Using a biological aerated filter to treat mixed water-borne volatile organic compounds and assessing its emissions

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Abstract

A biological aerated filter (BAF) was evaluated as a fixed-biofilm process to remove water-borne volatile organic compounds (VOCs) from a multiple layer ceramic capacitor (MLCC) manufacturing plant in southern Taiwan. The components of VOCs were identified to be toluene, 1,2,4-trimethylbenzene, 1,3,5-trimethylbenzene, bromodichloromethane and isopropanol (IPA). The full-scale BAF was constructed of two separate reactors in series, respectively, using 10- and 15-cm diameter polypropylene balls as the packing materials and a successful preliminary bench-scale experiment was performed to feasibility. Experimental results show that the BAF removed over 90% chemical oxygen demand (COD) from the influent with (1188 ± 605) mg/L of COD. A total organic loading of 2.76 kg biochemical oxygen demand (BOD)/(m³ packing-d) was determined for the packed bed, in which the flow pattern approached that of a mixed flow. A limited VOC concentration of (0.97 ± 0.29) ppmv (as methane) was emitted from the BAF system. Moreover, the emission rate of VOC was calculated using the proposed formula, based on an air-water mass equilibrium relationship, and compared to the simulated results obtained using the Water 9 model. Both estimation approaches of calculation and model simulation revealed that 0.1% IPA (0.0031–0.0037 kg/d) were aerated into a gaseous phase, and 30% to 40% (0.006–0.008 kg/d) of the toluene were aerated.

Key words: volatile organic compounds; biological aerated filter; total organic loading; emission

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Introduction

The submerged biological aerated filter (BAF) is a fixed-biofilm system used in the secondary and tertiary biological treatment of wastewater. Such a filter has three phases: a solid phase that supports microbial growth, a liquid phase in which the solid material is submerged, and a gas phase generated by the aeration of air supplied to the reactor. The flow patterns within the packing of the BAF are similar to plug flows when packing materials of smaller-size (equivalent diameter < 3 mm) are used. A superficial velocity of 1–10 m/h (0.000278–0.00278 m/s) is typically operated for the full-scale BAF with media packed to a depth of 2–4 m (Leopoldo and Stephenson, 1999). Parker and Brathy (2001) stated that the U.S. Water Environment Federation and the American Society of Civil Engineers used total organic loading (TOL) in kg BOD₅ (five day biochemical oxygen demand)/(m³ packing-d) for BAF design. Equation (1) determines TOL.

\[
\text{TOL} = \left( C_i - C_e \right) \frac{Q}{V} \tag{1}
\]

where, \( C_i \) (kg/m³) is influent BOD concentration; \( C_e \) (kg/m³) is effluent BOD concentration; \( Q \) (m³/d) is influent flowrate, and \( V \) (m³) is volume of packed media. Furthermore, Parker and Brathy (2001) investigated the TOL of BAF in three large wastewater treatment plants (WWTPs) in Central Valley (Utah, USA), Bi City (Colorado, USA), and Annacis Island (Canada). The influent TOL was as low as 0.6 kg BOD₅/(m³ packing-d) and the soluble BOD removal efficiency was 88%. However, TOL was approximately 3.0 kg BOD₅/(m³ packing-d), and only 50% of soluble BOD was degraded by BAFs.

The tests performed by Hill et al. (1991) established that a packed column yields a lower volatile organic compound (VOC) emission loss to the air stream than a completely stirred tank reactor (CSTR). Hill et al. used a BAF packed with glass beads to treat an extremely high loading (8.64 kg/(m³ packing-d)) of phenol. Only 0.07% of phenol were detected in the air stream and a low proportion, 33%, of phenol were biodegraded. The observation using a scanning electronic microscope showed that the biomass almost gathered on the surface of polyethylene media, but did not grow on the inner anaerobic packing zone (Liu et al., 2000). Goedertetz et al. (1990) also determined that an extremely long hydraulic retention time (HRT) of 193 h is required to enable a pilot anaerobic BAF to remove over 90% COD (chemical oxygen demand) from a heavily COD loaded wastewater, ranging from 18.8 to 25.0 kg COD/(m³ packing-d). Goedertetz et al. (1990) and Liu et al. (2000) found an aerobic BAF, with the modest aeration supplied...
for bio-oxidation and a shorter HRT than the anaerobic
BAF, is an economical design for wastewater treatment.

BAF has been successfully used to degrade water-
borne biorefractory organic compounds, including phenol
and halogenated hydrocarbons (Friday and Portier, 1991;
Seignez et al., 1993; Yu and Chou, 2000), and wastewater
of dying (Liu et al., 2009), textile (Liu et al., 2008)
and leachate from a landfill (Wang et al., 2009). Pre-
liminary feasibility studies or pilot tests have often been
performed to determine the performance of BAF and
exactly evaluate the effects of different kinds of bio-phase
support media and operating conditions on the treatment
of industrial effluent. For example, Lahiere and Goodboy
(1993) established a pilot BAF with a ceramic membrane
to treat petrochemical wastewater. Wilderer et al. (2000)
determined four different types of support media including
blasted clay granules, granular activated carbon, zeolite
and small plastic rings in a series of feasibility studies and
pilot-scale test of BAF to remove nitrogen and phosphate
substrates from a tourist area.

In this study, a full-scale BAF with two beds in series
was used for treating multiple organic solvents, aggluti-
native agents, and a few detergents, flowing out from a
multiple layer ceramic capacitor (MLCC) manufacturing
plant in Kaohsiung, Southern Taiwan. The BAF was
designed according to the results of a successful feasibili-
ty study, in which the packing media were synthetic
polypropylene (PP) products. The organic compounds in
the wastewater were identified as aromatic hydrocarbons
(toluene, 1,2,4-trimethylbenzene, 1,3,5-trimethylbenzene
and bromodichloromethane) and isopropanol (IPA). Data
concerning the operations in the field, obtained over four
months, were used to determine optimal operating condi-
tions of BAF for polishing effluent COD at 100 mg/L,
and BOD at 30 mg/L. Additionally, the emission of VOC from
the reactors was evaluated by field sampling, and compared
to the amount of VOC fed in the aqueous phase and that
degraded by biomass.

1 Methodology

1.1 Experimental setup and materials

The preliminary feasibility study used PP balls (Tri-
Pack® No.1, Jaeger Products, Inc., USA) as packings to
treat the MLCC wastewater for over 35 d. The packing
material had a diameter of 5 cm and a porosity of 93%.
Each batch of collected water had a volume of around 120
L and fed over 10 d into the reactor in the laboratory. The
full-scale BAF was designed according to the data on the
successful operation of feasibility study.

Figure 1 schematically depicts the process diagram for
polishing the wastewater from an MLCC manufacturing
plant. The influent wastewater fed into the BAF was pre-
treated using a coagulation-settlement package to remove
the suspended solids of ceramic powder. The BAF reactor
was built from stainless steel and the dimensions of the
packing in TK-01A/B were 5.5 m (L) × 1.7 m (W) × 4.5 m
(H). The PP packing materials in the first BAF tank (TK-
01A) had a diameter of 10 cm (porosity 0.95), and those
in the following tank (TK-01B) had a diameter of 15 cm
(porosity 0.93). Both packings were locally manufactured.
The packed bed was flooded beneath 10 cm of water.

For the suddenly high organic load from the manufac-
turing process, a sufficient margin was considered of a low
superficial velocity in the packed bed of BAF. Therefore,
the mean superficial velocity in the packed zone was
estimated to be 0.223 m/h, a typical operation range below
1–10 m/h, with a low hydraulic loading of 50 m³/d. The
hydraulic retention time was 1.68 d, based on the total
packing volume 84 m³ in TK-01A/B. The air from blowers
(BL-01A/B, 2.2 kW, local manufacturer, Taiwan and one
spare) flowed at 81 m³/h was provided to maintain 4–
6 mg/L dissolved oxygen in the packed zone. The pH
of the water was maintained at range 6.5–8.0 using the
analytical-indicating controllers to ensure microorganisms
could survive.

![Fig. 1 Schematic flow diagram of wastewater treatment. TK-01A/B: BAF reactor (two tanks in series); TK-02: settlement tank; TK-03: sandfilter influent tank; TK-04: sandfilter tank; P-01A/B: sandfilter influent Pump; P-02: sludge transfer pump; P-03: bubble deforming pump; B-01A/B: blowers.](image-url)
1.2 Analysis

Routine sampling and analysis of the liquid phase were essential in evaluating the performance of the system. COD and BOD in influent and effluent were analyzed to identify the organic compounds. COD and total organic carbon (TOC) of the circulating water with height variations in the packed zone were also checked during the preliminary feasibility test to determine the flow pattern in the zone. All aqueous analysis followed Standard Methods for the Examination of Water and Wastewater, established by American Public Health Association (APHA, 1992). TOC was analyzed using a Model 700 analyzer, O. I. Analytical Corporation (USA). A gas chromatograph/mass spectrum (GC-MS, 6890 and 5973 Series, Agilent, USA) was used to identify the components of the water-borne multiple VOCs in the effluent from the MLCC manufacturing process. The procedures were performed using a purge and trap system (O. I. Analytical Corporation, USA), and then the purged gaseous phase was injected into a gas chromatograph equipped with a flame ionization detector (GC-FID) (6890 Series, Agilent, USA), connected to a mass spectrometer system (5973 Series, Agilent, USA).

1.3 Measurement of VOC emission

The VOC emission rate \( N \) (g/h) was measured according to the estimation equation of Cheng and Chou (2003) (Eq. (2)).

\[
N = \frac{Q_{g}}{C_{e}^{*}} \quad (2)
\]

where, \( Q_{g} \) (m³/h) is aerated rate of a blower, and \( C_{e}^{*} \) (g/m³) is gaseous VOC concentration in equilibrium in the aqueous phase, which is determined by the gas-liquid equilibrium experiment (Fig. 2). A 42-mL opaque glass vial was submerged into the water for liquid sample collection in the field. This was performed as gently and slowly as possible to prevent the generation of gas bubbles. \( C_{e}^{*} \) was measured using a GC-FID (6890 Series, Agilent, USA). The concentration \( C_{e} \) of emitted VOC was also determined in the field, followed the description of Cheng and Chou (2003), using a portable GC-FID (TVA-100B, Thermo, USA) with a sampling tube 10 cm above the water, and undisturbed by the aerated bubbles.

The emission rate of specific constituents of VOC from the air-water interface in BAF system was estimated using the model Water 9 (2001), revised by the USEPA. Water 9 model requires following parameters: (a) water content parameters, including hydraulic loading (m³/d), water temperature \( T_{w} \), total dissolved solids (TDS), total suspended solids (TSS) and aqueous VOC concentrations \( C_{i} \), surface wind and ambient temperature; (b) equipment specifications, including tank dimensions (length \( \times \) width \( \times \) packed depth), rate of aeration, total bed porosity; and (c) properties of the target VOC components and the biological reaction coefficients taken from the model database. Typically, the related outputs as exhaust rate (g/s) include the fractions emitted in gaseous phase, taken in biomass, and dissolved in water.

2 Results and discussion

2.1 Feasibility study of laboratory-scale BAF

A bench-scale BAF, with 5 cm PP packing material, was operated more than 850 h in a feasibility study. Field batch collection revealed a stable influent COD from 120 to 368 mg/L (average 225 mg/L). The BAF system degraded more than 80% of the influent COD, and the effluents had COD of (43 ± 37) mg/L. An average TOL of 0.165 kg BOD/(m³ packing-d) was obtained over all of the runs according to Eq. (1), with an effective packed volume ratio of 0.07 (porosity 93%), a space velocity 0.33 d⁻¹ and an average BOD/COD of approximately 0.3 ((0.225–0.043) kg COD/m³ × 0.3 (kg BOD/(kg COD))/0.33 d⁻¹ = 0.165 kg BOD/(m³ packing-d)). Notably, the effluent COD < 100 mg/L implies that BAF is feasible and practical for polishing the effluent of biorefractory organics extracted from the existing coagulation-settlement system.

2.2 Startup of full-scale BAF

During the startup period, the packed media of BAF were initially seeded with sludge. Raw sludge was transferred from WWTP in the local area to manufacturing electronic parts. The startup period of 104 d was divided into two periods, according to the organic removal efficiency, and plotted in Fig. 3. The influent COD was (1910 ± 1031) mg/L and less than 20% COD was removed during the first 26 d. The low COD removal efficiency resulted from the poor performance of the fresh microorganisms. In fact, the target wastewater included complex effluent organics (solvents, agglutinative agents, and detergents) with unstably high COD loads in the manufacturing process, as well as additional acidic and basic ions from the blow-down streams of the scrubbing and deionization system.

BAF was reseeded with sludge at day 28 and COD feeding was kept as constant as possible by preventing an extremely high-load organics from the processing line.
Figure 3 shows that during the period of day 64 to day 90, the influent COD was (1188 ± 605) mg/L and more than 90% COD was removed from the effluent. Meanwhile, a satisfactory TOL of 2.76 kg BOD/(m$^3$ packing-d) was obtained with an effective packed volume ratio of 0.06 (average porosity 94%), a space velocity of 1.2 d$^{-1}$ and an average BOD/COD of 0.26 ((1.188 kg COD/m$^3$ × 0.9 × 0.26 (kg BOD/(kg COD)) × 50 m$^3$/d)/84 m$^3$ packing/(1–0.94) = 2.76 kg BOD/(m$^3$ packing-d)). These results are similar to the TOL data, compiled by Parker and Brathy (2001). However, during the period after day 99, a decreasing degradation of COD was resulted from acidic and basic substances feeding into the BAF from the deionization.

### 2.3 Performance characteristics of the BAF

Biomass was sampled from the top, middle, and bottom of the packed zone in the bench-scale BAF to evaluate the biomass characteristics. Two packing balls were randomly selected from each sampling site. A dry-base biomass density of 4.65 kg/m$^3$ in the packed zone was measured. The result is close to 6.0 kg/m$^3$ given in the Contacted Aeration Treats Industrial Wastewater (1991).

The hydraulic flow pattern in the packed zone is specified by the variations of aqueous COD and TOC with different heights in the packed zone of the bench-scale reactor. Figure 4 indicates that neither COD nor TOC vary with the depth of the packed zone, showing that the flow pattern of the BAF reactor approaches that of a CSTR, which is like the hydraulic flow pattern model of BAF developed by Kim et al. (2008). The hydraulic flow pattern, almost as the same as a CSTR, resulted from the loose packing of large-sized balls with diameters of 5 cm and a high total porosity of 93%. Two types of large packing balls (with diameters of 10 or 15 cm, and a total porosity of 94%) were used to simulate the flow pattern of CSTR for the full-scale BAF.

According to the findings, alcohols are likely to promote the depletion of DO and reduce the extent of aerobic degradation of aromatic hydrocarbon (Corseuil et al., 1998; Ramirez-Lopez et al., 2000; Powers et al., 2001). Moreover, the accumulation of toxic secondary metabolites (acetaldehyde, acetic acid, ethyl acetate) results in a low bioactivity, then results in an extremely low BOD/COD of under 0.3.

### 2.4 Estimation of VOC emissions from the BAF system

Additional samplings were taken in the field to evaluate the VOC emission rates from the full-scale BAF. $C_g$ above the water and $C_{g*}$ of the liquids in the BAF were determined. The emitted $C_g$ was (0.97 ± 0.29) ppmv (as methane) during a period of 120 min in the field. The $C_{g*}$ values of IPA and toluene were (0.037 ± 0.012) and (0.020 ± 0.0062) g/m$^3$, respectively. The $C_{g*}$ values of all other VOC components were under 0.0001 g/m$^3$. Furthermore, the $C_L$ values of IPA and toluene were 142 and 0.40 mg/L, respectively, according to analysis using a GC-MS system. For a hydraulic loading of 50 m$^3$/d, 7.1 kg IPA and 0.02 kg toluene were dissolved into the water phase of BAF daily. Notably, the partitioning coefficient of toluene ($C_{g*}/C_L$) was 0.02/0.40 = 0.05, which is much lower than 0.23 for pure toluene dissolved in water (Cheng et al., 2004). The co-solute effect, for toluene in aqueous IPA of high concentration, was conspicuous (Cheng et al., 2003).

Gupta and Thodos (1962) estimated the reduction percentage ($\eta_1$) of VOC emission for aeration passing through a packed bed of a high porosity. $\eta_1$ depends on the Reynolds’s number (Re) and the porosity ($\epsilon$) of the packed bed. Re can be calculated using Eq. (3).

\[
Re = \frac{\epsilon \mu D_e}{\nu} = \frac{\rho Q D_e}{\epsilon \mu A_c}
\]  

(3)

where, $\rho$ is a density of water (approximately 996 kg/m$^3$ at 27°C); $\nu$ (m/s) is superficial velocity passing through the packed bed; $D_e$ is equivalent diameter of a square section of the packed bed (approximately 3.21 m, estimated from the cross section of 5.5 m (L) × 1.7 m (W)); $\mu$ is viscosity of water (about 0.00086 kg/(m·s) at 27°C); $Q$ is influent water rate (50 m$^3$/d), and $A_c$ (m$^2$) is an area of the cross section of the packed bed. The density and viscosity of water at 27°C are taken from the book of Perry and Green (1997).

With Re = 245 calculated from Eq. (3) and the porosity of 0.94 in the packed bed, $\eta_1$ is found to be about 0.046, according to Gupta and Thodos (1962). Table 1 summarizes the emission rate of VOC, as estimated by Eq. (2), multiplied by two coefficients, $\eta_1$ (0.046) and $\eta_2$ (0.95, an assumed aeration efficiency of the installed blower). Furthermore, the model Water 9 (2001) was also
applied to determine the emission rate from the BAF. Notably, the $C_L$ of ethanol and toluene, were lower than the detected limits to be neglected in the treated stream. The aerated phase of IPA represents less than 0.1% of the liquid phase. That is, the biomass degraded almost all IPA. In contrast, the proportion of toluene that is aerated ranges from 30% to 40%, based on the two assessment approaches. The competition between toluene and IPA for oxygen may be a key reason for the weak biodegradation of toluene by microorganism. Additionally, a large aqueous ratio of IPA/toluene (Table 1, 7.1/0.02 = 350) resulted in the dominant microorganism for the degradation of IPA, but not for toluene.

### 3 Conclusions

BAF is proven to be an effective technique for polishing the multiple VOC-rich wastewater from an electronic parts manufacturing plant. A TOL of 2.76 kg BOD/(m³ packing-d) was obtained for a ratio of BOD/COD as low as 0.26 in wastewater. Actually, more than 90% of COD in the feeding stream can be treated within the packed bed, containing PP ball and having a total porosity of 94%. A COD removal capacity was 10.6 kg/(m³ packing-d) within the media.

The aqueous solvents from the MLCC processing were effectively treated by the BAF system, and less than 0.1% of the IPA were aerated from the BAF system. However, 30%–40% of the toluene were stripped into the atmosphere. This finding indicates that toluene in ethanol-rich wastewater is slightly biodegraded, because IPA is a stronger donor of electrons in an aerobic condition comparing to toluene. Notably, the total gaseous emission rates ofIPA and toluene were 0.0097–0.011 kg/d, which were very limited emission rates comparing to the feeding organic matters.

### References


### Table 1 Fate of VOCs in the BAF system

<table>
<thead>
<tr>
<th>VOC component</th>
<th>Liquid phase $L_1$ (kg/d)</th>
<th>Effluent rate (kg/d)</th>
<th>Gaseous phase $A_1$ (kg/d)</th>
<th>$A_2$ (kg/d)</th>
<th>Emission ratio $A_1/L_1$ (%)</th>
<th>$A_2/L_1$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>IPA</td>
<td>7.1</td>
<td>–</td>
<td>0.0031</td>
<td>0.0037</td>
<td>0.044</td>
<td>0.052</td>
</tr>
<tr>
<td>Toluene</td>
<td>0.02</td>
<td>–</td>
<td>0.008</td>
<td>0.006</td>
<td>40</td>
<td>30</td>
</tr>
</tbody>
</table>

$L_1$: influent rate. $A_1$: emission rate calculated using Eq. (2), and $\eta_1$ and $\eta_2$, where $C_p^*$ for ethanol and toluene are 0.037 and 0.02 g/m², respectively. $Q_L = 81$ m³/h, $\eta_1 = 0.046$; and $\eta_2 = 0.95$. For example, IPA emission rate is calculated as $(0.037 \text{ g/m}^2) \times (81 \text{ m}^3/\text{h}) \times (24 \text{ h/d}) \times (1 \text{ kg/1000 g}) \times 0.046 \times 0.95 = 0.0031 \text{ kg/d}$. $A_2$: emission rate simulated by Water 9 model.

$\cdots$: aqueous ethanol and toluene were not detected in the effluent stream of BAF.


