



## Application of Fibrous Activated Carbon Filter in Continuous-Flow Unit for Removal of Volatile Organic Compounds under Simulated Indoor Conditions

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### ABSTRACT

This work explored the feasibility of applying fibrous activated carbon (FAC) for use as volatile organic compound filters in portable indoor air cleaning devices under simulated indoor conditions. The saturation time for benzene, toluene, ethyl benzene, and xylene (BTEX) decreased from ~11 to ~3 h, ~17 to ~8 h, ~52 to ~17 h, and ~54 to ~27 h, respectively, as the relative humidity (RH) increased from 20 to 90%. Consistently, the adsorption capacities for BTEX decreased as the RH increased. The saturation time for BT decreased from ~11 to ~2 h and ~70 to ~22 h, respectively, as the initial concentration (IC) increased from 0.1 to 2.0 ppm. In contrast, the adsorption capacities for BT increased from 1.8 to 7.5 mg/g and 8.9 to 53 mg/g, respectively, as the IC increased from 0.1 to 2.0 ppm. The saturation time for BT decreased from ~24 to < 4 h and ~132 to ~22 h, respectively, as the stream flow rate (SFR) increased from 1 to 7 L/min. In addition, the adsorption capacities for BT decreased from 55 to 3.1 mg/g and 308 to 36 mg/g, respectively, as the SFR increased from 1 to 7 L/min. The regeneration efficiencies for both target compounds increased as the desorption temperature increased from 50 to 200°C, while the regeneration times decreased. Overall, this study provides information regarding the optimal operational and regeneration conditions of FAC.

**Keywords:** Adsorption capacity; Regeneration; Saturation time; VOC filter; Indoor condition.

### INTRODUCTION

Volatile organic compounds (VOCs) are commonly detected in residential and occupational indoor environments (Jia *et al.*, 2008; Caro and Gallego, 2009). Benzene, toluene, ethyl benzene, and xylene (BTEX) comprise a VOC group that is usually present at higher concentrations in urban indoor air than in outdoor air (Jia *et al.*, 2008). These elevated indoor BTEX concentrations are ascribed to emission of a range of indoor sources, including consumer products, cigarette smoke, and/or building materials, as well as the penetration of outdoor air polluted with these compounds (Jia *et al.*, 2008; Schlink *et al.*, 2010). Moreover, BTEX are hazardous to human health and have the potential to induce a variety of types of DNA damage (Chen *et al.*, 2008). As such, methods for the control of BTEX are necessary to reduce their indoor concentration levels.

Several methods have been developed to remove VOCs in the gaseous phase, the primary ones being bio-filtration (Cai *et al.*, 2007), thermal or catalytic incineration

(Salvador *et al.*, 2006; Wang *et al.*, 2006), scrubbing (Yang *et al.*, 2011), and physical adsorption (Tsai *et al.*, 2008; Shiue *et al.*, 2010). Adsorption via granular-type activated carbon (GAC) is typically used to control indoor VOCs in non-industrial buildings (Haghighat *et al.*, 2008; Howard-Reed *et al.*, 2008; Bastani *et al.*, 2010). However, despite its widespread utilization, GAC is hampered by the high-pressure drop that occurs as a result of air flow through the packed media. Fibrous activated carbon (FAC) is a recent alternative adsorbent that overcomes this drawback (Sidheswaran *et al.*, 2012). In addition, FAC has a larger specific surface area than GAC, higher adsorption capacity, faster equilibrium for adsorption, and low pressure drop (Das *et al.*, 2004; Fournel *et al.*, 2005; Lorimier *et al.*, 2005; Tsai *et al.*, 2008). These advantages make FAC a promising candidate for use in VOC filters in portable indoor air cleaning devices, as well as in building heating, ventilation and air conditioning (HVAC) systems.

Several studies (Das *et al.*, 2004; Tsai *et al.*, 2008; Águeda *et al.*, 2011) have suggested that the adsorption performance of VOCs can be varied according to different parameters, such as adsorbate type, initial concentrations (ICs) and stream flow rates (SFRs) as well as relative humidity (RH). Tsai *et al.* (2008) reported the effect of adsorbates at high IC ranges (43–7800 ppm) on the adsorption characteristics of FAC and other adsorbents. Das *et al.* (2004) also reported

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the dependence of ICs of selected VOCs at 2000–10,000 ppm and SFRs at 0.2–1.0 L/min on the adsorption capacity of FAC. Moreover, Águeda *et al.* (2011) found that RH decreased the adsorption capacities of AC monoliths for dichloromethane at unknown concentrations at the breakthrough and saturation time. However, these studies mainly examined low to hundred ppm concentration levels, which are more common for industrial emission concentrations than indoor air concentrations. The extrapolation of adsorption properties obtained at concentrations substantially greater than those for the desired purpose is not always appropriate. Therefore, studies to confirm whether the patterns previously obtained at high IC conditions were still applicable to those of low indoor air quality (IAQ) levels were conducted. Although, Sidheswaran *et al.* (2012) reported that FAC could be employed to effectively control VOCs at IAQ levels, their study was carried out to explore the application of FACs to VOC cleaning in building HVAC systems, but not for portable indoor air cleaning devices.

Consequently, the present study explored the feasibility of applying FAC for use as VOC filters in portable indoor air cleaning devices. This was accomplished using a continuous-flow unit under simulated indoor conditions. The target adsorbing species included four different VOCs (BTEX), which are known to be common indoor pollutants (Jia *et al.*, 2008; Schlink *et al.*, 2010). The RH range investigated in this study (20–90%) covers both dry and humid conditions that can be encountered in actual indoor environments. Moreover, ICs were tested to include typical IAQ levels (Jia *et al.*, 2008; Schlink *et al.*, 2010). Specifically, this study determined the adsorption capacity and saturation period of a commercially-available FAC for BTEX mixture adsorption. In addition, the saturated FACs were regenerated by applying heated air at different temperatures to desorb previously adsorbed BTEX from the FAC filter and their regeneration efficiencies were estimated. The results of this study are expected to be applied to determination of a reduced rate for FAC exchanges for indoor uses, as well as

to establish a control strategy for indoor VOC levels in non-industrial buildings.

## METHODOLOGY

### Study Design

This study consisted of two different experiments: an adsorption test of FAC under simulated indoor conditions and a regeneration test of saturated FAC. For these experiments, a continuous-flow adsorption unit containing FAC (0.1 cm thickness and 6.2-cm diameter) was prepared using a double-cylindrical Pyrex tube (Fig. 1). A felt-form FAC (Viscose rayon base) was purchased from Korean Activated Carbon Fiber Ltd. The micropore diameter, surface area, and carbon content of the FAC were 20 Å, 1000 m<sup>2</sup>/g, and 70 g/m<sup>2</sup>, respectively.

To test the adsorption of FAC, simulated indoor conditions were determined based on the RH and IC. Gas sampling was conducted at the down- and up-stream ports of the adsorption unit until the ratios of outlet to inlet concentrations of BTEX or BT mixture were equal to or greater than 1. The RHs investigated in this study were 20, 45, 70, and 90%. Humidified air was prepared using water-containing impingers partially immersed in a water bath fixed at 35°C. The SFR was adjusted to 1.0, 3.0, 5.0, or 7.0 L/min and four different ICs (0.1, 0.5, 1.0, and 2.0 ppm) were accomplished using an auto-programmed syringe pump (Model 210, KdScientific Inc.). In addition, the SFR ranged from 1.0–7.0 L/min to cover a wide range of face velocities (0.3–2.1 m/min). To test each parameter, the RH, SFR, and IC were adjusted to 45%, 5 L/min, and 1 ppm, respectively.

To test the regeneration of saturated FAC, humidified air (45%) was passed through saturated FAC filters heated at four different temperatures (50, 100, 150, and 200°C) until no chemicals were observed downstream of the regeneration unit. A fresh FAC filter was saturated with BT mixture on the basis of the adsorption time determined in the adsorption experiments. For this test, the experimental conditions

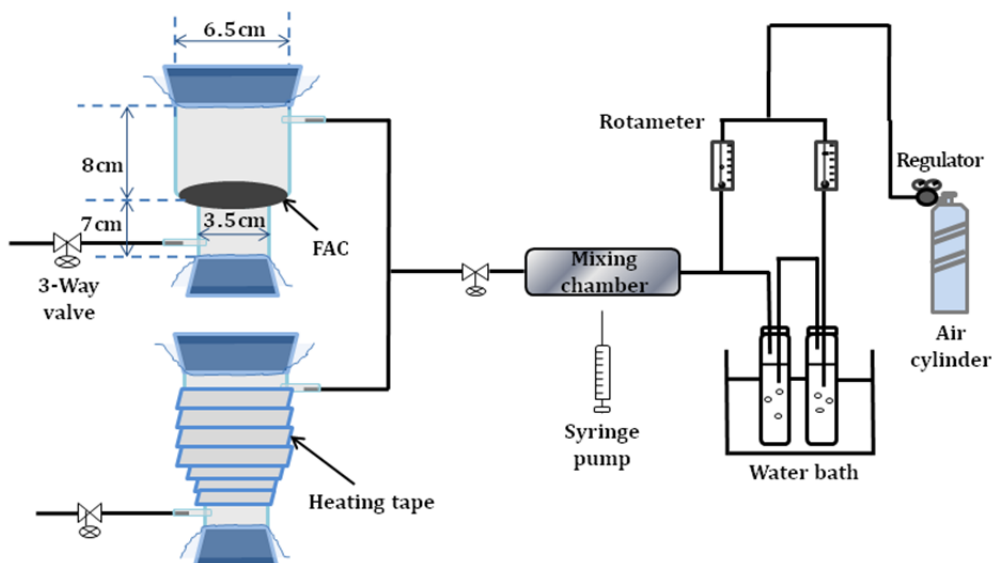


Fig. 1. Schematic diagram of the experimental set-up.

were adjusted to the above representative values of RH, SFR, and IC, and the saturated FAC filter was regenerated by passing humidified air through it while heating the filter at the specified temperature. The air flow for regeneration of saturated FACs was directed opposite to that of the adsorption tests. Ten-min of air sampling was conducted at the outlet of the regeneration unit.

### BTEX Measurements

Air sampling was conducted using an evacuated Teflon bag, after which the air was drawn into a Tenax TA (0.3 g) trap via a personal air sampler (SKC Aircheck Sampler Model 224-PCXR8). The sampling flow rates were 0.02–0.1 L/min. Downstream sampling was conducted via a 3-way valve installed at the downstream port of the plug-flow reactor. The collected target VOCs were analyzed using a gas chromatograph (GC, Hewlett Packard 7890) and a flame ionization detector (HP 5890II) or a mass spectrometer (HP MSD5973) system coupled to a thermal desorption system (Perkin Elmer ATD 400).

The quality control for measurement of gas-phase chemicals involved analyses of lab blank and standard samples. On each day, a blank sample was examined for contamination and one standard sample was analyzed daily to identify the instrumental response of the analytical system. A new calibration curve was generated, if the instrumental sensitivity differed by more than  $\pm 10\%$  from that estimated by the pre-determined calibration equation. The detection limits ranged from 0.2 to 0.6 ppb, depending on the chemical types.

### Estimation of Adsorption Capacity and Regeneration Efficiency

The adsorption capacity of the FAC was calculated by linking the inlet and outlet concentrations of BTEX obtained during the adsorption process to the following equations:

$$C_{ai} = M_{ai}/M_f \quad (1)$$

$$M_{ai} = \sum_{ij} \quad (2)$$

$$m_j = \int_0^t (C_i - C_{o,j}(t))Qdt \quad (3)$$

where,  $C_{ai}$  is the adsorption capacity of chemical  $i$  on the FAC (mg/g);  $M_f$  is the mass of the FAC (g);  $M_{ai}$  is the total mass of chemical  $i$  adsorbed on the FAC (mg);  $m_{ij}$  is the mass of chemical  $i$  adsorbed on the FAC (mg) during the  $j^{\text{th}}$  adsorption period;  $C_i$  is the inlet concentration of chemical  $i$  ( $\text{mg}/\text{m}^3$ );  $C_{o,j}(t)$  is the outlet concentration of chemical  $i$  ( $\text{mg}/\text{m}^3$ ) obtained downstream of the FAC during the  $j^{\text{th}}$  adsorption period; and  $Q$  is the air flow rate for adsorptions ( $\text{m}^3/\text{min}$ ).

The regeneration efficiency of the FAC saturated with BT mixture was estimated using the outlet concentrations obtained during the desorption process, and the following equations:

$$R_c = M_d/M_a \quad (4)$$

$$M_d = \sum m_{d,j} \quad (5)$$

$$M_a = \sum m_{a,j} \quad (6)$$

$$m_{d,j} = \int_0^t (C_{\text{out},d,j}(t))Qdt \quad (7)$$

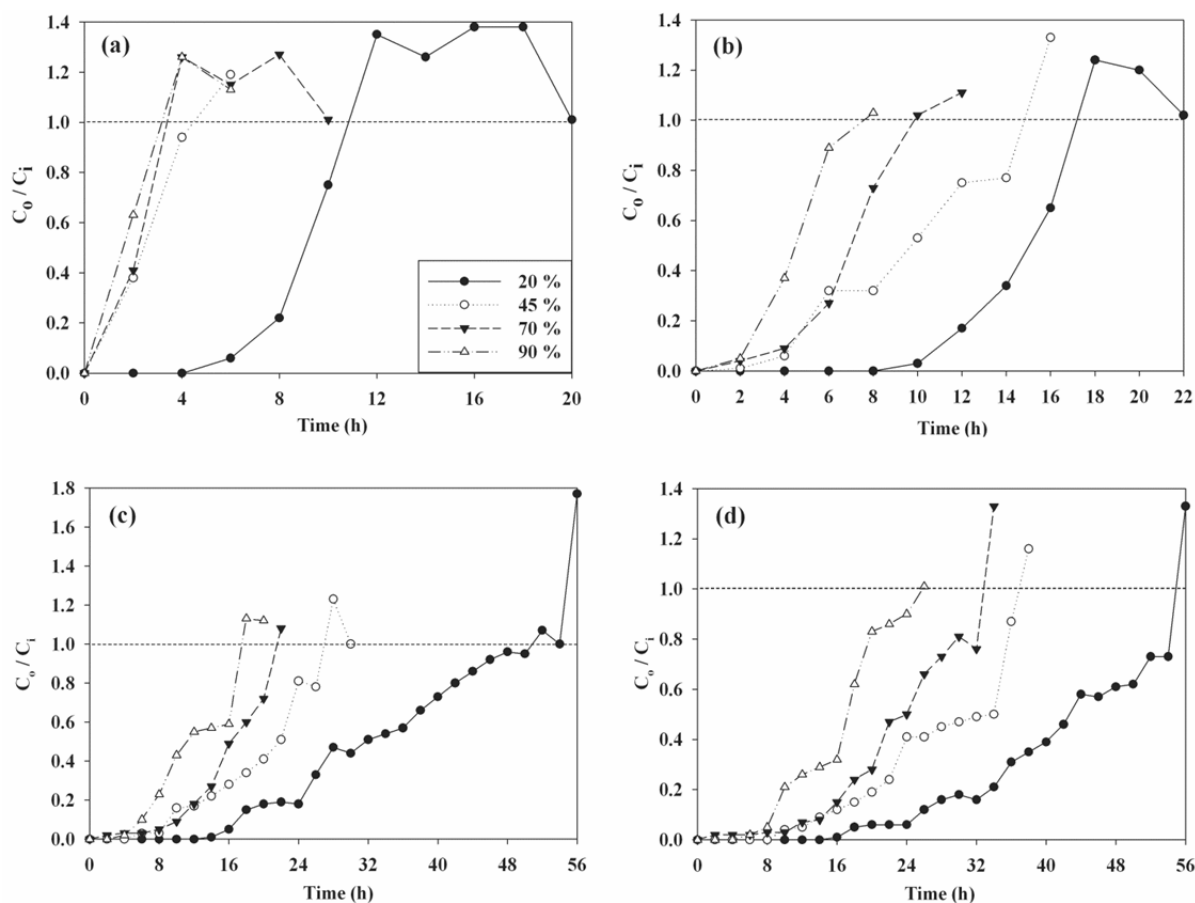
$$m_{a,j} = \int_0^t (C_{\text{in}} - C_{\text{out},a,j}(t))Qdt \quad (8)$$

where,  $R_c$  is the regeneration efficiency of FAC for benzene or toluene;  $M_d$  is the total mass of benzene or toluene desorbed from the FAC (mg);  $M_a$  is the total mass of benzene or toluene adsorbed on the FAC (mg);  $m_{d,j}$  is the mass of benzene or toluene desorbed from the FAC (mg) during the  $j^{\text{th}}$  adsorption period;  $m_{a,j}$  is the mass of benzene or toluene adsorbed on the FAC (mg) during the  $j^{\text{th}}$  adsorption period;  $C_{\text{out},d,j}(t)$  is the benzene or toluene concentration ( $\text{mg}/\text{m}^3$ ) obtained downstream of the FAC during the  $j^{\text{th}}$  desorption period;  $C_{\text{in}}$  is the inlet concentration of benzene or toluene ( $\text{mg}/\text{m}^3$ );  $C_{\text{out},a,j}(t)$  is the benzene or toluene concentration ( $\text{mg}/\text{m}^3$ ) obtained downstream of the FAC during the  $j^{\text{th}}$  adsorption period; and  $Q$  is the air flow rate for adsorptions ( $\text{m}^3/\text{min}$ ).

## RESULTS AND DISCUSSION

### Effect of RH and IC on Adsorption Performance of FAC

The FAC installed in a continuous-flow unit was applied to purification of BTEX under a range of conditions determined based on the RH and IC. No reacted VOCs other than BTEX were observed at the outlet of the plug-flow reactor. Fig. 2 shows the adsorption breakthrough profile of the FAC filter for the BTEX mixture according to RH. Both the saturation and breakthrough times for BTEX decreased with increasing RH. The saturation time for BTEX decreased from  $\sim 11$  to  $\sim 3$  h,  $\sim 17$  to  $\sim 8$  h,  $\sim 52$  to  $\sim 17$  h, and  $\sim 54$  to  $\sim 27$  h, respectively, as the RH increased from 20 to 90%. Consistently, the adsorption capacity of the FAC filter revealed a descending trend with increasing RH (Table 1). The adsorption capacities for BTEX decreased from 29 to 4, 55 to 16, 138 to 52, and 183 to 71 mg/g, respectively, as the RH increased from 20 to 90%. The descending trend in both the saturation time and adsorption capacity is ascribed to the competition of water molecules and BTEX molecules for the active adsorption sites on the FAC surface. More water molecules would be adsorbed on the FAC surface under highly humidified conditions. Similarly, previous studies (Tao *et al.*, 2006; Jo and Yang, 2009; Águeda *et al.*, 2011) showed that the AC adsorption capacity of several species decreased with increasing RH. Moreover, after the ratios of inlet to outlet concentrations of target compounds reached to 1, which reflect a saturation condition, they rolled-up. These roll-up phenomena were attributed to off-gassing of the compounds previously



**Fig. 2.** Adsorption breakthrough profile of fresh FAC filter for benzene (a), toluene (b), ethylbenzene (c), and xylene (d) according to the relative humidity (20, 45, 70, and 90%).

**Table 1.** Adsorption capacity (mg/g) of the FAC filter for BTEX according to the relative humidity.

Compound	20%	45%	70%	90%
Benzene	29	7	6	4
Toluene	55	36	25	16
Ethyl benzene	138	85	70	52
Xylene	183	122	101	71

adsorbed onto the surface of the FAC, which might have been displaced by water molecules. Águeda *et al.* (2011) also reported a roll-up effect when AC monoliths were used for the adsorption of gaseous dichloromethane.

Fig. 2 also shows the saturation and breakthrough time for the target compounds in ascending order: BTEX. Specifically, the saturation time for benzene was ~11 h under 90% RH conditions, whereas it was ~56 h for xylene. The adsorption capacity for these compounds also showed the same ascending trend under the same RH conditions (Table 1), suggesting that it depends on chemical type. This suggestion is supported by Fuertes *et al.* (2003), who reported that the adsorption capacity showed a strong dependence on the nature of the adsorbate. For example, the adsorption capacity for benzene was 29 mg/g under 20% RH, whereas it was 183 mg/g for xylene. This order is ascribed to the solubility and molecular weight of these

compounds. Daifullah and Girgis (2003) reported that for GAC the water solubility order was BTEX in descending order. A decrease in water solubility indicates a decrease in the affinity to water molecules on the adsorbent surface (Cal *et al.*, 1996), which results in a higher FAC adsorption capacity for compounds with low water solubility. In addition, species with higher molecular weight are preferred for the adsorption sites (Daifullah and Girgis, 2003), resulting in a higher FAC adsorption capacity for compounds with high molecular weight.

The breakthrough and saturation time and adsorption capacity for BTEX obtained in this study were compared with those of previous studies. The saturation and breakthrough time of the present study were longer than those (< 2 h) reported by Das *et al.* (2004), which was mainly attributed to differences in the initial concentrations of the two studies. Specifically, Das *et al.* (2004) used 2000–10000 ppm of toluene, which was  $10^3$ – $10^5$  times higher than the values used in the present study and resulted in their having a shorter breakthrough and saturation time. In addition, the maximum toluene adsorption capacity obtained in the present study (55 mg/g) was lower than that (81.6 mg/g) reported by Yao *et al.* (2009). Unlike the current study, this previous study was performed using the adsorption of toluene alone. Sidheswaran *et al.* (2011) found that individual VOCs and mixtures of VOCs did not follow the same isotherm

adsorption processes, which suggests that for VOC adsorptions, the single-component adsorption model would be different from the multi-component adsorption model. For the mixed VOCs, the adsorption capacity would likely be lower due to competitive adsorption of different VOC molecules (Sidheswaran *et al.*, 2011). In contrast, the sum (250 mg) of the BTEX masses adsorbed onto 1 g of FAC obtained from the present study under the 45% RH conditions was higher than the sum of the six VOC mixture (BTX, 1-butanol, undecane, and formaldehyde) masses (90.7 mg) per gram of FAC reported by Sidheswaran *et al.* (2012) under 30% RH. This difference is ascribed to differences in experimental parameters, such as the characteristics of FACs and the nature of the VOC mixture.

Fig. 3 exhibits the adsorption breakthrough profile of the FAC filter for the BT mixture according to IC. The saturation and breakthrough time for BT decreased with increasing IC. Specifically, the saturation time for BT decreased from ~11 to ~2 h and ~70 to ~22 h, respectively, as the IC increased from 0.1 to 2.0 ppm. Consistently, Das *et al.* (2004) reported that the saturation time decreased from 70 to 40 min as the initial concentration of toluene increased from 2000 to 10000 ppm. This descending trend in saturation time with increasing IC was attributed to the amount of BT molecules transferred to the FAC surface per unit of time. For low ICs, less BT molecules enter the macro-pores of the FAC, thereby hastening saturation of the FAC. In contrast, the adsorption capacity for these compounds revealed an ascending trend with increasing IC (Table 2). The adsorption capacities for BT were increased from 1.8 to 7.5 mg/g and 8.9 to 53 mg/g, respectively, as the IC increased from 0.1 to 2.0 ppm. This ascending trend of adsorption capacity was ascribed to higher competitive adsorption of BT molecules on the FAC

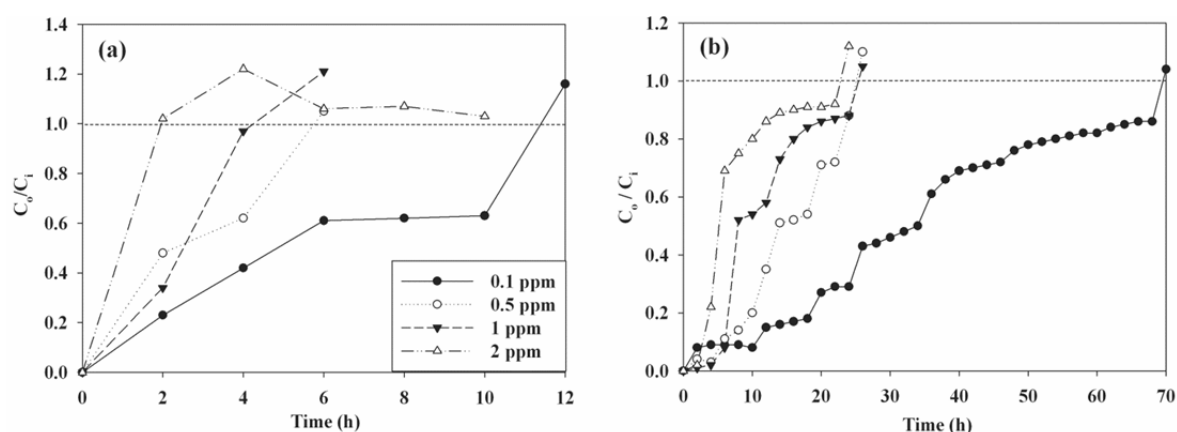
surface under high IC conditions.

#### Effect of SFR on Adsorption Performance of FAC

Fig. 4 shows the adsorption breakthrough profile of the FAC filter for the BT mixture according to SFR. The saturation and breakthrough time for BT decreased as the SFR increased. Specifically, the saturation time for BT decreased from ~24 to < 4 h and ~132 to ~22 h, respectively, as the SFR increased from 1 to 7 L/min. Similarly, Das *et al.* (2004) found that the saturation time of toluene decreased from 20 to 3 min as the SFR increased from 0.25 to 1 L/min. As expected, the early saturation of FAC observed under higher SFR conditions was ascribed to greater amounts of adsorbate being transferred to the FAC per unit of time. Moreover, the adsorption capacity of these compounds revealed a descending trend with increasing SFR (Table 2). Specifically, the adsorption capacities for BT decreased from 55 to 3.1 mg/g and 308 to 36 mg/g, respectively, as the SFR increased from 1 to 7 L/min. The adsorbent particle mass transfer and pore diffusional resistances usually control the adsorption process of various chemical species (Tsibranska *et al.*, 2000). Several studies (Davini, 2003; Das *et al.*, 2004; Gaur *et al.*, 2006) have reported that the FAC particle mass transfer resistance would be negligible for the adsorption rates of gas-phase compounds, such as VOC or SO<sub>2</sub>, on FAC. Therefore, the descending adsorption capacity observed for BT was ascribed to low pore diffusion of BT molecules under high SFR conditions (Gaur *et al.*, 2006).

#### Regeneration Efficiency

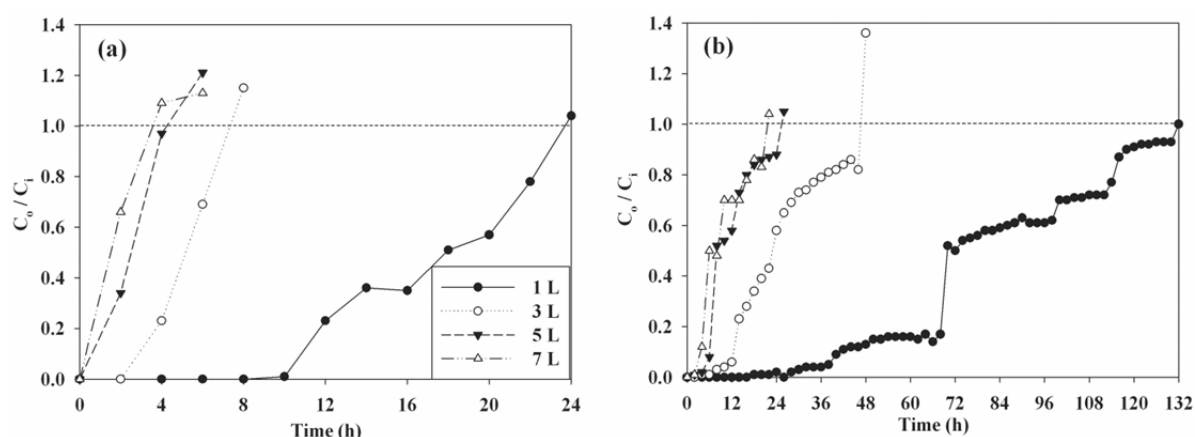
The regeneration efficiency of the FAC filter for the BT mixture was estimated under different regeneration conditions using Eq. (4). Table 3 lists the regeneration



**Fig. 3.** Adsorption breakthrough profile of fresh FAC filter for benzene (a) and toluene (b) according to initial concentration (0.1, 0.5, 1.0, and 2.0 ppm).

**Table 2.** Adsorption capacity (mg/g) of the FAC filter for benzene and toluene according to initial concentration and stream flow rate.

Compound	Initial concentration, ppm				Stream flow rate, L/min			
	0.1	0.5	1.0	2.0	1	3	5	7
Benzene	1.8	3.7	6.6	7.5	55	17	6.6	3.1
Toluene	8.9	28	43	53	308	94	43	36



**Fig. 4.** Adsorption breakthrough profile of fresh FAC filter for benzene (a) and toluene (b) according to stream flow rate (1, 3, 5, and 7 L/min).

**Table 3.** Regeneration efficiency and regeneration time of the FAC filter for benzene and toluene according to desorption temperature.

Compound	Temperature, °C	Regeneration efficiency, %	Regeneration time, h
Benzene	50	54	12
	100	57	4
	150	78	1
	200	88	0.3
Toluene	50	44	82
	100	53	62
	150	73	4
	200	82	1

times and regeneration efficiency of the FAC filter for the target compounds according to desorption temperature. The regeneration efficiencies for both target compounds increased as the desorption temperature increased from 50 to 200°C, while the regeneration times decreased. Das *et al.* (2004) reported a similar regeneration efficiency for selected VOCs on desorbing temperature under their specified experimental conditions. For the current study, the highest regeneration efficiencies of the FAC filter heated at 200°C for BT were 88 and 82%, respectively, while the lowest regeneration efficiencies of the FAC filter heated at 50°C for BT were 54 and 44%, respectively. In addition, the shortest regeneration times of the FAC filter heated at 200°C for BT were 0.3 and 1 h, respectively, while the longest regeneration times of the FAC filter heated at 50°C for BT were 12 and 82 h, respectively. The regeneration efficiency determined in this study was higher than that determined by Sidheswaran *et al.* (2012), who reported that regeneration efficiencies of FAC filters heated at 150°C with an air face velocity of 5.2 m/min for BT were 41–44% and 55–64%, respectively, while those for a lower air face velocity (0.52 m/min) were 31–45% and 37–52%, respectively. In the present study, the regeneration efficiencies of FAC filters heated at 150°C with an air face velocity of 1.7 m/min for BT were 68 and 73%, respectively. The difference in regeneration temperature between the two studies was primarily a result of the difference in FAC filter type and size, regeneration time, VOC mixture, and

air face velocity. For the FAC used in the present study, the surface area, carbon content, thickness, and diameter of the FAC were 1000 m<sup>2</sup>/g, 70 g/m<sup>2</sup>, 0.1 cm, and 6.2 cm, respectively, whereas for the FAC used in the previous study they were 1000–2000 m<sup>2</sup>/g, 120 g/m<sup>2</sup>, 0.04 cm, and 8.0 cm, respectively. The regeneration times of BT for the current study were 1 and 4 h, respectively, whereas for the previous study it was 2 h for both BT. The present study used a BT mixture, whereas the previous study used a mixture of BTX, 1-butanol, undecane, limonene, and formaldehyde.

Benzene exhibited a higher regeneration efficiency and shorter regeneration time than toluene (Table 3). Specifically, at a desorption temperature of 50°C, the regeneration efficiency was 1.2 times higher for benzene than toluene and the regeneration time for toluene was longer than that for benzene. Similar to the difference in adsorption properties of the two compounds described above, these differences in regeneration efficiency and time were primarily ascribed to the difference in water solubility and molecular weight between these compounds (Cal *et al.*, 1996; Daifullah and Girgis, 2003).

## CONCLUSIONS

The present study explored the feasibility of applying FAC for use as VOC filters in portable indoor air cleaning devices under simulated indoor conditions. This was achieved by determining the quantitative relationship between

adsorption characteristics (saturation and breakthrough times, and adsorption capacity) of FAC and indoor conditions on the basis of indoor humidity and indoor VOC concentration levels to provide information regarding optimal operational conditions of FAC. This study also confirmed that when FACs are applied as VOC filters in portable indoor air cleaning devices, their effectiveness depends on SFR entering into air cleaning devices and the indoor environmental conditions, such as RH and IC. Moreover, the regeneration efficiencies of the FAC filter for the BT mixture were quantitatively determined under different regeneration conditions to help select the optimum regeneration conditions for saturated FACs.

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