

การกำจัดไซลินด้วยวิธี Colloidal Gas Aphrons

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บทคัดย่อ

วิธีการแยกไซลินจากอากาศมีหลายวิธีด้วยกัน แต่การแยกสารด้วย Colloidal Gas Aphron (CGA) ก็เป็นอีกวิธีหนึ่งที่มีศักยภาพสูงที่ควรได้รับความสนใจ ในงานวิจัยที่ผ่านมาส่วนใหญ่ได้ใช้ CGA ในการแยกสารที่อยู่ในของเหลว ส่วนการแยกสารจากอากาศด้วย CGA นั้นมีอยู่น้อยมาก ดังนั้นงานวิจัยนี้จึงสนใจศึกษาการกำจัดไซลินด้วย CGA ซึ่งได้ทำการแยกโดยให้ไซลินไหลทางเดียวกับ CGA ในขั้นตอนได้ทำการศึกษาถึงความเสถียรและขนาดของ CGA ที่เตรียมจาก Triton X-100 จากการทดลองพบว่าความเสถียรของฟองเพิ่มขึ้นเมื่อความเข้มข้นของสารลดแรงตึงผิวเพิ่มขึ้น ความเร็วรอบในการกวนเพิ่มขึ้น แต่ระยะเวลาการกวนลดลง ขนาดของ CGA จะเพิ่มขึ้นเมื่อความเข้มข้นของสารลดแรงตึงผิวเพิ่มขึ้น ความเร็วรอบในการกวนต่ำ และระยะเวลาการกวนสั้น ในการศึกษาการแยกไซลินได้ทำการศึกษาดัชนีต่างๆ ที่มีผลต่อการแยกไซลิน ได้แก่ ชนิดของสารลดแรงตึงผิว ความเข้มข้นของสารลดแรงตึงผิว ระยะเวลาของ CGA ในคอลัมน์ ความเข้มข้นของไซลิน และอัตราการไหลเชิงมวลของ CGA จากการทดลองพบว่าสารลดแรงตึงผิวชนิดไม่มีประจุให้ประสิทธิภาพในการแยกไซลินสูงสุด โดยที่ประสิทธิภาพในการกำจัดเพิ่มขึ้นเมื่อเพิ่มความเข้มข้นของสารลดแรงตึงผิว อัตราการไหลเชิงมวลของ CGA เพิ่มขึ้น และระยะเวลาของ CGA ในคอลัมน์เพิ่มขึ้น สภาวะการดำเนินการที่ให้ประสิทธิภาพในการแยกสูงสุดคือ ความเข้มข้นของ Triton X-100 17 mM ระยะเวลาของ CGA ในคอลัมน์ 13.3 นาที อัตราการไหลเชิงมวลของ CGA 83 $\mu\text{mole/s}$ และความเข้มข้นของไซลิน 40 ppm

คำสำคัญ : Colloidal Gas Aphrons / ไซลิน / สารลดแรงตึงผิว / การกำจัดสาร VOC

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Xylene Separation by Using Colloidal Gas Aphrons

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Abstract

Xylene separation has been performed by many techniques. Colloidal Gas Aphron (CGA) showed highly potential for a number of applications in aqueous phase but few in gas phase. To our best knowledge, the xylene separation using CGA were not yet studied. In this study, separation of xylene was performed co-currently in a CGA separation column. In the preliminary experiment, CGA generated from Triton X-100 was subjected to the study of its stability and size. We found that the high stability of non-ionic CGA was obtained with high surfactant concentration, stirring speed and short stirring time. Regarding the size, however, the size of CGA was increased with high surfactant concentration, low stirring speed and short stirring time. For the xylene separation experiments, we assessed the system parameters of surfactant type, surfactant concentration, CGA retention time, xylene concentration and CGA mass flow rate to investigate the effect of these parameters on the removal efficiency. The non-ionic surfactant gave the highest yield. The removal efficiency increased by increasing surfactant concentration, CGA retention time and mass flow rate. The condition with 17 mM Triton X-100, CGA retention time of 13.3 min, CGA mass flow rate of 83 $\mu\text{mole/s}$, and 40 ppm of xylene concentration gave the highest efficiency.

Keywords : Colloidal Gas Aphrons / Xylene / Surfactant / VOC removal

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1. Introduction

The release of the volatile organic compounds (VOC) into the environment becomes an increasingly environmental problem in many industries such as petrochemical, printing, fine, and heavy chemicals industry. Benzene, toluene, and xylene (BTX) are ubiquitous materials in many industrial fields. These aromatic hydrocarbons are neurotoxin and cause diseases of the nervous system and sterility [1].

Conventional VOC elimination techniques are essentially of three types: adsorption, combustion and biological degradation. Adsorption techniques are useful when the recuperation of VOCs is economically worthwhile. Combustion techniques are also applicable when the VOC concentration is sufficiently high to maintain combustion. The principle of biotechnological processes is to use a gas-liquid contactor to transfer VOCs from the gas phase to the liquid phase, where they are degraded by specific microorganisms. The removal of xylene has been reported by many techniques: adsorption on zeolite [2], adsorption on soil mineral [3], solubilization in hydroxypropyl-cyclodextrins [4], separation by membrane [5] and biodegradation in biofilters [6]. Time consuming in biofiltration and high cost of zeolite are the drawbacks of these techniques. Colloidal gas aphrons (CGA) is another high potential for organic compound removal technique.

CGA are microbubbles (10-100 μm) encapsulated in a soapy film. They are created by intense stirring at 5,000-10,000 rpm. The attractive features of CGA are firstly their small sizes, resulting in a large surface area to volume ratio and secondly the existence of a soluble film of surfactant encapsulating the gas, retarding the bubbles from coalescing. CGA has been applied in many areas of separation

such as removal of heavy metal from aqueous solution [7], removal of organic dyes from wastewater [8], removal of hazardous oily waste from a soil matrix [9,10], clarification of suspensions [11] and lysozyme separation [12]. The electrostatic and/or hydrophobic interaction was thought to be the major mechanism of separation. When comparing with the other VOC removal techniques such as biofiltration, separation by CGA has many advantages in terms of rapid process, higher efficiency and low cost. Moreover, the separated VOC could be recovered from the liquid drainage.

In this work, Xylene was separated from air using CGA as continuous mode. The characterisation of CGA was firstly investigated in terms of stability and size. Then, effect of surfactant type and concentration, CGA retention time, xylene concentration, and CGA mass flow rate on xylene separation was examined.

2. Experimental

2.1 Materials

Triton X-100, cetyltrimethylammonium bromide (CTAB), and sodium dodecyl sulphate (SDS) were purchased from Sigma. Xylene was purchased from Lab Scan Asia Co. Ltd. All other chemicals were analytical grade purchased from Sigma.

2.2 CGA formation and characterisation

The CGA generator, as shown in Fig. 1, was the same as Sebba model [13], and comprised a Perspex cylindrical beaker (150 x 250 mm) with two baffles, a spinning disc of 5 cm in diameter connected to a motor of which speed is controlled by an inverter. CGA was generated by rotating the spinning disc at the speed of 4,800-6,000 rpm. The Triton X-100 surfactant solution of 3,000 ml with

the concentration varied at 10, 14 and 27 mM was added to the beaker and stirred for 3, 5, and 10 min respectively. The CGA formed by Triton X-100 was characterized by the half-life method and a light microscopy to investigate the stability and size respectively. The half-life was defined as the time required for half the amount of original liquid to

drain. Size of CGA was determined by using a light microscopy. CGA sample was placed on a microscope slide, magnified using a 20x magnification objective. Within 3 minutes, 5 images from different areas of each sample and standard scale were captured by a camera.

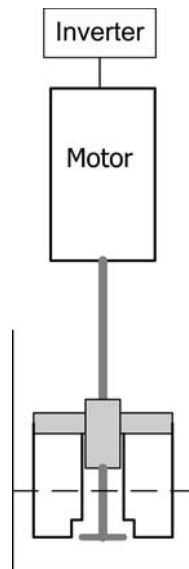


Fig. 1 CGA generator.

2.3 Xylene separation

The separation column comprised a Perspex cylinder of 60-mm internal diameter (ID) with a total length of 200 cm. A 50-mm stainless steel gas distributor was put at the bottom of column. In

order to simulate the xylene vapour as occurring in industry, pure xylene was fed into a stainless steel chamber with a high-pressure pump through atomizer. The vapour was then fed to the column by a connected air blower as shown in Fig. 2.

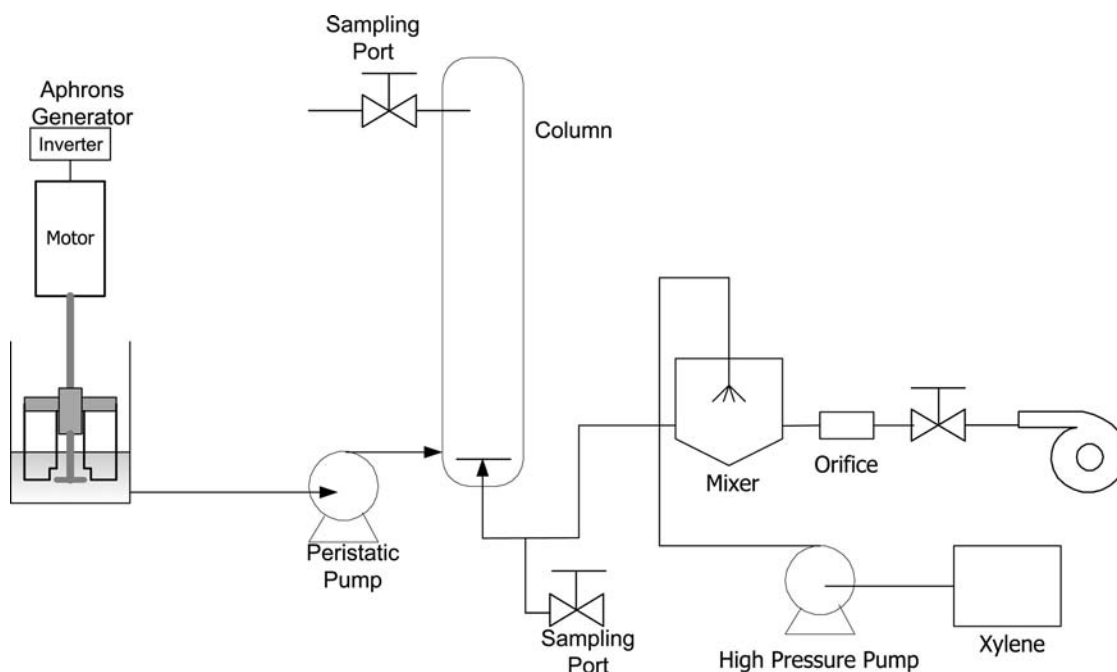


Fig. 2 Schematic diagram of xylene separation column.

The separation was conducted in co-current mode. The generated CGA with 6,000-rpm stirring time and 10 min stirring time was fed at predetermined flow velocity into the column by peristaltic pump until completely filled up. Subsequently, the xylene vapour mixed with air at predetermined concentration was fed at the bottom of the column with constant flow velocity of 74.63 cm/s (reten-

tion time of 1.4 s). Samples of xylene vapour at the input and output of the column were taken through sampling ports when the system reached the steady state (after 20 min). The system parameters of surfactant type and concentration, CGA retention time, xylene concentration, and CGA mass flow rate were subjected to study and to investigate their effects on the xylene separation as shown in Table 1.

Table 1 Experimental conditions

System parameters	Surfactant type and concentration	CGA Retention time (min) (Column height (cm))	Xylene concentration (ppm)	CGA mass flow rate ($\mu\text{mole/s}$)
Surfactant type	10 mM Triton X 100, CTAB, SDS	7	90	65
Surfactant conc.	Triton X 100 of 12, 14,15,17,18,26, 34 mM	7	90	65
Retention time (Column height)	Triton X 100 of 17 mM	2, 3.7, 5.3, 7, 13.3 (30, 55, 80, 105, 200)	90	65
Xylene conc.	Triton X 100 of 17 mM	105	30, 40, 50, 90, 100	65
CGA mass flow rate	Triton X 100 of 17 mM	105	90	48, 58, 65,83

2.4 Xylene concentration determination

Xylene was taken from sampling port at inlet and outlet of the column by Gas Tight syringe. Then, the concentration of xylene in vapour phase of both inlet ($C_{X(in)}$, ppm) and outlet ($C_{X(out)}$, ppm) were measured with gas chromatography (Shimadzu QC-9A). Then, the removal efficiency of xylene was calculated by the following equation.

$$\text{Removal efficiency} = \left(C_{X(in)} - C_{X(out)} \right) / C_{X(in)} * 100$$

3. Results and discussion

3.1 Characterization of CGA

Triton X100 CGA was subjected to characterize because it gave the highest removal efficiency (see effect of surfactant type in the next section). Fig. 3 shows the effect of surfactant concentration, stirring time, and stirring speed on the CGA stability presented in terms of the half-life. The higher the concentrations of surfactant, the better stability was obtained. This might be attributed to the increase of elasticity at higher concentration of non-ionic

surfactant [14]. However, the CGA stability slightly increased with increasing of stirring time but it started to decrease at 10 min of stirring time with high surfactant concentration. After CGA formation, continuously stirring may inversely destabilize the CGA as reported by Save [15]. Sebba (1985) [13] suggested that stirring speed should be lowered after complete formation of CGA from economic point of view.

The effects of stirring time and surfactant concentration on CGA size are shown in Fig. 4. The diameter of CGA increased with higher surfactant concentration, shorter stirring time and lower speed. When comparing the size with stability, the smaller size of CGA is, the better stability should be. This may be explained by longer coalescent time of the small bubbles. Under the conditions of 10-min stirring time, 6,000 rpm stirring speed and 10 mM Triton X100 concentration, the size of CGA became smallest resulting in the highest stability (6 min of half life), as shown in Fig. 3. Hence, in the separation, small CGA was favorable.

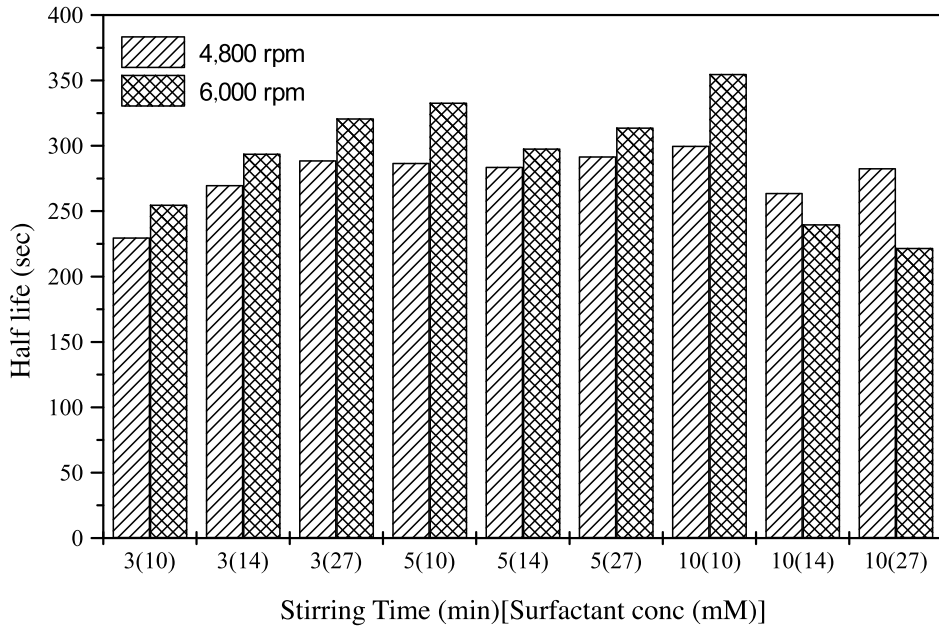


Fig. 3 Effect of stirring time, stirring speed, and surfactant concentration on stability.

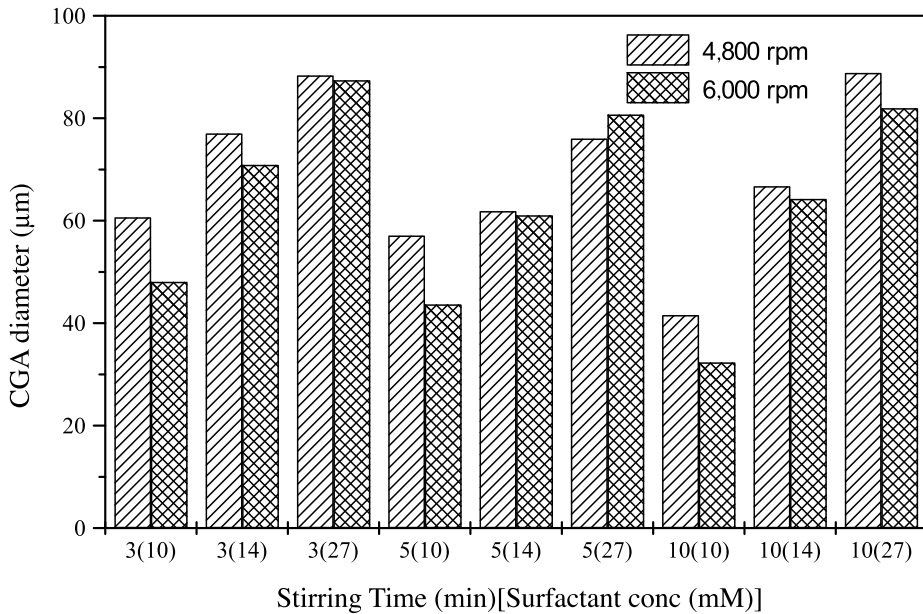


Fig. 4 Effect of stirring time, stirring speed, and surfactant concentration on CGA size.

3.2 Xylene separation

3.2.1 Effect of surfactant type

Fig. 5 shows the effect of surfactant type on the percentage of removal. CGA prepared from all type of surfactants could remove xylene from vapour phase. CGA using Triton X100 was the most effective for the xylene separation. The yield of CGA using Triton X100 was 65% while those of CTAB and SDS were 40% and 42%, respectively. Based on the chemical properties of the surfactant, the good yield removal of CGA generated by Triton X100 can be attributed to the

number of carbon atoms at hydrophobic side, which were 14, 14 and 12 for Triton X100, CTAB, and SDS, respectively. In addition, the aromatic structure and non-ionic characteristic of Triton X100 may be involved in the enhancement of the solubility of xylene [14]. In terms of separation time, the removal efficiency by Triton X-100 reached steady state within 20 min. This indicated that CGA separation was a rapid process. After drainage of CGA, adsorbed xylene could be recovered by separation from water.

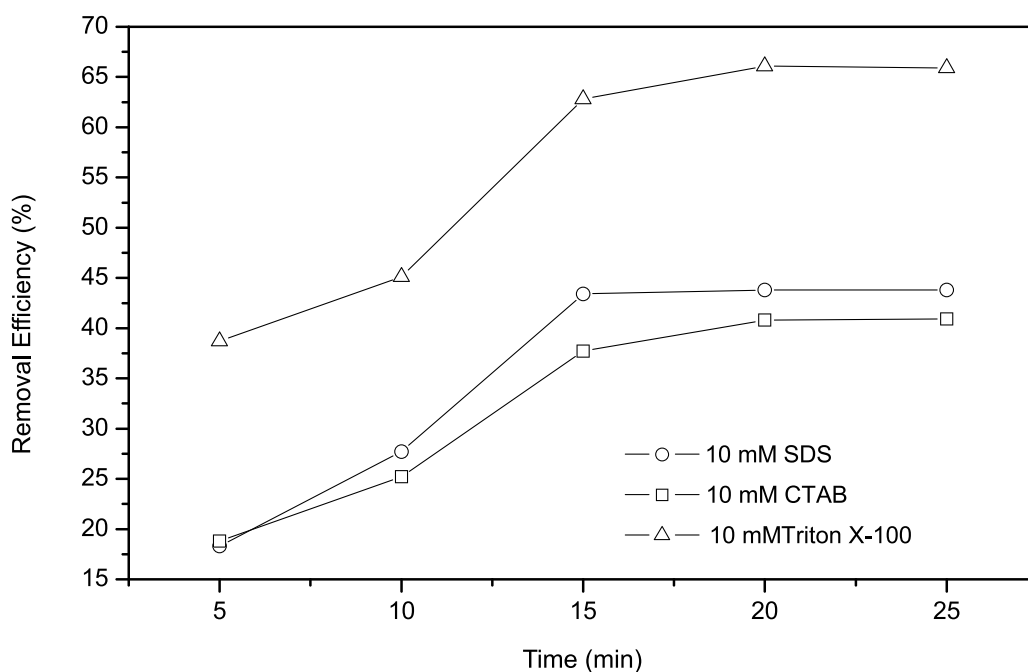


Fig. 5 Effect of surfactant type on xylene separation at CGA retention time of 7 min, CGA mass flow rate of 65 $\mu\text{mole/s}$, and 90 ppm xylene.

3.2.2 Effect of surfactant concentration

In order to study the effect of surfactant concentration on the separation, various Triton X100 concentrations were investigated. Fig.6 demonstrates that the removal efficiency increased with higher surfactant concentration, and approached to constant separation of 68% at 34 mM Triton X100.

The increment was attributed to the larger amount of CGA formed and higher stability of CGA at higher surfactant concentration [16]. However, the saturated removal efficiency at higher concentration (e.g. 68%) may be limited by column height resulting in the short retention time.

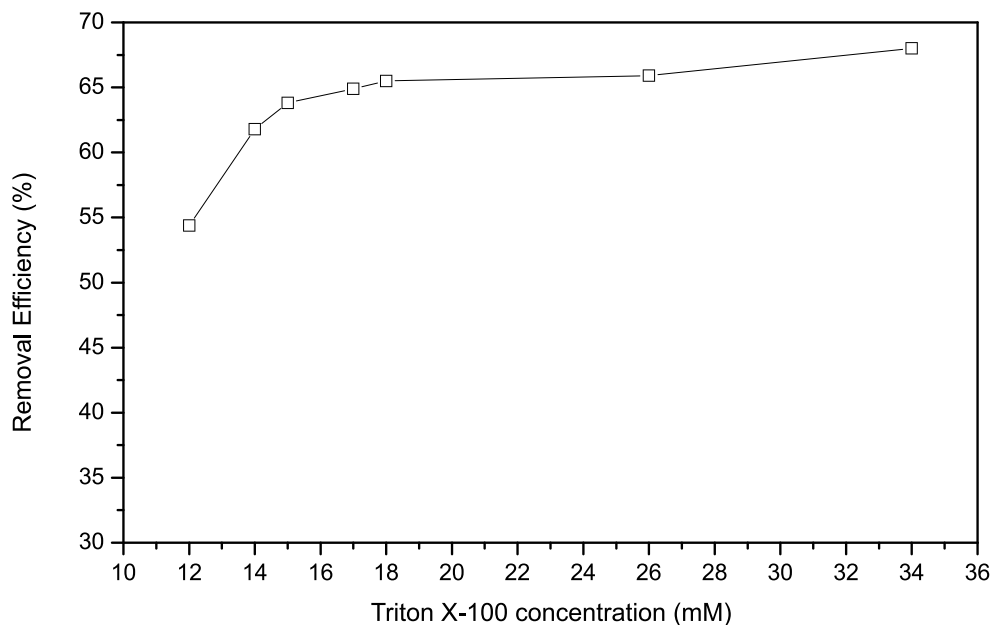


Fig. 6 Effect of surfactant concentration on xylene separation at CGA retention time of 7 min, CGA mass flow rate of 65 $\mu\text{mole/s}$, and 90 ppm xylene.

3.2.3 Effect of CGA retention time

The effect of column height on removal efficiency was examined in the range of 30-200 cm, while the xylene concentration and CGA mass flow rate were kept at 90 ppm and 65 $\mu\text{mole/s}$, respectively. Retention time of CGA at each column height was 2, 3.7, 5.3, 7, and 13.3 min, respectively while the retention time of xylene was constant at 1.4 s. Increasing the retention time resulted in a greater removal efficiency of xylene, as shown in Fig. 7. The adsorption was proportional to the retention time, whereas the rate of adsorption slightly declined when increasing the retention time. This could be ascribed to the longer contact time between xylene

and CGA in the column due to the longer column height. However, CGA coalesced and depleted more with the increasing height resulting in slightly reduction of the rate of removal efficiency. At the highest retention time of 13.3 min, percentage of adsorption was 90. With the condition of CGA formed (17 mM Triton X-100, 6,000 rpm stirring speed and 10 min stirring time), the half life of CGA was 6 min. Comparing the half life time with the retention time of each height, fed CGA should be less deplete with the shorter column. Therefore, stability of CGA should be an important parameter for the adsorption.

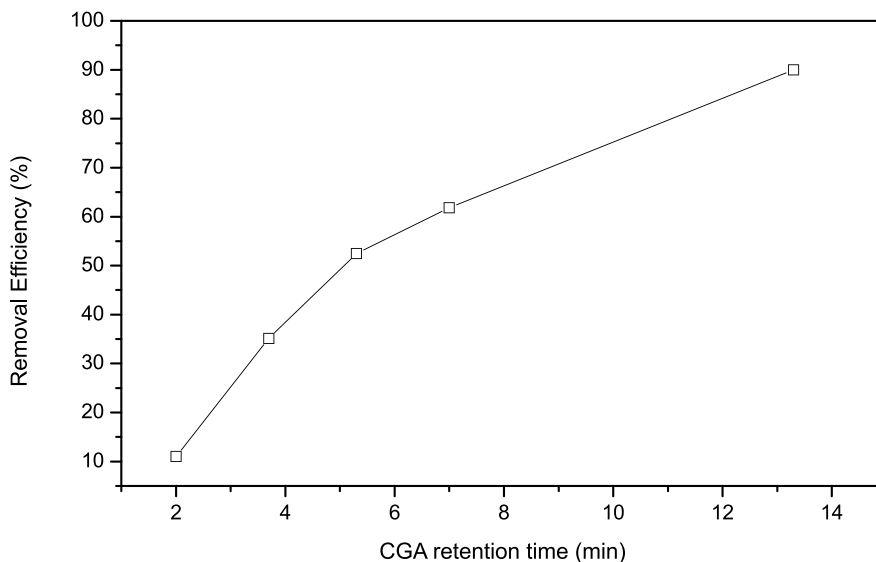


Fig. 7 Effect of CGA retention time on xylene separation at 17 mM Triton X100, CGA mass flow rate of 65 μ mole/s, and 90 ppm xylene.

3.2.4 Effect of xylene concentration

The concentration of xylene was varied in order to investigate CGA adsorption efficiency (as shown in Fig. 8). With increasing xylene concentration from 30 to 50 ppm, the removal efficiency slightly decreased, while the yield rapidly declined when xylene concentration greater than 90 ppm. The

reduction of removal efficiency was attributed to the constant volume of CGA resulting in limitation of adsorption capacity. Based on the volume of column used, these results indicated that 2.1×10^4 mole of CGA could remove 0.3 mole of xylene and gave removal efficiency of 62-65%.

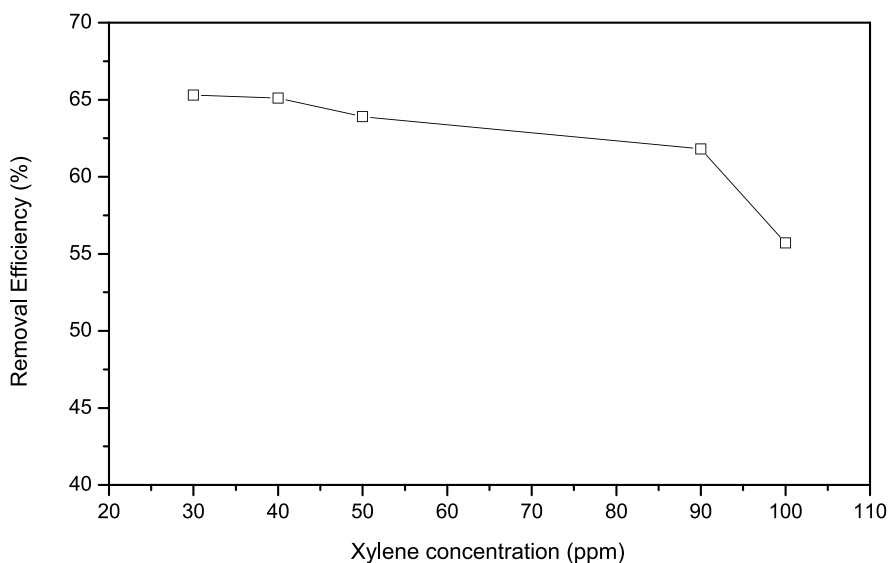


Fig. 8 Effect of xylene concentration on xylene separation at 17 mM Triton X100, CGA retention time of 7 min, and CGA mass flow rate of 65 μ mole/s.

3.2.5 Effect of CGA mass flow rate

Fig. 9 shows the effect of CGA mass flow rate on the percentage of adsorption at 17 mM Triton X-100 and air flow velocity of 74.63 cm/s. The removal efficiency increased with increasing CGA mass flow rate. This may be explained by

replacement of adsorbed CGA with the fresh one resulting in increasing the chance of fed xylene to be adsorbed by the fresh CGA. Therefore, the adsorption was enhanced with co-current flow operation.

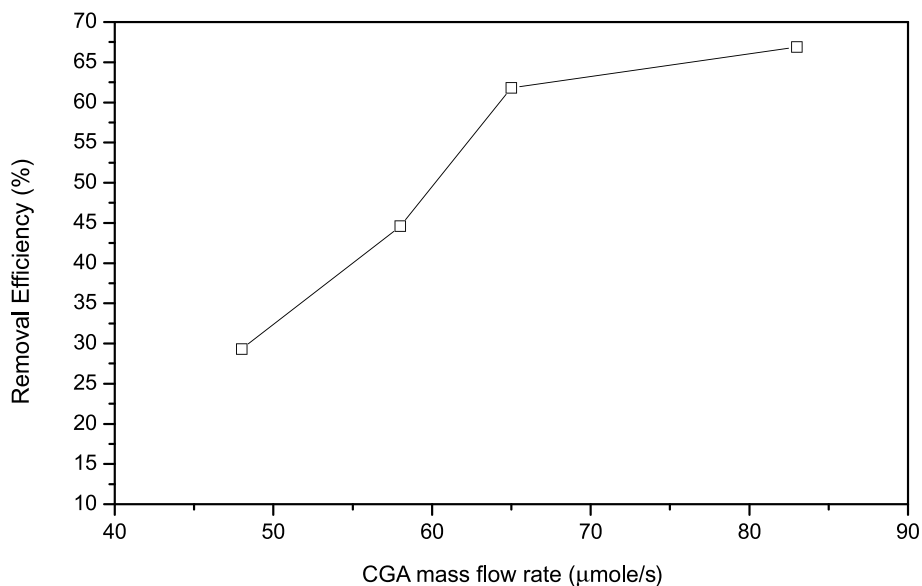


Fig. 9 Effect of CGA mass flow rate on xylene separation at 17 mM Triton X100, CGA retention time of 7 min, and 90 ppm xylene.

4. Conclusions

The characterization of CGA was carried out by studying the effect of surfactant concentration, stirring speed and time on stability and size of CGA. High stability CGA was obtained at high surfactant concentration, high stirring speed, and low stirring time. However, at low surfactant concentration (more than critical micellar concentration), and high stirring speed and time, the smaller CGA was obtained.

The non-ionic surfactant, Triton X-100, gave the highest yield of xylene removal. At high surfactant concentration, high CGA mass flow rate and retention time, the removal efficiency increased. The

highest yield of separation (65%) was obtained with the condition: 17 mM Triton X-100, CGA retention time of 13.3 min, CGA mass flow rate of 83 $\mu\text{mole/s}$, and 40 ppm of xylene concentration. Base on the condition, 2.1×10^4 mole of CGA could remove 0.3 mole of xylene.

5. References

1. White R.F. and Proctor S.P., 1997, "Solvents and Neurotoxicity", *Lancet*, Vol. 349, pp. 1239-1243.
2. Sakai H., Tomita T., and Takahashi T., 2001, "*p*-Xylene Paration with MFI-type Zeolite Membrane", *Separation and Purification Technology*, Vol. 25, pp. 297-306.

3. Ruiz J., Bilbao R., and Murillo M.B., 1998, "Adsorption of Different VOC onto Soil Minerals from Gas Phase: Influence of Mineral, Type of VOC, and Air Humidity", *Environ. Sci. Technol.*, Vol. 32, pp. 1079-1084.
4. Tanada S., Nakamura T., Kawasaki N., Torii Y., and Kitayama S., 1999, "Removal of Aromatic Hydrocarbon Compounds by Hydroxypropyl-cyclodextrin", *J. Colloid Interface Sci.*, Vol. 217 pp. 417-419.
5. Burggraai A.J., Vroon Z.A.E.P., Keizer K., and Verweij H., 1998, "Permeation of Single Gases in Thin Zeolite MFI Membranes", *Journal of Membrane Science*, Vol. 144 pp. 77-86.
6. Jorio H., Bibeau I., Viel G., and Heitz M., 2000, "Effects of Gas Flow Rate and Inlet Concentration on Xylene Vapors Biofiltration Performance", *Chem. Eng. J.*, Vol. 76, pp. 209-221.
7. Amiri M.C. and Woodburn E.T., 1990, "A Method for the Characterisation of Colloidal Gas Aphron Dispersions", *Trans. Inst. Chem. Eng.*, Vol. 68A, pp. 154-160.
8. Roy D., Valsaraj K.T., and Kottai S.A., 1992, "Separation of Organic Dyes from Wastewater by Using Colloidal Gas Aphrons", *Sep. Sci. Technol.*, Vol. 27, pp. 573-588.
9. Roy D., Kommalapati R.R., Valsaraj K.T., and Constant W.D., 1997, "Soil Flushing of Residual Transmission Fluid: Application of Colloidal Gas Aphron Suspensions and Conventional Surfactant Solutions", *Water Res.*, Vol. 31, pp. 2161-2170.
10. Kommalapati R.R., Valsaraj K.T., Constant W.D., and Roy D., 1998, "Soil Flushing using Colloidal Gas Aphron Suspensions Generated from a Plant-based Surfactant", *J. Haz. Mat.*, Vol. 60, pp. 73-87.
11. Hashim M.A. and Gupta B.S., 1998, "The Application of Colloidal Gas Aphrons in the Recovery of Fine Cellulose Fibers from Paper Mill Wastewater", *Bioresource Technology*, Vol. 64, pp. 199-204.
12. Jauregi P. and Varley J., 1996, "Lysozyme Separation by Colloidal Gas Aphrons", *Progr. Colloidal Polym Sci.*, Vol. 100, pp. 362-367.
13. Sebba F., 1985, "An Improved Generator for Micron-sized Bubbles", *Chem. Ind.*, Vol. 3, pp. 91-92.
14. Rosen M.J., 1978, *Surfactants and Interfacial phenomena*, John Wiley & Sons.
15. Save S.V. and Pangarkar V.G., 1994, "Characterisation of Colloidal Gas Aphrons", *Chem. Eng. Comm.*, Vol. 127, pp. 35-54.
16. Jauregi P. and Varley J., 1998, "Colloidal Gas Aphrons: A Novel Approach to Protein Recovery", *Biotechnol. Bioeng.*, Vol. 59, pp. 471-481.