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Removal Characteristics of Ethyl Acetate and 2-Butanol by a Biofilter Packed with Jeju Scoria

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Abstract The removal characteristics of ethyl acetate and 2-butanol were investigated in a bench-scale down-flow biofilter packed with Jeju scoria medium. Various inlet concentrations and gas flow rates were tested. The adaptation times of microorganisms to the change of the influent concentration of ethyl acetate and 2-butanol gas were found to be about 3 days. At the inlet concentration of 300 ppmv and empty bed contact time (EBCT) of 15 sec, the removal efficiencies of the biofilter for ethyl acetate and 2-butanol were above 99.9%. The maximum removal capacity of the biofilter for ethyl acetate was 316–318 g/m^3/h and that for 2-butanol was 245–251 g/m^3/h. Overall, the removal capacity of the biofilter for ethyl acetate was 50–70 g/m^3/h larger than that for 2-butanol. During the operation of 65 days, the pressure drop through the biofilter column was maintained below 13 mmH_2O/m. Although the pH in the drain water decreased from 7.2 to 5.0, the pH drop did not affect the removal of the gases. From the above results, the biofilter using Jeju scoria as a packing material seemed to very effectively treat waste gases such as ethyl acetate and 2-butanol.

Key words: Biofiltration, biofilter, scoria, ethyl acetate, 2-butanol

Artificial chemical substances such as volatile organic compounds (VOCs) are emitted to the atmosphere in various forms and influence the environment according to their chemical reactivity in the air. Considerable quantities of VOCs are produced mainly from industrial sources such as printing and coating facilities, tape and paint manufacturing, and food waste plant [9]. In particular, VOCs such as ethanol, ethyl acetate, 2-butanol, chloroform, toluene, and benzene are produced from food waste plant. Releasing these substances to the ambient air may lead to an adverse environmental impact on air quality, thus endangering public health and welfare [18].

Biofiltration has been known to be an effective and economical technology among different air pollution control techniques to reduce VOCs and odors in air. Bifiltration has significant advantages over conventional technologies such as absorption, condensation, activated carbon adsorption, and thermal and catalytic oxidation. Biofiltration is particularly suited and cost-effective for the treatment of large volumes of waste gases containing low concentrations of VOCs. Furthermore, it is environmentally friendly because the contaminants are completely converted into non-hazardous final products such as carbon dioxide and water [6].

Biofilter systems for VOCs control are greatly affected by the selection of the packing media and microorganisms. Since the inoculation of specifically adapted microorganisms into biofilter media reduces the start-up period and may increase the biofilter performance [24], activated sludge, extracts from contaminated soils, and specially grown cultures have been used as inoculums [7, 16]. The ideal packing media are characterized by high specific surface area, low volumetric density, minimal pressure drop, and suitable surface for the attachment of microorganisms [26].

Various kinds of materials have been used as a packing material in biofilter's. Although natural organic media such as compost, peat, and wood chip have many advantages, including high physical adsorption capacity, excellent water holding capacity, favorable biological media for microbial growth and activity for their nutrient supply [19], they may be subjected to operation difficulties such as compaction and channeling [27], thus leading to lower process efficiency and short exchange cycle [10, 25]. To overcome such problems, some synthetic materials including ceramic, glass, polystyrene, perlite, activated carbon, sandstone, and clay are recently widely used as main constituents of the packing media or mixed with natural organic materials [2, 8, 10].

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The reliability of the biofilter process for the treatment of waste gas streams containing VOCs has been demonstrated in a number of experimental studies [22]. Researches have focused on the treatment of odors and VOCs such as benzene [13], toluene [11, 12], xylene [10], BTX [16], BTEX [19], ethanol [5], ethyl acetate [8, 18], 2-butanol [12], and odors [6, 12]. Although Lee *et al.* [14] and Cho *et al.* [4] used Jeju scoria as the packing media of the biofilter and they reported its good performance as biofilter media in the removal of H_2S and toluene, little research has been done to compare the removal of each of ethyl acetate and 2-butanol as a single gas by a biofilter packed with Jeju scoria.

The aim of this study was to investigate the removal characteristics of each single gas, ethyl acetate and 2-butanol, by a biofilter using Jeju scoria as a packing medium. These removal characteristics are very useful information for industrial application. Thus, the removal characteristics of changes of both influent concentration and influent flow rate of each gas were studied, and its maximum removal capacity was evaluated. Pressure drop and pH change in the biofilter bed during long-term operation were also investigated.

Materials and Methods

The schematic diagram of an experiment apparatus is shown in Fig. 1; it is composed of a biofilter, syringe pump, mixing chamber, and nutrient pump. The biofilter column was made using a circular acryl tube with inside diameter of 5 cm and column height of 75 cm. Jeju scoria (volcanic stone) was sieved to obtain a constant size (12–17 mm), and one liter was packed in the biofilter column. In this study, ethyl acetate and 2-butanol were selected as the test gases, and the experiment was performed separately. Each test gas flowed into the upper part of the biofilter bed, and then discharged at the bottom part of the biofilter. A spray nozzle for nutrient was installed at its upper part. A constant amount of each gas (in a liquid state) whose purity was over 99% was supplied by a syringe pump (Ken-a Mechatronics, KMSP-15MP) and the liquid was vaporized by air supplied from an air pump. Then, it was mixed in a mixing chamber to make a constant concentration. The gas with the constant concentration flowed into the upper part of the biofilter, and its inlet flow rate was controlled by a flow meter. The inlet concentration of each gas was first at a low value and then increased stepwise after the biofilter was stabilized. EBCT was changed

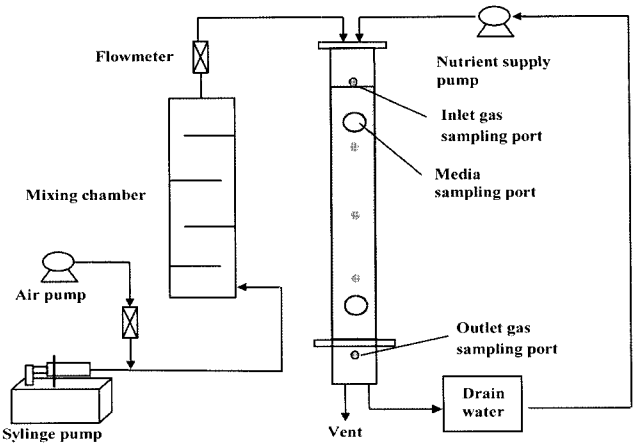

Fig. 1. Schematic diagram of an experimental apparatus.

to 60, 30, 15, 10, and 7.5 sec at constant inlet concentration of 300 ppmv. The experiment under each EBCT condition was performed for one week.

Return sludge, of which the cell concentration was 5,730 mg/l as MLVSS, obtained from a wastewater treatment plant was inoculated to scoria media through circulating by a nutrient-supplying pump for about 3 days. The final amount of microorganisms attached to the media was 0.457 g-MLVSS/g-dry material. Nutrients for the microorganisms used in this study were the mixture of KH_2PO_4 (2.5 g/l), K_2HPO_4 (2.5 g/l), NH_4Cl (2.5 g/l), $MgSO_4 \cdot 7H_2O$ (6.8 g/l), $CaCl_2 \cdot 2H_2O$ (0.5 g/l), $FeSO_4 \cdot 7H_2O$ (0.3 g/l), and KNO_3 (1.5 g/l), and 5 ml/min of the nutrients was supplied continuously.

The biofilter was operated at a constant temperature of 28–32°C. The pressure drop across the biofilter bed was measured with a manometer (Dwyer Instrument, MM. 400). The pH of drain water discharged from the biofilter was measured with a pH meter (Orion, 420A). Gas samples from gas sampling ports were collected into a gas bag, and their concentrations were then analyzed with a gas chromatograph (HP 5890 series II, U.S.A.) with flame ionization detector (FID). The capillary column used was an 'HP-5' (30 ml, 0.32 mm ID), and the flow rate of N_2 gas (99.99% purity) as the carrier gas was 40 ml/min. Temperatures of injection, oven, and detector were 70, 150, and 250°C, respectively. The detection limit by the gas chromatography for ethyl acetate and 2-butanol was below 1 ppmv.

The scoria used as a packing media in this experiment is a volcanic stone produced by fusion of porous volcanic materials, volcanic sand, and other volcanic ashes spouted from volcanos and it is widely distributed in Jeju Island

Table 1. Chemical composition of Jeju scoria (wt%).

SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	TiO_2	MnO	P_2O_5	LOI#
47.48	16.93	13.00	7.45	4.37	1.63	3.54	2.65	0.16	0.64	1.65

LOI#: Loss on ignition.

[14]. The chemical composition of the scoria is tabulated in Table 1. The scoria was mainly composed of coarse grains whose particle sizes were of several centimeters, and its main components (about 75%) were SiO_2, Al_2O_3, and Fe_2O_3. It had a stable chemical composition, so that the content of organic impurities was relatively low. Since the scoria had high porosity, its water content was about 31.7%. When the scoria in the range of 12–17 mm was packed in the biofilter, the apparent density and the packing density were 2 and 0.8 g/cm^3, respectively, with a void volume ratio of 0.5.

RESULTS AND DISCUSSION

Removal Characteristics on Change of Influent Concentration

The concentration of VOCs produced at a real site generally varies widely. The performance of a biofilter to remove VOCs depends on both the influent concentration and the type of substance used. Furthermore, in order to obtain good and stable removal capacities, microorganisms attached onto the media need acclimation time to adapt to the new environment during the start up of biofilter operation, after changes in operational parameters such as an increase of inlet concentration, shock loads, or reduction of empty bed contact time (EBCT) [20]. Acclimation time influences the overall performance of the biofilter system, and it depends on the flow types of gas, characteristic of gaseous pollutant, or characteristic of medium. Morales *et al.* [21] reported long acclimation periods, ranging from several weeks to months. However, the start-up time for systems inoculated with well-adapted cells varies from one day to a week.

To investigate the removal performance and acclimation time of microorganisms as the function of changes in the inlet concentration of each of ethyl acetate and 2-butanol, the biofilter was first operated at EBCT of 30 sec. The inlet concentrations of ethyl acetate and 2-butanol were increased stepwise from 100 ppmv to 1,000 ppmv and to 1,100 ppmv, respectively. Figure 2 shows the inlet and outlet concentration profiles of each of ethyl acetate and 2-butanol under different inlet concentrations as a function of operation time, and Fig. 3 shows the removal efficiency for each of ethyl acetate and 2-butanol as a function of change of inlet concentration. As seen in Fig. 2, the microorganisms seemed to be adapted to the change of the inlet concentration within 3–4 days. The acclimation time observed for each of ethyl acetate and 2-butanol in the present study was shorter than the 5–6 days in the toluene removal by a biofilter using Jeju scoria [14] and the 9 days in the toluene removal by a biofilter using compost [20], indicating that ethyl acetate and 2-butanol are more biodegradable than toluene.

As seen in Fig. 2, a high outlet concentration of ethyl acetate was detected after 1 day of the biofilter operation, and it was not detected after then. In the experiment of 2-butanol, it was detected for the first 3 days of the biofilter operation, and it was completely removed after that. These results indicate that the adaptation time of microorganisms for each gas was somewhat different and the biofilter was stabilized after the initial adaptation of microorganisms was completed. When the ethyl acetate concentration was increased stepwise up to 500 ppmv, it was not detected at the outlet biofilter under a steady state. When the inlet concentration of ethyl acetate was increased to 640 ppmv, its initial outlet concentration was 100 ppmv; however, about average 4 ppmv was detected after complete adaptation. When the inlet concentrations were further increased to 750 and 1,000 ppmv, the average concentrations of about 100 and 280 ppmv were detected, respectively. For the inlet concentrations of 650, 750, and 1,000 ppmv, the average

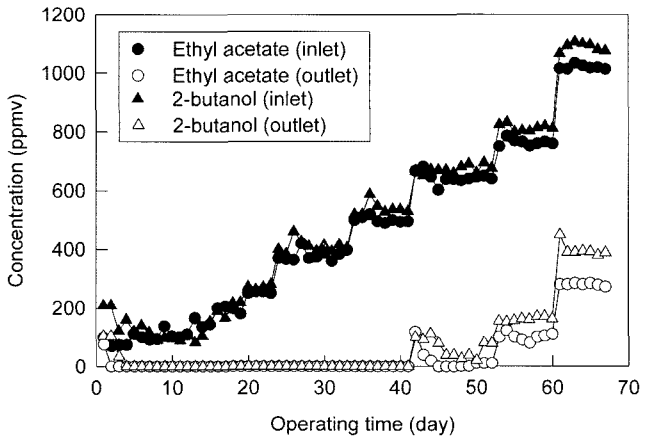

Fig. 2. Inlet and outlet concentration profiles of ethyl acetate and 2-butanol under different inlet concentrations at EBCT of 30 sec.

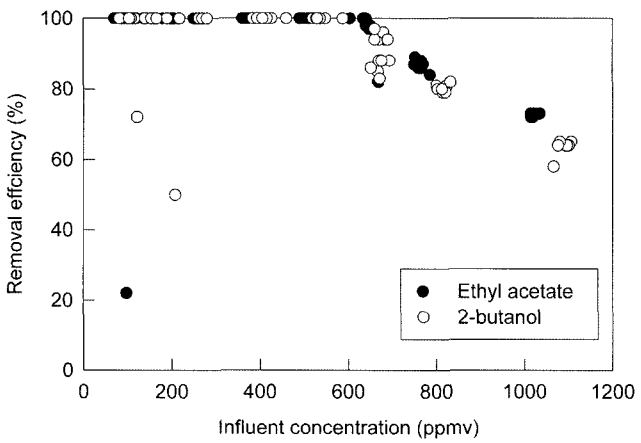

Fig. 3. Removal efficiency of ethyl acetate and 2-butanol under different inlet concentrations at EBCT of 30 sec.

removal efficiencies were calculated to be 99%, 87%, and 73%, respectively (Fig. 3).

In the case of 2-butanol, it was not detected at the outlet biofilter when the inlet concentration was stepwise increased up to 550 ppmv. At higher inlet concentrations of 2-butanol, the average outlet concentrations were found to be about 50, 160, and 390 ppmv for the inlet concentrations of 700, 800, and 1,100 ppmv, respectively, and their average removal efficiencies were 93%, 80%, and 63%, respectively. As seen in Fig. 3, the maximum inlet concentration for each of ethyl acetate and 2-butanol was about 650 ppmv, in which each gas was completely removed by the biofilter. At higher inlet concentrations, the average removal efficiency of ethyl acetate was 5–10% higher than that of 2-butanol. This means that the removal capacity of the biofilter for 2-butanol was somewhat lower than that of ethyl acetate.

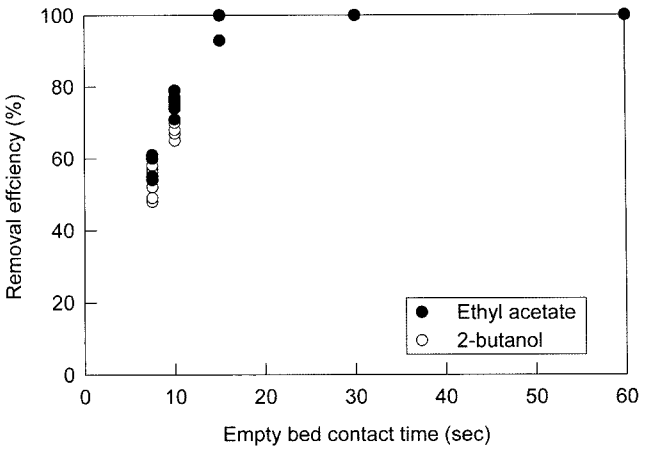

Fig. 5. Removal efficiency of ethyl acetate and 2-butanol under different EBCT at 300 ppmv of the inlet concentration.

Removal Characteristics on the Change of EBCT

When the flow rate of inlet gas is increased, the time for the gas to contact with the surface of packing media (EBCT) is shortened, leading to the reduction of removal efficiency of the biofilter. Therefore, to find the optimum flow rate of inlet gas is important in biofilter operation. The removal characteristics of each of ethyl acetate and 2-butanol as a function of change of the influent flow rate (namely, change of EBCT) were investigated, and the results are shown in Figs. 4 and 5. Figure 4 shows the inlet and outlet concentration profiles of ethyl acetate and 2-butanol under different EBCT, in which the inlet concentration of each gas was fixed at 300 ppmv. Each of ethyl acetate and 2-butanol was completely removed under 30 and 60 sec of EBCT. When EBCT was reduced to 15 sec from 30 sec, about 20 ppmv of each gas was not degraded at the first day, and it was then completely removed by the biofilter

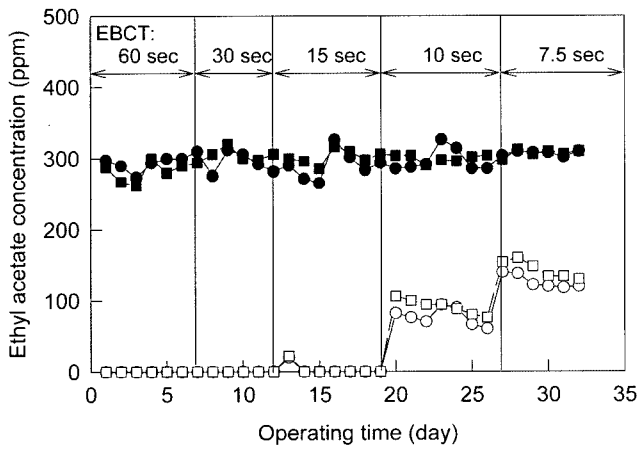

Fig. 4. Inlet and outlet concentration profiles of ethyl acetate and 2-butanol under different EBCT at 300 ppmv of the inlet concentration.
Circle indicates ethyl acetate and rectangular indicates 2-butanol, closed symbol indicates inlet and open symbol indicates outlet.

after that. It is quite likely that this was due to the shock of inlet load coming from the double inlet loading rate. The ethyl acetate and the 2-butanol not degraded by the biofilter were detected at the outlet of the biofilter, and the average outlet concentrations of ethyl acetate and 2-butanol were 76 and 120 ppmv, and 86 and 137 ppmv for 10 and 7.5 sec of EBCT, respectively. The average removal efficiencies of ethyl acetate and 2-butanol were calculated to be 75% and 71%, and 61% and 56% for 10 and 7.5 sec of EBCT, respectively (Fig. 5). In all ranges of EBCT, the removal efficiency of the biofilter for ethyl acetate was higher than that for 2-butanol. This result was similar to that obtained in the experiment of removal characteristics on change of the inlet concentration in the previous section.

The above results indicate that the biofilter with scoria as a packing media gives stable efficiency because of fast adaptation for stepwise reduction of EBCT, compared with most commercial biofilters that are operated within the EBCT range of 50–150 sec. The biofilter can effectively treat waste gases such as ethyl acetate and 2-butanol at high flow rates, and this will result in a size reduction of biofilter equipment.

Evaluation of Maximum Removal Capacity

Maximum removal capacity is the capacity that the system can stand without inhibition of microbial activity, and it depends on operation conditions of the system and types of packed media used and is an important parameter in the design of a real biofilter system. The removal capacity of the biofilter for each of ethyl acetate and 2-butanol as a function of influent loading rate under different inlet concentrations is shown in Fig. 6: The solid line indicates the theoretical 100% removal. The removal efficiencies of ethyl acetate and 2-butanol increased with increase of influent loading rate. Removal efficiencies of more than 99.9% were achieved with the inlet loading rate of ethyl acetate

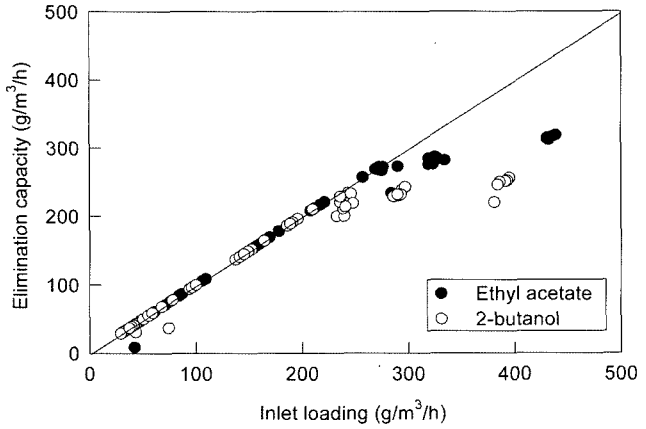

Fig. 6. Relationship between influent loading rate and removal capacity of the biofilter for each of ethyl acetate and 2-butanol under different influent concentrations.

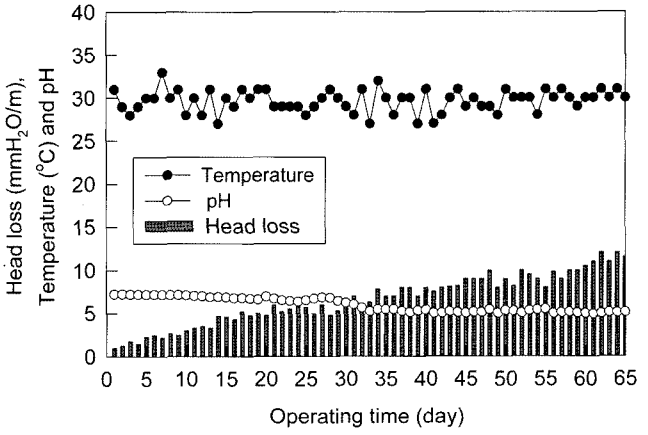

Fig. 8. Variations of head loss, pH, and temperature during operation times.

and 2-butanol below 260 and 210 g/m^3/h, respectively. When the maximal influent loading rates of ethyl acetate and 2-butanol were 434 and 390 g/m^3/h, the maximum removal capacities were determined to be 316 and 251 g/m^3/h, respectively. Therefore, the maximum removal capacity of ethyl acetate was about 50 g/m^3/h higher than that of 2-butanol at the high inlet loading rate.

Figure 7 shows the relationship between the inlet loading rate and the removal capacity as a function of change of EBCT. The removal capacity increased with increase of the inlet loading rate. Complete removals were achieved with the inlet loading rate of ethyl acetate and 2-butanol below 270 and 210 g/m^3/h, respectively. When the influent loading rates of ethyl acetate and 2-butanol were maximally 522 and 440 g/m^3/h, the maximum removal capacities were determined to be 318 and 245 g/m^3/h, respectively. Thus, the maximum removal capacity of ethyl acetate was about 70 g/m^3/h higher than that of 2-butanol at a high inlet loading rate. Overall, the removal capacity of the biofilter for ethyl acetate was 50–70 g/m^3/h higher than that for 2-butanol. At the same inlet loading rate, the removal capacity of the biofilter for gas at high concentration with relatively low EBCT was somewhat higher than that for gas at low concentration with high EBCT. This means that both ethyl acetate and 2-butanol gas are dissolved and easily diffused into biofilm, thus resulting in a fast overall reaction rate.

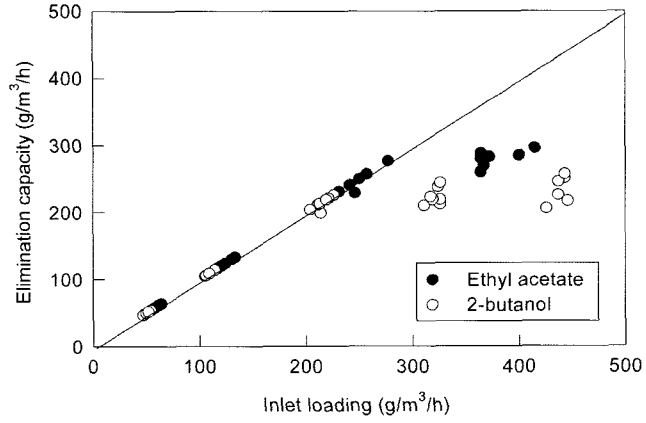

Fig. 7. Relationship between the influent loading rate and the removal capacity of the biofilter for each of ethyl acetate and 2-butanol under different EBCT.

In this study, the maximum removal capacities of the biofilter with Jeju scoria as the packing media for ethyl acetate and 2-butanol were 316–318 g/m^3/h and 245–251 g/m^3/h, respectively. The maximum removal capacity of ethyl acetate was higher than the 170–200 g/m^3/h rate shown by a biofilter packed with the mixed media of compost and wood chip [8], and the 79–96 g/m^3/h rate by a biofilter packed with the mixed media of compost and polystyrene [22]. Baltzis and Androutsopoulou [2] reported that the maximum removal capacity of 2-butanol by a biofilter packed with the mixed media of peat and perlite was 24–26–g/m^3/h, and Sabo [23] reported that the maximum removal capacity of 2-butanol by a biofilter packed with the mixed media of compost and clay was 70 g/m^3/h. Thus, the maximum removal capacity of 2-butanol obtained in this study was 2–3 times higher than those of these two mentioned studies. Consequently, the Jeju scoria seems to be appropriate as a packed medium for a biofilter, since it showed an excellent removal capacity for both ethyl acetate and 2-butanol.

pH and Pressure Drop

pH is an important factor that affects the performance of any biofilter, because microorganisms generally cannot tolerate extreme pH conditions. In addition, pH of an environment influences the dissociation and solubility of many molecules that indirectly influence microorganisms [1]. Optimal pH

levels for most microorganisms involved in a biofilter are usually between 6 to 8, and a rapid change in pH can damage most microorganisms or decrease the activity of microorganisms [22]. Figure 8 shows the changes of pH in drain water, pressure drop, and temperature in the biofilter during the biofilter operation for the treatment of ethyl acetate and 2-butanol gases. The pH changed from 6.21 to 7.23 during the initial 30-day operation, and then decreased to 4.98, concordant with the result of Liu *et al.* [27] in which pH was lowered from 6.71 to 5.5 after a 3-month operation, when ethyl acetate was treated by a biofilter using compost as the packing media. The reasons of why the pH decreased during continuous operation of the biofilter could be found in the following. Firstly, the influent loading rate during operation was increased owing to increase of the influent concentration that was increased from 100 ppmv to 200, 250, 400, 500, 650, 750, and 1,000 ppmv. Secondly, pH decreased during long-term operation, because some acids could be produced by microorganisms [3]. However, in this study, the reduction of removal efficiency or activity of microorganism was not observed as pH decreased. Whenever pH continuously decreases in a biofilter system, a buffer solution should be added in order to maintain the pH in a favorable range for the microorganisms.

Pressure drop across a biofilter generally depends on the property of flow, the flow rate of gas, and the nature of a packing material. Since the resistance of flow in a biofilter bed causes reduction of removal capacity and energy consumption as well, it is desirable to maintain the pressure drop to as low as possible. Controlling the amount of biomass is important and, sometimes, back washing is required [28]. Generally, pressure drop occurs when the biofilm gets thicker because of growth of microorganisms attached on the packing media. Increases in pressure drop have been reported in a number of studies. Leson *et al.* [15] reported that pressure drop increased from 4 to 25 cmH_2O/m over 6 months, and Wolstenholme and Finger [29] also reported a high pressure drop of 10–15 cmH_2O/m in the treatment of VOCs by an open biofilter system. In this study, however, the pressure drop was maintained well below 13 mmH_2O/m during 67 days of operation, and any reduction of removal efficiency or clogging of packing media in the biofilter bed was not observed (Fig. 8).

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