

Absorption of a volatile organic compound by a jet loop reactor with circulation of a surfactant solution: Performance evaluation

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Abstract

Biofiltration shows high efficiency for the removal of industrial waste gases and reliable operational stability at low investment and operating cost, especially when the VOC concentration is low, such as 100 ppmv ($\mu\text{L L}^{-1}$) or less. However, it has been reported that the abrupt change in VOC concentrations leads to the failure of the biofilter. Hence, the pretreatment of waste gases is necessary to ensure the stable operation of the biofilter. The objective of this study is to develop a jet loop reactor (JLR) with circulation of a surfactant solution to lower the concentration of VOCs, especially hydrophobic VOCs. Toluene and Tween 81 were used as a model industrial waste gas and a surfactant, respectively. Among several non-ionic surfactants tested, Tween 81 showed the most rapid dissolution of toluene. When a JLR is replaced with fresh Tween 81 solution (0.3% w/v) every hour, it successfully absorbed for 48 h over 90% of the toluene in an inlet gas containing toluene at 1000 ppmv ($\mu\text{L L}^{-1}$) or less. Therefore, JLR with circulation of a surfactant solution is believed to ensure the stable operation of the biofilter even with the unexpected increase in the VOC concentrations.

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1. Introduction

Air emissions from a variety of industries contain volatile organic compounds (VOCs) and have been subject to increasingly stringent environmental regulations over the last two decades [1]. For example, benzene, toluene, ethyl benzene, and xylene (BTEX), which are often detected at significant levels in industrial exhaust gases, are regarded as predominant pollutants by the Environmental Protection Agency (EPA).

A number of methods have been developed to remove VOCs in the gas phase. Among them, biofiltration, which is a technology based on the biological oxidation of VOCs using microorganisms, is widely used when the concentration of VOC is relatively low [2,3]. For biofiltration, microorganisms are immobilized on porous solid particles such as peat, compost, and wood chips and these immobilized particles are packed in

a column called a biofilter [4–10]. Compared to non-biological techniques such as incineration, condensation, scrubbing, and adsorption, the major advantage of biofiltration is that the pollutants can be converted into harmless oxidation products. Moreover, biofiltration shows high removal efficiency and reliable operational stability at low investment and operating cost, especially when the VOC concentration is low, such as 100 ppmv ($\mu\text{L L}^{-1}$) or less. However, a stable biofilter performance relies on constant loading [11]. In practice, most industrial waste gases have variable flow rates and abrupt change in VOC concentrations leading to the failure of biofilter [12–17]. The abrupt increase in the VOC concentration from 100 to 1000 ppmv for example, decreases the number of viable microorganisms. Hence, the pretreatment of industrial waste gases to lower VOC concentrations prior to biofiltration is necessary to guarantee the stable operation of the biofilter [4].

There have been numerous studies of activated carbon bed for dampening fluctuations in VOC loading [18–22]. However, the costs of activated carbon and the recovery process are inhibiting the combined use of the biofilter and the activated carbon

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bed. A reactor for VOC absorption could be another candidate for the pretreatment of industrial waste gases. When hydrophobic VOCs such as BTEX are to be removed from industrial exhaust gases, an absorption reactor filled with pure water is not efficient due to the low solubility of hydrophobic VOCs in water. Surfactants composed of a hydrophilic head group and a hydrophobic tail can be well solubilized in water forming a micelle. The hydrophilic groups are headed to water while the hydrophobic tails aggregates in the middle of the micelle. As hydrophobic VOCs are captured in the hydrophobic tails due to the hydrophobic interaction, their solubility in the surfactant solution is much larger than that in pure water. Hence, an absorption reactor filled with the surfactant solution is more efficient than one with pure water [23]. When using a biodegradable surfactant for the absorption of VOCs, the surfactant solution saturated with VOCs can be treated in a wastewater treatment facility. The surfactant can be reused by ultrafiltration [24–28]. The more plausible method for regeneration of the surfactant is simply to meter clean air through the jet loop reactor with another biofilter to treat that VOC-laden air. If the VOC concentration in the inlet waste gas decreases back to a normal level such as below 100 ppmv ($\mu\text{L L}^{-1}$), the gas can be used to strip VOCs from the surfactant solution.

In the field of biochemical engineering, various types of reactors, including an air-lift loop reactor, a bubble column reactor, a stirred tank reactor, and a jet loop reactor, have been studied for oxygen absorption to facilitate the supply of dissolved oxygen to aerobic microorganisms [29]. A jet loop reactor (JLR) contains a central draft tube inside the reactor. A two-fluid (liquid and gas) nozzle is located at the top or bottom of the reactor in a structure of two concentric cylinders. The gas delivered through one cylinder is dispersed by the liquid jet stream delivered through the other cylinder. The liquid and/or the gas can be circulated back to the reactor to increase the concentration of the gas species in the liquid phase. For dissolving oxygen in the water, a JLR was found to be superior to other reactors in terms of the oxygen transfer rate per unit power input [30]. Since JLRs have another advantage of a small area requirement for installation, they have become increasingly important in chemical, biochemical, and even environmental process industries [31–36].

In this study, a JLR with circulation of a surfactant solution was used to improve the transfer of a hydrophobic VOC from the gas phase to the water phase. Toluene was used as a model hydrophobic VOC. A surfactant showing a higher dissolution rate and greater solubility of hydrophobic VOCs with less foaming should be selected. Surfactants with a hydrophile–lipophile balance (HLB) value between 8 and 18 are generally applied to

make oil-in-water emulsions [37]. Hence, non-ionic surfactants with an HLB of 8–18 are appropriate for this purpose [37].

Non-ionic surfactants are largely divided into polyoxyethylene-based and polyol-based surfactants. Among the numerous polyoxyethylene-based surfactants, LA5 and LA7 were used in this study. Among the numerous polyol-based surfactants, Span 20, Tween 20, and Tween 81, which are an ester and ethoxylated esters of fatty acids with sorbitan, respectively, were used. The purposes of this study were first to determine the optimum surfactant showing the most rapid dissolution of a hydrophobic VOC (toluene) and then to investigate the influence of several operational variables of the JLR, including the gas/liquid flow rate and the surfactant concentration, on the efficiency of toluene removal. As 1000 ppmv is the concentration of VOCs that usually causes biofilter malfunction in the treatment of waste gases (communication with industrialists), 500–1500 ppmv was chosen as the range of toluene concentrations in the inlet gas. A down-flow JLR, in which the gas was introduced from the top into the surfactant solution, was used to increase the gas-phase residence time.

2. Materials and methods

2.1. Dissolution of toluene in a surfactant solution

The physical properties of surfactants used are summarized in Table 1. LA5 and LA7 were purchased from Korea Polyol Co. Span 20, Tween 20, and Tween 81 were all purchased from Sigma–Aldrich. Into a vial with a magnetic bar, 30 mL of a surfactant solution (1% w/v) and 20 mL of toluene (HPLC grade) were added. After 30 min of mixing at room temperature, the mixture was centrifuged at 2000 rpm for 20 min to separate undissolved toluene. By measuring the toluene concentration in the aqueous phase, the dissolution rate of toluene in the surfactant solution was determined.

The equilibrium solubility of toluene in surfactant solutions was measured in a constant-temperature water bath in which the temperature was controlled to an accuracy of ± 0.1 °C. To 20 mL of surfactant solution at various concentrations (0.1%, 0.5%, 1.0%, 1.5%, and 2.0% w/v), an excess amount (~ 20 mL) of toluene was added to ensure maximum solubility. Vials containing the mixture were then shaken for 48 h and subsequently centrifuged at 2000 rpm for 20 min to separate undissolved toluene.

The amount of toluene dissolved in the surfactant solution was analyzed by high-performance liquid chromatography (Waters Model 510 HPLC) using a differential refractometer.

Table 1
Physical properties of surfactants used in this study

Name	Chemical name	HLB [38–40]	CMC (mg L^{-1}) [38,41,42]	pH of 1% (w/v) solution
LA5	Pentaethylene glycol monododecyl ether	10.5 [38]	25.2 [38]	5.0–7.5 [38]
LA7	Heptaethylene glycol monododecyl ether	12.2 [38]	36.1 [38]	5.0–7.5 [38]
Span 20	Sorbitan monolaurate	8.6 [39,40]	21.1 [41]	4.8–4.9 ^a
Tween 20	Polyoxyethylene sorbitan monolaurate	16.7 [40]	60.0 [42]	5–7 ^a
Tween 81	Polyoxyethylene sorbitan monooleate	10.0 [39,40]	33.7 [42]	6.3–6.5 ^a

^a Measured in this study.

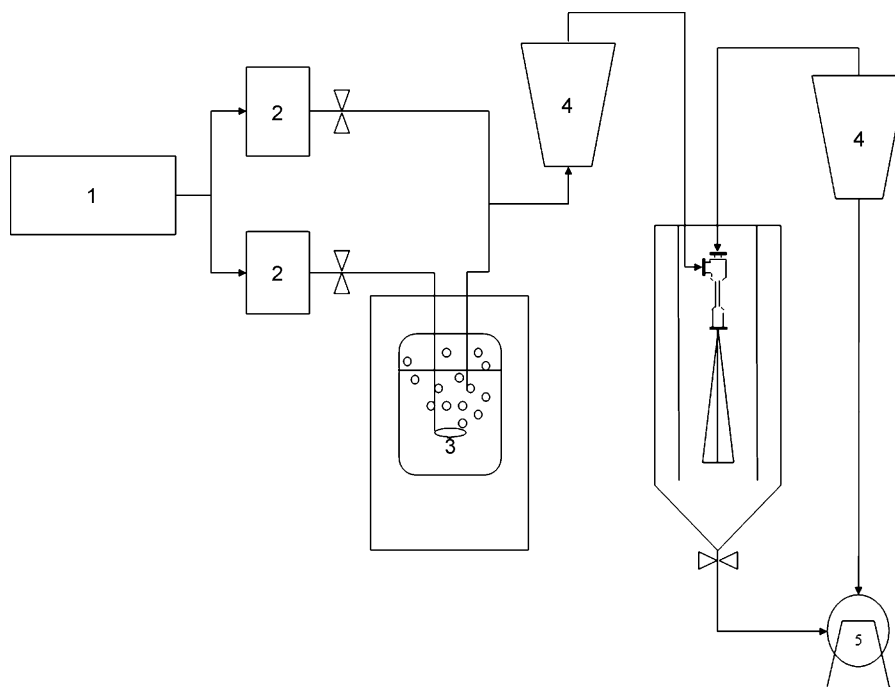


Fig. 1. Schematic diagram of the overall jet loop reactor system: (1) N₂ gas cylinder; (2) MFC; (3) VOC chamber; (4) flow meter; (5) pump.

Separation was performed on an EnviroSep-PP column (Phenomenex, 4.6 mm × 125 mm) at 20 °C. The samples were eluted under isocratic conditions with 65% acetonitrile (HPLC grade) and 35% deionized water at a constant flow rate of 1 mL min⁻¹. The viscosity of the surfactant solutions was measured using a Brookfield viscometer.

2.2. Experimental set-up for the jet loop reactor

A schematic diagram of the overall JLR system is shown in Fig. 1. It consists of a toluene gas generator and the JLR. In the toluene gas generator, a nitrogen gas flow was divided into two streams: one passed through a chamber filled with toluene liquid, and the other stream by-passed the toluene chamber and diluted the former stream to adjust the toluene concentration to a desired level. Gas flow rates were controlled using mass flow controllers (MFCs). The inner diameter and the height of the acrylic JLR were 0.25 and 1.0 m, respectively. A draft tube was located axially in the center of the reactor column. An ejector (Anico Co., Seoul, Korea) made of SUS (stainless steel) was fixed at the top of the tube and the surfactant solution containing mineral oil at 40 ppm was forced to circulate to the nozzle by means of a rotary vane pump. A detailed schematic diagram of the ejector is shown in Fig. 2. The maximum head and flow rate of the pump were 138 m and 14 L min⁻¹, respectively. The volume of liquid in the JLR was 50 L. The liquid flowed inside the draft tube and consequently entered the annular space between the inner and outer tubes. The flow rates of the Tween 81 solution and the toluene-containing nitrogen gas tested are summarized in Table 2. Due to the venturi effect at the ejector, the high gas flow rate could be obtained without using a gas-pump (see Fig. 1) and the gas flow rate was proportional to the liquid flow rate. All the experiments were performed at room temperature.

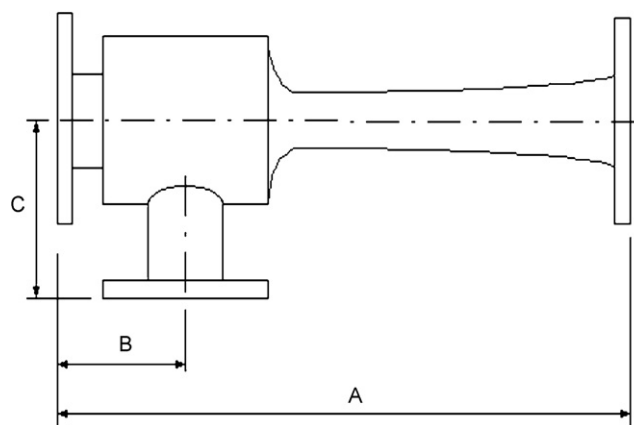


Fig. 2. Detailed schematic diagram of the ejector. The dimensions of the ejector are A = 180 mm; B = 70 mm; and C = 70 mm.

Toluene in the gas phase was analyzed by gas chromatography (HP6890, Hewlett Packard) using a flame ionization detector. For a GC-column and a carrier gas, an HP-5 column and nitrogen gas were used, respectively. Both injector and detector temperature were 250 °C. Based on the standard curve for toluene prepared by GC, Micro FID (Photovac Inc., USA) was

Table 2
Flow rates of Tween 81 solution and toluene-containing gas

Liquid flow rate (L min ⁻¹)	Gas flow rate (L min ⁻¹)
10.0	8.5
11.0	9.5
12.0	10.5
13.0	11.5
14.0	12.0

used to measure toluene concentrations in the outlet gas stream. Measurements were performed semi-continuously with a time interval of 1–5 min. From the graphical integration of the data, the removal efficiency of toluene was calculated.

3. Results and discussion

3.1. Dissolution rate of toluene in surfactant solutions

The concentrations of toluene in various surfactant solutions after 30-min dissolution experiments are shown in Fig. 3. Tween 81 exhibited the most rapid dissolution of toluene. Hence, Tween 81 was used for further experiments. For reference, toluene solubility in water is 0.62 g L^{-1} (6.74 mM) at 25°C .

3.2. Solubility of toluene in Tween 81 solution and viscosity of the Tween 81 solution

The solubility of toluene in Tween 81 solution at various temperatures is summarized in Fig. 4. As the HLB and critical micelle concentration (CMC) of Tween 81 were 10.0 and 33.7 mg L^{-1} , respectively, toluene was dissolved in the Tween 81 solution tested via an oil-in-water emulsion. Fig. 5 shows

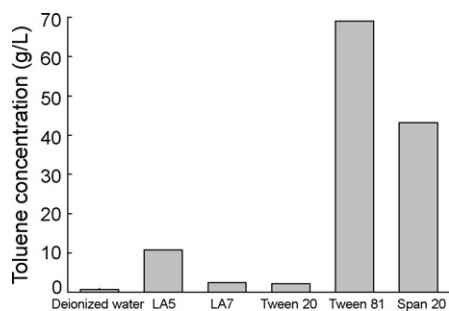


Fig. 3. Dissolution rate of toluene in various surfactant solutions measured as toluene concentration in the aqueous phase after mixing toluene and surfactant solutions for 30 min at 298 K.

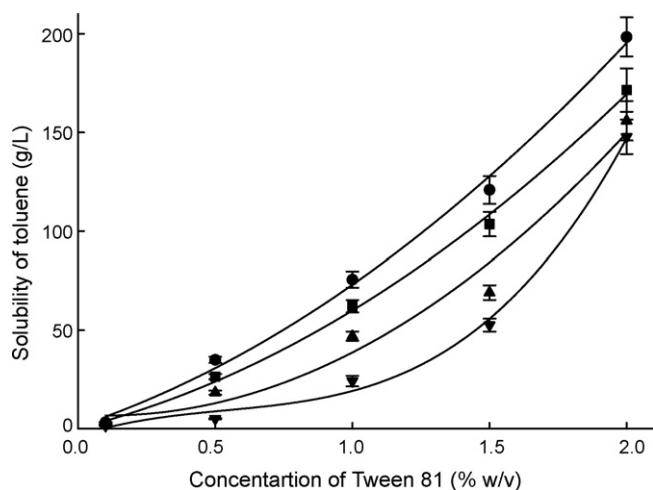


Fig. 4. Solubility of toluene in Tween 81 solutions: 298 K (●); 303 K (■); 308 K (▲); and 313 K (▼). Each data point and error bar denote the average value of duplicate measurements and the deviation, respectively.

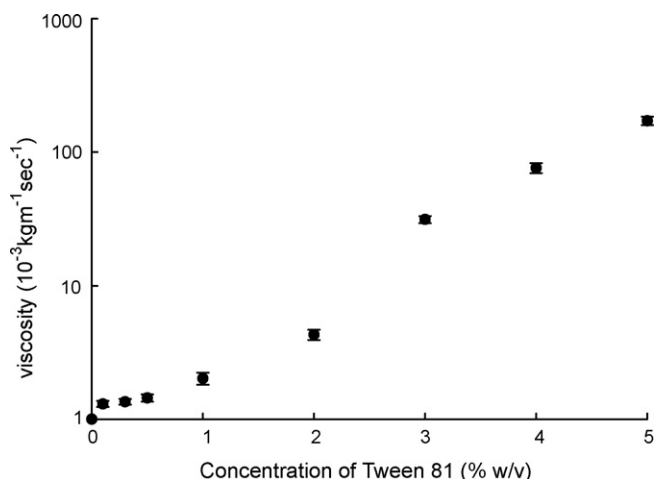


Fig. 5. Viscosity of the Tween 81 solution at 298 K. Each data point and error bar denote the average value of duplicate measurements and the deviation, respectively.

the viscosity of the Tween 81 solutions as a function of concentration at room temperature. As expected, the viscosity of the surfactant solution increased with the increase in the surfactant concentration.

3.3. Effect of toluene concentration in the inlet gas on the removal efficiency

Toluene concentrations in the outlet gas of the JLR system were measured at various toluene concentrations in the inlet gas. The liquid flow rate, the Tween 81 concentration, and the ratio of the draft tube diameter to the column diameter were 10 L min^{-1} , 0.3% (w/v), and 0.48, respectively. The flow rate of gas generated by the liquid jet stream was 8.5 L min^{-1} . Every hour, the used Tween 81 solution in the JLR was replaced with the fresh

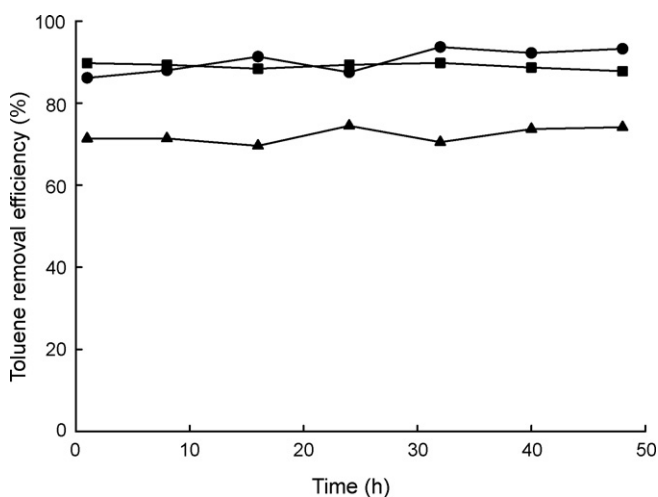


Fig. 6. Toluene removal efficiency of the jet loop reactor as a function of the toluene concentration in the inlet gas: 500 ppmv (●); 1000 ppmv (■); and 1500 ppmv (▲). The Tween 81 concentration, the liquid flow rate, and the ratio of the draft tube diameter to the column diameter were 0.3% (w/v), 10 L min^{-1} , and 0.48, respectively. The used surfactant solution in the JLR was replaced with the fresh one every hour.

solution. The toluene removal efficiency, which was defined in this study as the percentage of toluene absorbed by the surfactant solution in the JLR, was approximately 70% when the concentration of toluene in the inlet gas was 1500 ppmv (Fig. 6). However, when the concentration of toluene in the inlet gas was 1000 ppmv or less, over 90% of the toluene in the inlet gas was absorbed and the toluene concentration in the outlet gas was lower than 100 ppmv, which can be easily treated by biofiltration. As 1000 ppmv is the VOC concentration that usually causes biofilter malfunction in the treatment of waste gases, a JLR with circulation of a surfactant solution connected to the biofilter could be applied for the stable treatment of industrial waste gases, especially hydrophobic VOCs.

3.4. Effect of JLR operational variables and design parameters on the toluene removal efficiency

The liquid flow rate and the concentration of Tween 81 were chosen as the JLR operational variables. The ratio of the draft tube diameter to the column diameter was chosen as the design parameter of the JLR. The influence of the operational vari-

ables and design parameter on the toluene removal efficiency was investigated by running the JLR for 1 h while maintaining the toluene concentration in the inlet gas at 1000 ppmv. The experimental results are shown in Fig. 7. At the same Tween 81 concentration and the same ratio of the draft tube diameter to the column diameter, the toluene removal efficiency increased as the liquid flow rate increased. The increase in the gas absorption using the higher liquid flow rate has been already reported in many studies, especially oxygen absorption using a bubble column reactor or a jet loop reactor [30]. At the same liquid flow rate and the same ratio of the draft tube diameter to the column diameter, the toluene removal efficiency does not seem to be affected by Tween 81 concentrations. As the jet loop reactor was operated for a relatively short time (1 h), the amount of toluene absorbed was less than its saturation point, which was in equilibrium with the inlet gas and was greatly affected by the Tween 81 concentration. Also the viscosity of Tween 81 solution increased as the Tween 81 concentration increased (see Fig. 5). The rate of gas–liquid mass transfer is known to be lower in the more viscous solution [30]. Hence, the toluene removal efficiency was not apparently affected by Tween 81 concentrations.

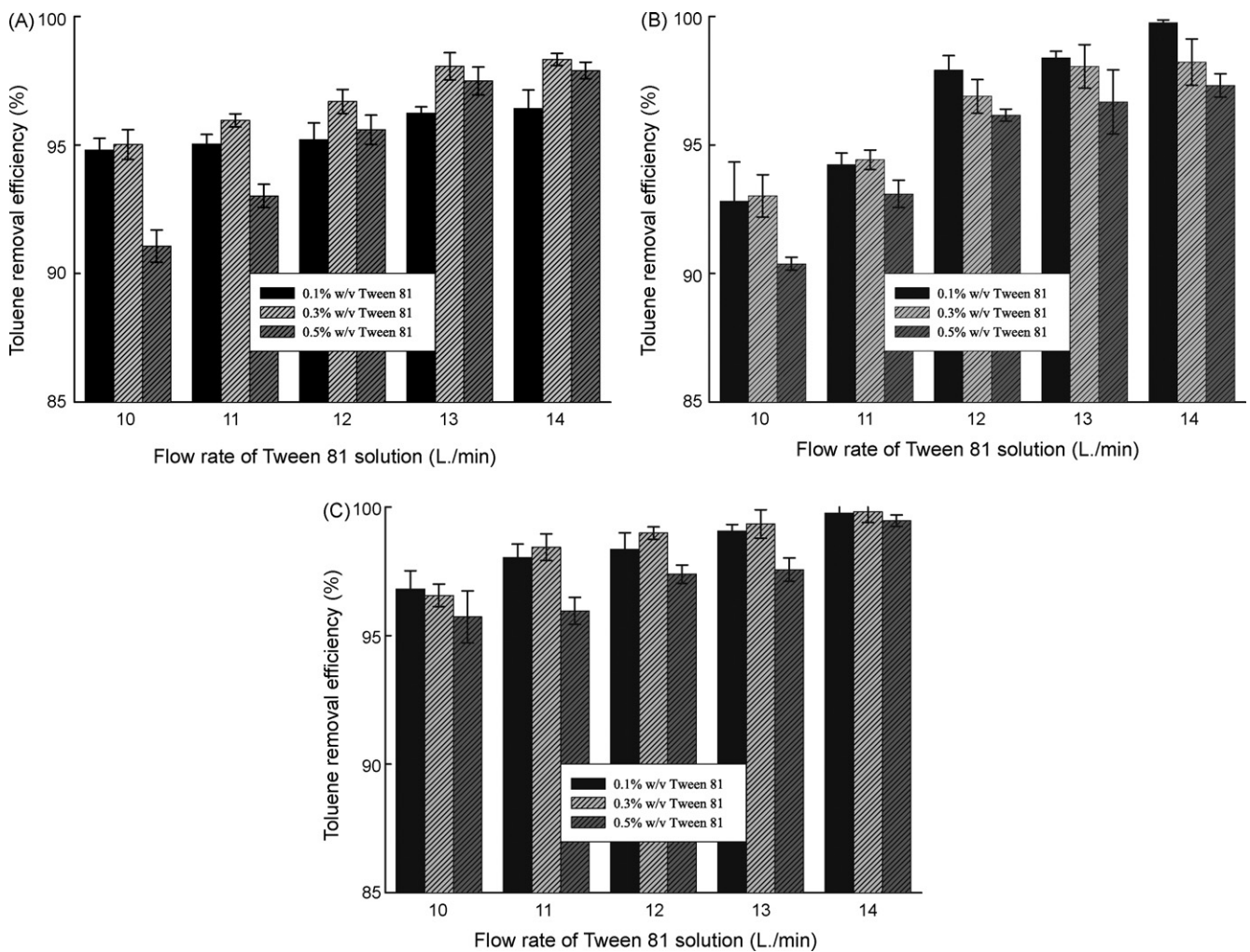


Fig. 7. Influence of the liquid flow rate and Tween 81 concentration on the toluene removal efficiency when the ratio of the draft tube diameter to the column diameter was: (A) 0.36; (B) 0.48; and (C) 0.60. Data shown in the plots were obtained for 1-h running of the jet loop reactor with an inlet gas containing toluene at 1000 ppmv. Each data and error bars denote the average value and the deviation, respectively.

In addition, the ratio of the draft tube diameter to the column diameter did not affect the toluene removal efficiency.

4. Conclusions

For stable operation of a biofilter regardless of fluctuations in VOC concentrations, especially hydrophobic VOCs such as toluene, a jet loop reactor with circulation of a surfactant solution is proposed. Among several non-ionic surfactants tested, Tween 81 showed the most rapid dissolution of toluene and was used in the JLR operation. For 48 h, JLR with 0.3% (w/v) Tween 81 solution successfully absorbed over 90% of the toluene in an inlet gas containing toluene at 1000 ppmv. The toluene removal efficiency increased as the liquid flow rate increased. However, the ratio of the draft tube diameter to the column diameter did not affect the toluene removal efficiency. To the best of our knowledge, this is the first study of the pretreatment of industrial waste gases containing hydrophobic VOCs with a JLR to guarantee stable operation of a biofilter.

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