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Biofiltration of isopropyl alcohol by a trickle-bed air biofilter

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Abstract

The performance of trickle-bed air biofilter (TBAB) for the removal of isopropyl alcohol (IPA) was evaluated in concentrations varying from 100 to 500 ppmv and at empty-bed residence time (EBRT) varying from 20 to 90 s. Nearly complete IPA removal could be achieved for influent carbon loading between 6 and 88 g/m^3·h. The TBAB appears efficient for controlling IPA emission under low-to-high carbon loading conditions. Carbon recoveries of 95-99% were achieved demonstrating the accuracy of results. Applicable operating conditions of TBAB for controlling IPA emission were suggested.

Introduction

Isopropyl alcohol (IPA) is a commonly used industrial chemical. In addition to its use as a solvent, it is frequently encountered in the Semi-conductor and Opto-electronic industries that are the major industries in Taiwan. Due to the lack of proper air pollution control device, many IPA vapors are released into the atmosphere during manufacturing process every year. IPA vapors are irritative and carcinogenic substances (Purdom 1980). Loss of these substances to ambient air may lead to an adverse effect on the air quality and thus endanger public health and welfare.

More stringent requirements for the removal of volatile organic compounds (VOCs) from waste gases in recent years necessitate the development of innovative, cost-effective treatment alternatives. Traditional VOC control technologies such as carbon adsorption, liquid scrubbing, condensation, thermal incineration, and catalytic incineration have been commonly used to remove VOC vapors from waste gases. However, these VOC control technologies may suffer from high operating costs and secondary waste stream issues (Corsi & Seed 1995).

The trickle-bed air biofilter (TBAB) is a type of biofilter process that employs synthetic, inorganic media and receives liquid nutrients through a spray nozzle system on the top of the TBAB. Due to better control of pressure drop across the bed, pH and nutrient feed, TBABs facilitate more consistent operation than do natural media biofilters. Furthermore, they do not suffer from the effects of aging as do natural media (Sorial et al. 1997a).

Smith et al. (1996) evaluated the performance of two similar TBABs with high toluene loading rates between 2.27 and 4.55 kg COD/m^3·d. They found that a sustained toluene removal efficiency of over 99% could be achieved and the effectiveness could be maintained over 200 days. Sorial et al. (1997b) studied the performance of a peat mixture biofilter for the removal of toluene in the concentration range of 20–2000 ppmv. The toluene removal efficiency was 99% for a loading of 0.45 kg COD/m^3·d and decreased to 57% as the loading was increased to 1.57 kg COD/m^3·d. Rihn et al. (1997) evaluated diethyl ether as a substrate for two TBABs and found that more than 99% removal efficiency could be obtained with a loading of 3.6 kg COD/m^3·d. Lu et al. (2000) evaluated the TBAB performance for treating acrylonitrile (AN) vapor. It was found that greater than 95% AN removal efficiency was achieved with influent AN loadings below 490 g/m^3·h, and the effectiveness could be

maintained over 24 weeks of laboratory operation. Lu et al. (2001) studied the TBAB performance for influent gases with styrene (SR) concentrations varying from 150 to 600 ppmv and at empty-bed residence time (EBRT) varying from 1.5 to 6 min. More than 90% removal efficiency could be achieved for influent SR loadings below 32 g/m^3·h.

The TBAB has been proven to be efficient for the removal of various individual VOCs by the foregoing researchers. However, studies on the biofiltration of gases containing IPA vapors are still unavailable in the literature. This research aims at evaluating the TBAB performance in the treatment of air contaminated by IPA in concentrations varying from 100 to 500 ppmv and at EBRT varying from 20 to 90 s. Applicable operating conditions of TBAB for controlling IPA emission are suggested according to experimental results obtained herein.

Materials and methods

Experimental set-up

The experimental set-up of the TBAB used for IPA removal is shown in Figure 1. It was made of stainless steel and had length of 100 cm and internal diameter of 10.85 cm. A 10-cm headspace was designed for the IPA inlet and for housing a nutrient spray nozzle, while a 10-cm bottom space was designed for the outlet of treated air and leachate. The TBAB was filled with a 7.86 L packing material consisting of coal particles with density of 0.318 kg/L, equivalent-volume diameter of 2 cm, average pore size of 23.25 μm, and surface area of 6.21 m^2/g (BET analyzer, ASAP 2000, Micromeritics, USA). The choice of coal particles as packing material was due to the following advantages: (1) cheap, large available surface area for biofilm accumulation, (2) proven capacity to maintain moisture content, (3) without aseptic conditions. The void fraction before biofilm attachment was 44% of the packed volume. In order to simulate the real-scale uncontrolled biofilter, the temperature inside TBAB was not controlled through the study.

Compressed air was passed first through a filtration device (Model LD-05A, Taiwan) to remove moisture, oil and particulate matter. After purification, the major air stream was mixed with a 2.4 mL/min nutrient solution and delivered into the headspace by a nozzle spray system. The minor air stream was passed through two glass bottles containing pure IPA solution (JT Baker, Actual Analysis, USA, 99.9% purity) and air, respectively, to produce IPA vapor. The IPA vapor was then mixed with the major air stream in the headspace and passed into the bed with the flows directed downwards. The major air stream rate was controlled by a rotameter, while the influent IPA concentration was controlled by regulating the minor air stream flow rate using mass flow controllers (MKS, Model 247C, Andover, MA, USA). The variations of influent IPA concentration were within 10%. The nutrient solution contained inorganic salts and $NaHCO_3$ as a buffer. The carbon mass ratio of influent IPA to nitrogen, phosphorus, sulfur and iron of nutrient solution was equal to 100:10:1:1:0.5. The composition of nutrient solution for a carbon-loading rate of 6 g-C/m^3·h (Run 1) are listed in Table 1, while those of other runs were proportionally increased according to the carbon-loading of influent gas to that of 6 g-C/m^3·h. In order to test the sorption capacity of IPA on coal particles, adsorption and desorption experiments were carried out before the TBAB start-up. For adsorption experiment, the reactor was fed at 7.86 L/min (EBRT = 60 s) using an air stream containing 500 ppmv IPA. The influent and effluent IPA concentrations were monitored every 4 hours. For desorption experiment, the TBAB was operated at 15.72 L/min (EBRT = 30 s) pure air. The effluent IPA concentration was monitored every hour.

Table 1. Composition of the nutrient feed for a carbon loading of 6 g-C/m^3· h (Run 1)

Constituents	Concentration
KNO_3 (g/L)	0.17
$Na_2HPO_4{\cdot}12H_2O$ (g/L)	0.01
$(NH_4)_2SO_4$ (g/L)	0.01
KH_2PO_4 (g/L)	0.01
$FeSO_4{\cdot}7H_2O$(mg/L)	3.38
$CaCl_2{\cdot}2H_2O$(mg/L)	3.00
$MgSO_4{\cdot}7H_2O$(mg/L)	2.00
$Na_2MoO_4{\cdot}2H_2O$(mg/L)	1.00
$MnSO_4{\cdot}H_2O$ (mg/L)	0.88
$NaHCO_3$ (g/L)	1.50

The TBAB was seeded with activated sludge having suspended solid (SS) of 42.07 g/L and volatile suspended solid (VSS) of 7.78 g/L, which was obtained from the sludge thickener of a wastewater treatment plant in Hsinchu Science-Based Industry Park (Hsinchu, Taiwan). Suspended solids were allowed

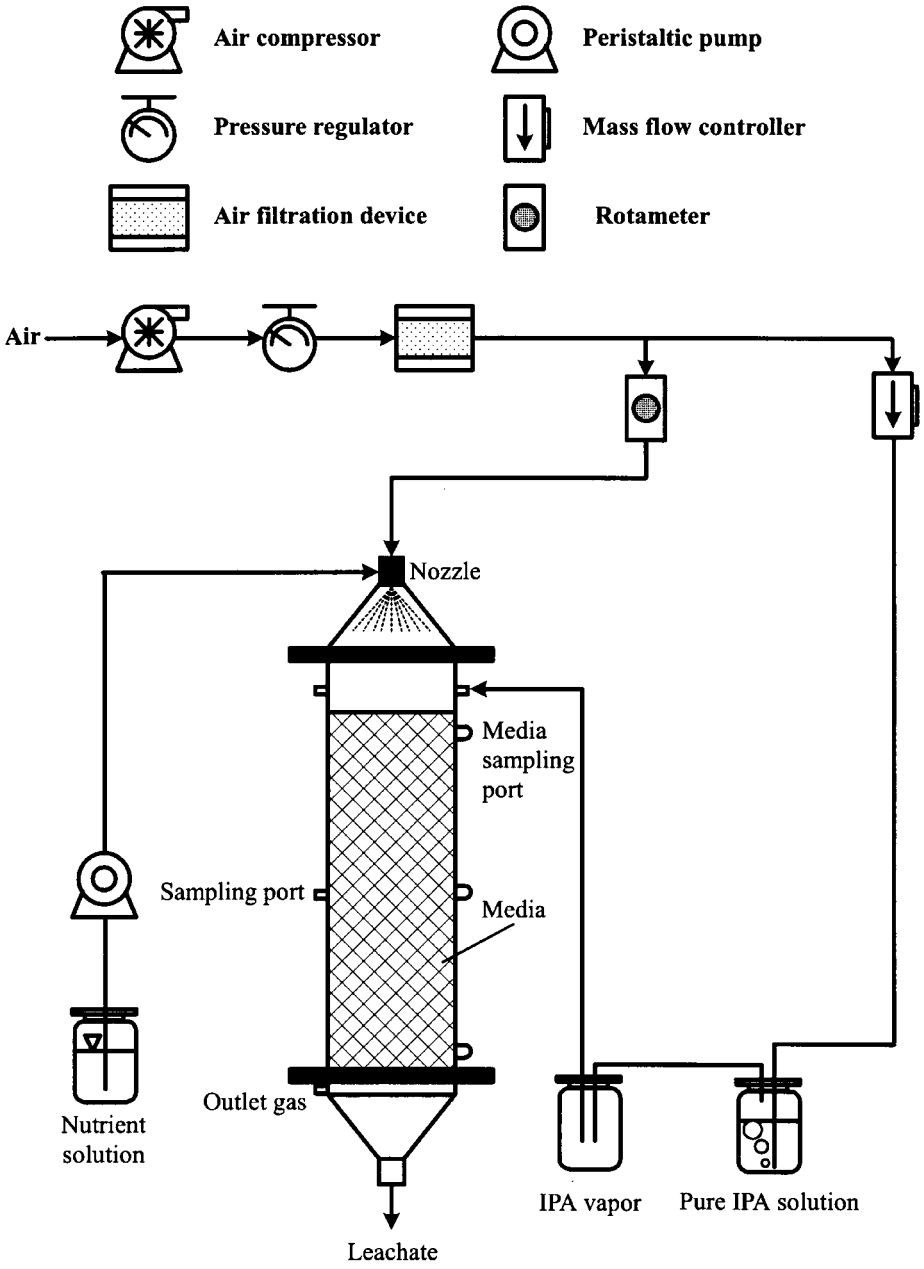

Figure 1. Schematic diagram of the trickle-bed air biofilter for the removal of isopropyl alcohol.

to settle for 4 h and the supernatant was discarded to obtain concentrated sludge. The seeding step consisted of mixing 500 L of concentrated sludge with coal particles and 125 g $CaCO_3$. Addition of $CaCO_3$ was used to prevent the acidification inside TBAB. Coal particles with biological attachment were placed into TBAB for about 3 h. After microbial seed, the TBAB was fed with a 2.4 mL/min nutrient solution and operated at a 5.24 L/min (EBRT = 90 s) air stream containing 100 ppmv IPA.

Analytical methods

IPA concentrations in the air stream were measured using a gas chromatograph (GC, China Chromatography 8900 Series, Taiwan) equipped with a flame ionization detector (FID). A 60-m SUPELCOWAX Fused Silica

capillary column (0.32 mm inside diameter, 1 μm film thickness) was used. Sampling ports were located at TBAB inlet and outlet, and middle height of TBAB. A 0.5 L effluent air sample was collected using 1.0 L Teflon bag (SKC Inc., PA, USA). Air samples (0.1 mL) were taken in this bag using a gas-tight syringe and injected into GC. The GC/FID was operated at injection temperature of 150 °C, detector temperature of 200 °C, and oven temperature of 70 °C.

The following parameters were determined according to Standard Methods (APHA 1995): soluble chemical oxygen demand (SCOD, 5220-D), and SS (2540-D) and VSS (2540-G). The pH value leachate was measured by a digital pH meter (SUNTEX SP-701, Taiwan). The CO_2 concentration in the air stream was determined by a CO_2 analyzer (TESTO Model 535, Germany). Pressure drop across the bed was measured using an oil-filled manometer (Dwyer Model 400, Michigan, USA).

A pair of tweezers was used to remove coal particles. Mass of the attached biofilm per unit volume of coal particle (X_a) was evaluated by drying coal particles before and after biofilm attachment at 80 °C for 24 h. The difference between the two measurements divided by the volume of the coal particle was equal to X_a.

Carbon balance analysis

Carbon balance was performed after achievement of the pseudo-steady-state conditions in each run. It can be written as:

$$Q_a C_i = Q_a C_e + Q_a C_c + \alpha Q_1 C_b + \beta Q_1 C_l \quad (1)$$

where Q and C represent the flow rate and carbon concentration, respectively, $Q_a C_i$ and $Q_a C_e$ are the influent and effluent carbon mass rates associated to IPA, $Q_a C_c$ is the effluent carbon mass rate associated to CO_2 production; $\alpha Q_1 C_b$ is the biomass production rate equivalent to carbon mass utilization rate; and $\beta Q_1 C_l$ is the effluent carbon mass rate of leachate. Assuming that the net accumulation of attached biomass in the TBAB was negligible (i.e., biomass production rate = biofilm detachment rate), the C_b level can be estimated from VSS of leachate. Since the predominant microorganisms in the biofilter decomposing VOCs are heterotrophic bacteria and fungi (Swanson et al. 1997), $NaHCO_3$, which was added into nutrient solution as pH buffering compound, was not included in the carbon balance. A typical cellular composition for a heterogeneous microbial population can be represented as $C_5H_7NO_2$ (Gaudy and Gaudy 1980), therefore, the conversion factor of biomass to carbon concentration, α, can be assumed to be equal to 0.53 (60/113). The C_l level can be approximated from SCOD of leachate. The conversion factor of SCOD to total organic carbon (TOC), β, can be evaluated from the stoichiometry for IPA oxidation

$$C_3H_8O + 9/2\ O_2 \rightarrow 3\ CO_2 + 4\ H_2O \quad (2)$$

From Equation (2), the β for IPA is estimated as 0.25 (36/144). The carbon recovery, R, is defined as the percentage ratio of the sum on the right-hand side of Equation (1) to $Q_a C_i$.

Table 2. Operating conditions of continuous tests of IPA removal in TBAB

Run no.	C_i (ppmv)	Q_a (L/min)	EBRT (sec)	L (g/m^3·h)
1	100	5.24	90	6
2	100	7.86	60	9
3	200	5.24	90	12
4	200	7.86	60	18
5	100	15.72	30	18
6	500	5.24	90	29
7	200	15.72	30	35
8	500	7.86	60	44
9	200	23.58	20	53
10	500	15.72	30	88

Note: C_i, IPA concentrations in the inlet gas phase; Q_a, air flow rate; EBRT, empty-bed residence time; L, carbon loading referred to the biofilter volume.

Experimental plan

The operating conditions are summarized in Table 2. The tests were conducted by following the order of runs in Table 2. The influent IPA was selected so as to simulate various emissions from Semi-conductor and Opto-electronic industries. The EBRT was varied from 20 to 90 s to establish the optimum operating conditions and the influent carbon loading was in the range of 6 (Run 1) to 88 g-C/m^3·h (Run 10).

Results and discussion

Figure 2 shows the breakthrough curve for adsorption of IPA vapor on coal particles and the curve for desorption of IPA vapor from coal particles. The effluent

IPA concentration started to increase fast after 50-h operation and the influent IPA concentration could be reduced from 500 to below 80 ppmv by coal particles. After 64-h operation, the effluent IPA concentration reached steady values (approximately 95% of the influent IPA concentration). The operating capacity of coal particles to remove IPA was equal to 18.7 mg-IPA/g-coal. The desorption curve indicated that the IPA was slowly released from coal particles. After 22-h operation, the effluent IPA concentration was reduced from 300 to 15 ppmv. Based on these results, it could be concluded that the coal particles appear to be efficient for buffering the fluctuation in IPA concentration.

The temperature inside TBAB, the pH of leachate, and the influent and effluent IPA concentrations were monitored every 2 to 3 days. The temperature inside TBAB ranged from 17 to 26 °C while the pH of the leachate ranged from 7.5 to 8.5. Figure 3 shows the TBAB performance for IPA removal as a function of the operating time. Note that with the exception of Run 1 (including the period for starting TBAB) each run was operated for 1–2 weeks to reach the pseudo steady state. Pseudo steady state was presumed when the changes in the IPA removal efficiency were within 5% for three successive samples. With the exception of Run 2, the IPA removal efficiency in each run increased gradually, reached steady values, and then decreased rapidly after a sudden change of EBRT or influent IPA concentration. The operating time to reach pseudo steady state is shorter for the runs with a lower IPA feed or at a longer EBRT (a lower influent IPA loading). Nearly complete IPA removal could be attained at the end of each run. Hence, the TBAB appears to be very efficient for controlling IPA emission under low-to-high carbon loading conditions. Upon reaching a pseudo steady state, the TBAB was extensively sampled and analyzed, and the results are discussed in the following paragraphs.

The CO_2 concentrations of influent and effluent air streams were evaluated and listed in Table 3. The difference between the two concentrations was equal to concentration of CO_2 production. The theoretical concentrations of CO_2 production estimated from Equation (2) are also presented. The data shows that, at identical EBRTs, the CO_2 production has a linear relationship with the influent IPA concentration. This was because that the TBAB was operated under conditions of carbon source limitation. The concentrations of CO_2 production ranged from 206 to 1185 ppmv. The data also shows that fair agreement was obtained between the concentrations of measured and theoretical CO_2 production. The measured results were slightly lower than the theoretical results and the discrepancies were in the range of 1.4–6.5%. There are two possible reasons to explain the discrepancies. First, some of the IPA vapor was dissolved into the nutrient solution. Second, some of the IPA vapor was utilized and converted to microbial cell.

Figure 4 shows the values of X_a and P as a function of the operating time. As can be seen, the X_a level increased as the operating time increased. The X_a value of each run ranged from 2.02 to 42.75 mg/cm^3. The X_a increased as influent IPA concentration increased or EBRT decreased (an increase of influent IPA loading), likely because the biofilm growth was directly related to IPA elimination capacity. With a higher influent IPA loading, the IPA elimination capacity was higher. Therefore, more attached microorganisms were yielded under a higher influent IPA loading. The P value significantly increased as EBRT decreased. This was due to the fact that the P value has a linear relationship with the superficial gas flow rate (Crawford 1976). The P value slightly increased with the increase in influent IPA concentration. This can be attributed to the fact that more microorganisms were yielded under a higher IPA feed, which might have minimized the external porosity of coal particles and thus led to higher-pressure drops across the bed. With the exception of Runs 3, 8 and 10 similar trend was observed for the P level. This was because the EBRTs of these runs are longer than those of the previous runs. However, The P level did not drop at Run 6. This was because the carbon loading of Run 6 was increased from 17.65 to 29.41 g/m^3·h which leads to the increase of X_a from 15.32 to 20.76 mg/cm^3. The TBAB was operated about 120 days from Runs 1 to 10. During this period, the P value increased from 0.01 to 8.13 cm-H_2O, which had no influence on the performance of TBAB. Therefore, the TBAB was not backwashed in this study.

The leachate production rate was approximately 3 L per day. The SCOD of leachate can be mainly related to dissolved IPA vapor. The SCOD of leachate ranged from 126 to 289 mg/L. Figure 5 shows the results of SS and VSS in leachate and mass ratio of VSS to SS (η) as a function of the operating time. The VSS of leachate can be mainly related to sloughed biofilm. The SS and VSS levels in leachate ranged from 18.94 to 159.9 mg/L and 14.39 to 153.5 mg/L, respectively. As can be seen, the η value was in the range of 0.76–0.98 indicating that most of the SS in leachate came

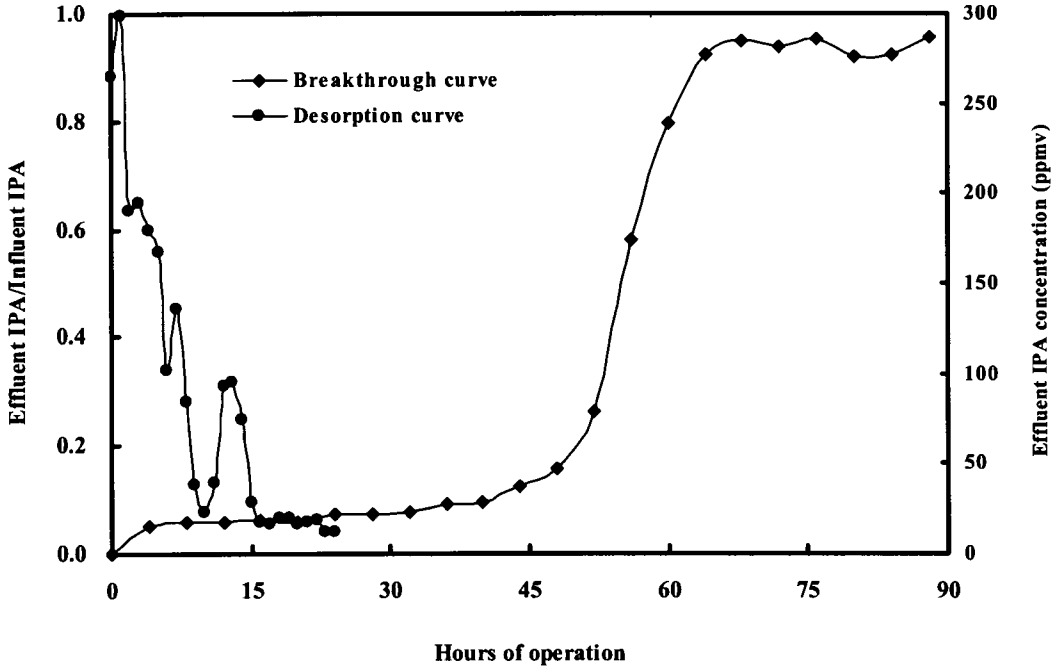

Figure 2. The breakthrough curve of isopropyl alcohol vapor on coal particles and the desorption curve of isopropyl alcohol vapor from coal particles.

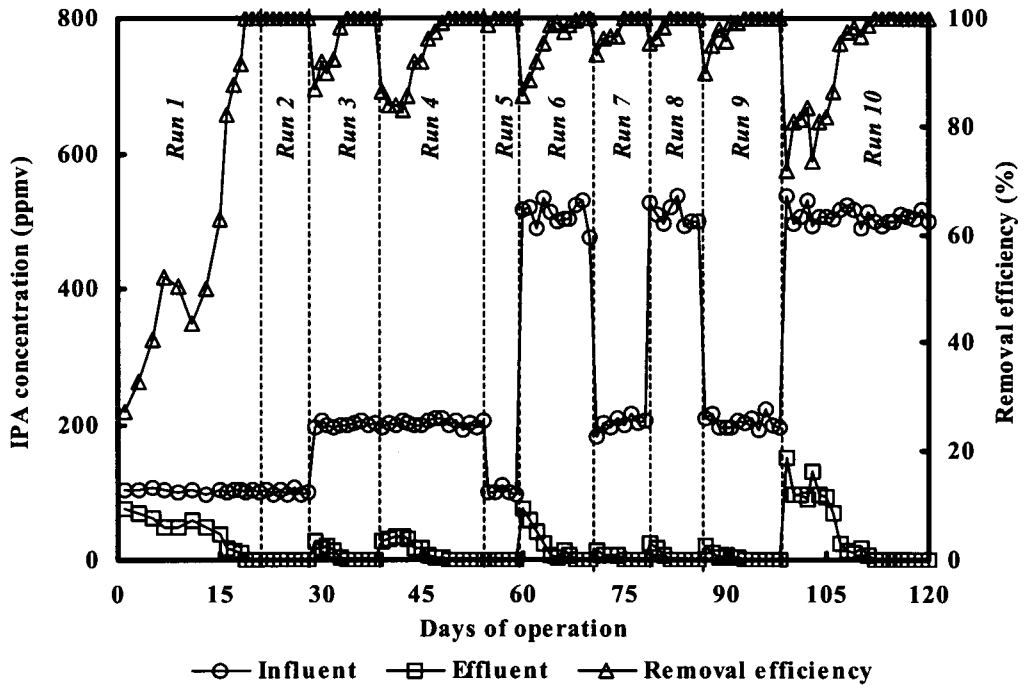

Figure 3. The performance of trickle-bed air biofilter for isopropyl alcohol removal.

Table 3. Carbon dioxide results of continuous tests of IPA removal in TBAB

Run no.	Influent CO_2 (ppmv)	Effluent CO_2 (ppmv)	Measured CO_2 Production (ppmv)	Theoretical CO_2 Production (ppmv)	Difference (%)
1	425	634	209	220.65	5.28
2	407	618	211	220.97	4.51
3	409	842	433	445.57	2.82
4	403	834	431	452.69	4.79
5	401	601	200	213.95	6.52
6	408	1418	1010	1057.15	4.46
7	397	839	442	451.67	2.14
8	377	1464	1087	1106.84	1.79
9	408	826	418	434.90	3.89
10	405	1498	1093	1108.51	1.40

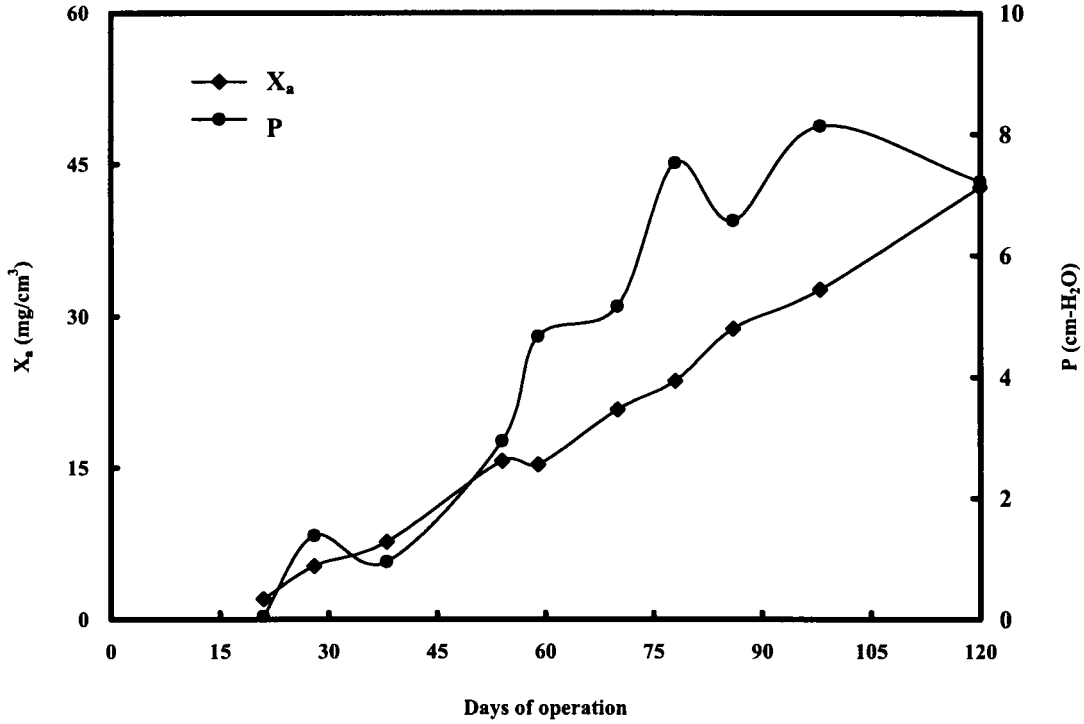

Figure 4. The results of mass of the attached biofilm per unit volume of coal particles (X_a) and pressure drop across the bed (P).

from sloughed biofilm. The SS and VSS levels were relatively higher at shorter EBRT. This can be attributed to higher fluid shear stress caused by a higher superficial gas/liquid flow rate.

The microbial yield coefficients (Y) of each run are listed in Table 4. The Y value was much less than unity, indicating that the fraction of IPA associated to cells production was usually very small. The Y values for Runs 8–10 ranged from 0.005 to 0.009 g-VSS/g-COD, smaller than those of other runs. This may be attributed to an inhibitory effect on the growth of microbial cell at high IPA loadings. The influent COD loading was in the range of 0.56–8.47 kg COD/m^3·d. The resulting Y value ranged from 0.005 to 0.041 g-VSS/g-COD with an average value of 0.014 g-VSS/g-COD, which was smaller than the

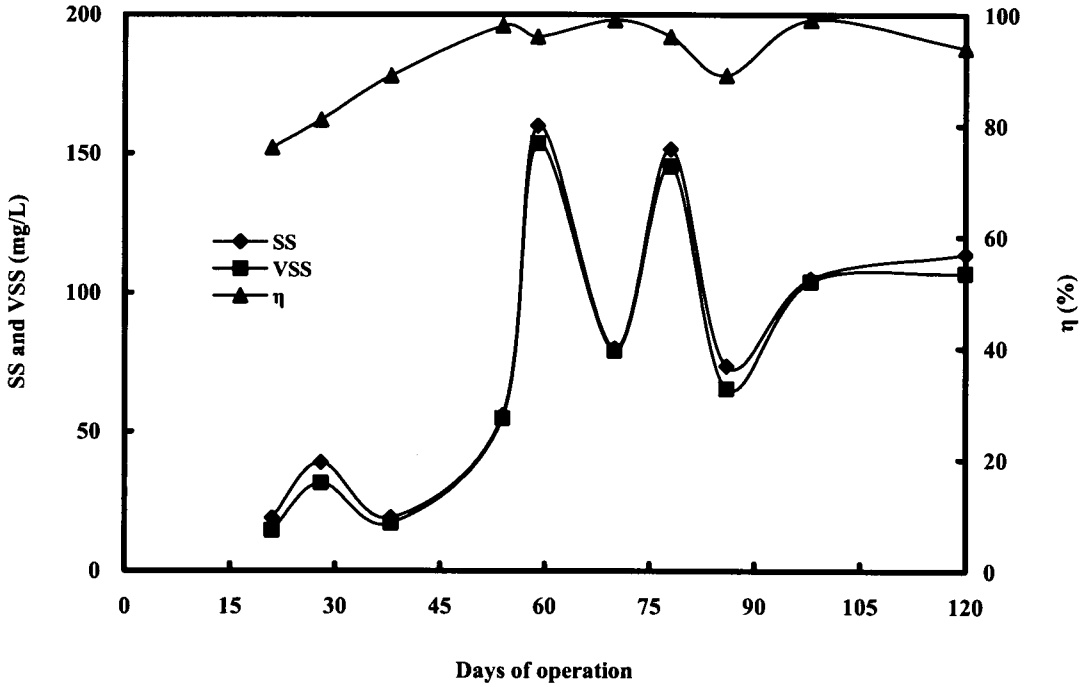

Figure 5. The results of suspended solid (SS) and volatile suspended solid (VSS) in leachate and mass ratio of VSS to SS.

Table 4. The yield coefficients of each run

Run no.	SRR (g-COD/day)	BPR (g-VSS/day)	*Y* (g-VSS/g-COD)
1	4.476	0.050	0.011
2	6.725	0.109	0.016
3	9.041	0.059	0.007
4	13.778	0.189	0.014
5	13.025	0.530	0.041
6	21.451	0.273	0.013
7	27.494	0.502	0.018
8	33.686	0.226	0.007
9	39.710	0.359	0.009
10	67.394	0.370	0.005

Note: SRR, substrate removal rate; BPR, biomass production rate; *Y*, microbial yield coefficient.

typical value of 0.078 g-VSS/g-COD for the removal of benzene, toluene, ethyl benzene and xylene (BTEX) mixtures by TBAB systems in the COD loading range of 0.45–6.2 kg COD/m^3·d (Sorial et al. 1997a). This indicated that the fraction of IPA that is channeled into new cell during growth is smaller than that of BTEX.

The results of the carbon balance listed in Table 5 show that carbon recovery (R) was particularly high (95-99%), thus indicating the accuracy of test results. Most of the effluent carbon was from CO_2 production (above 99%). This implies that the aerobic digestion of IPA vapor was almost complete in TBAB. The carbon mass rate of the liquid effluent only ranged from 0.06 to 0.14 mg-C/min, which was approximately 2–3 orders of magnitude less than that of the CO_2 effluent (12.19–191.26 mg-C/min). Therefore, it could be concluded that the dissolved IPA and its derivatives in leachate were not significant in TBAB.

Applicable operating conditions of TBAB for IPA removal are suggested in Table 6. Removal efficiencies of 90-99% could be achieved under following operating conditions: temperature 25–35 °C, relative humidity 85-95%, EBRT 20-30 s, mass loading 50–90 g-C/m^3·h, and surface loading 4–7 m^3/m^2·h.

Conclusions

The following conclusions could be drawn from this study:

1. Coal particles appear to be efficient for buffering the fluctuation in isopropyl alcohol (IPA) concentration.
2. Nearly complete IPA removal could be achieved for influent carbon loading between 6 and 88 g/m^3·h demonstrating the trickle-bed air biofilter

Table 5. Carbon balance analysis of each run

Run no.	Q_aC_i (mg-C/min)	Q_aC_e (mg-C/min)	Q_aC_c (mg-C/min)	αQ_lC_b (mg-C/min)	βQ_lC_l (mg-C/min)	R (%)
1	12.74	0.00	12.19	0.01	0.06	96
2	19.14	0.00	18.46	0.03	0.12	97
3	25.73	0.00	25.26	0.02	0.08	97
4	39.21	0.00	37.71	0.05	0.12	97
5	37.07	0.00	35.00	0.15	0.12	95
6	61.05	0.00	58.91	0.08	0.12	97
7	78.25	0.00	77.35	0.15	0.12	99
8	95.88	0.00	95.11	0.07	0.13	99
9	113.01	0.00	109.72	0.10	0.14	97
10	192.04	0.00	191.26	0.11	0.06	99

Note: Q_aC_i, influent carbon rate; Q_aC_e, effluent carbon rate; Q_aC_c, effluent carbon rate of CO_2 gas; αQ_lC_b, biomass production rate equivalent to carbon utilization rate; βQ_lC_l, effluent carbon rate of leachate; R, carbon recovery.

Table 6. Applicable operating conditions of TBAB in treating IPA

Parameter	Range	Typical
Temperature (°C)	25–35	30
Relative humidity (%)	85–95	90
EBRT (sec)	20–30	25
Mass loading (g-C/m^3·h)	50-90	
Surface loading (m^3/m^2·h)	4-7	
Removal efficiency (%)	90–99	95
Elimination carbon capacity (g/m^3·h)	45-89	

(TBAB) could be very efficient for controlling IPA emission under low-to-high carbon loading conditions.

3. From carbon balance analysis, high carbon recoveries (R) of 95-99% were achieved indicating the accuracy of test results. Most of the carbon in the effluent was from CO_2 production (above 99%). The carbon mass rate of the liquid effluent was approximately 2–3 orders of magnitude less than that of the CO_2 effluent indicating the dissolved IPA and its derivatives in leachate were not significant in TBAB.
4. High removal efficiencies of IPA could be achieved under following operating conditions: temperature 25–35 °C, relative humidity 85–95%, EBRT 20-30 s, mass loading 50–90 g-C/m^3·h, and surface loading 4–7 m^3/m^2·h.

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