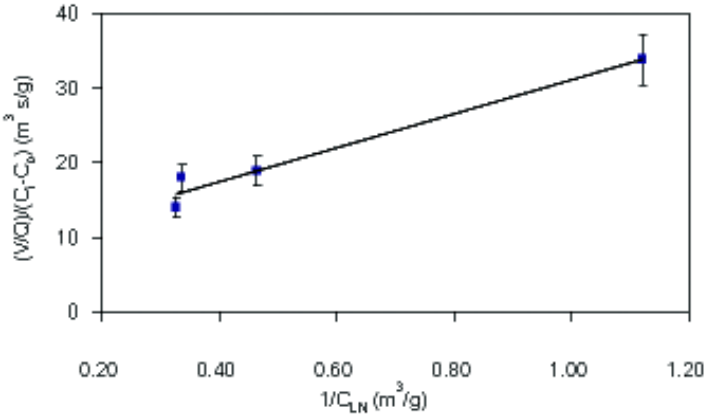

Figure 5. Macrokinetic determination from steady state data on IPA biodegradation (with ±10% error).

Conclusions

The purpose of this work was to use Wani's assumption to macrokinectically determine the kinetic parameters of IPA biodegradation since this method was successfully applied in the methanol removal system using biofiltration (Krailas, *et al*. 2002). The simple Monod kinetics appeared to fit best with the experimental results of IPA removal system. The maximum reaction rate per unit volume (R_m) and the Monod constant (K_M) were found to be 0.12 g/m^3 -s and 2.72 g/m^3 respectively.

A good performance of the biofilter in this study was also observed in terms of the IPA elimination capacity. The maximum IPA elimination capacity (EC) of 276 g/m^3 h and acetone production rate of 56 g/m^3 h were reported at the inlet mass loading of 342 g/m^3 h. In addition, the IPA biodegradation was found to yield acetone as an intermediate. The maximum level of acetone generation in this study was 1.06 g/m^3 at the inlet IPA concentration of 7.98 g/m^3. The amount of acetone generated was found to increase with the increase of inlet IPA concentration.

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