Process parameters for operating 1-butanol gas stripping in a fermentor

Ying-Chen Liao, Kuan-Ming Lu, and Si-Yu Li*

Department of Chemical Engineering, National Chung Hsing University, Taichung 402, Taiwan

In this study, effects of the agitation speed, the flow rate, and type of non-polar gases on the performance of gas stripping was systematically investigated. Macroscopically, the stripping rate of butanol is linearly proportional to the concentration of butanol in the feed solution. Nevertheless, a decrease in butanol selectivity was observed with the increasing butanol concentrations up to 0.01 g/cm³. This can be attributed to the thermodynamics reason that with increasing butanol concentrations in the feed, more stripping gas will dissolve in the feed solution that decrease the activity of butanol for mass transfer from liquids to gas bubbles. This can be supported by the use of highly soluble gas of carbon dioxide as the stripping gas where the $K_S a$ dropped 48% compared to the nitrogen stripping. By the parameter sensitivity analysis, it has been shown that the dominant variable is the flow rate. The best strategy of maximizing the performance of 1-butanol gas stripping at a given flow rate is to bubble the gases at a high superficial velocity, which leads to a less resistance on the liquid side for mass transfer.

Gas stripping is a chemical separation process where target compounds were removed from an aqueous solution by bubbling gas from the bottom. The mass transfer occurs at the interface of liquid and vapor phases in which the target compounds volatilize faster than water to achieve the separation. Because of low cost, simple operation, and biocompatibility, gas stripping has been widely used in different biological processes (1—5) as well as in the application of environmental remediation (6). One important application of gas stripping is to serve as the *in situ* butanol removal technique during acetone−butanol−ethanol (ABE) fermentation (7—10). 1-Butanol has shown substantial inhibition effect on the bacterial growth at the concentration above 0.006 g/cm³ and the threshold concentration is about 0.010−0.015 g/cm³ (10−12). By circumventing the accumulation of 1-butanol during ABE fermentation through *in situ* gas stripping, 20−100% improvement in butanol productivity of ABE fermentation can be achieved (2,3,13,14).

Macroscopically, the process of gas stripping can be described by the first order kinetics as shown in Eq. 1 (15). R_S represents the stripping rate of the solute, where C_S and $K_S a$ are solute concentrations in the aqueous solution and stripping rate constant, respectively. Eq. 1 indicates that the stripping rate of solute is proportional to its concentration in the bulk liquid, which has been verified in many studies (13,15,16).

$$R_S = -\frac{dC_S}{dt} = K_S a \cdot C_S \tag{1}$$

$$K_S a = \frac{a}{V} \cdot \left[\frac{1}{k_l} + \frac{RT}{H' k_g} \right]^{-1} \tag{2}$$

$$K_S a = b \frac{Q}{V} (H_c)^m \tag{3}$$

The stripping rate constant, $K_S a$, has been modeled by two equations as shown in Eqs. 2 (15,17) and 3 (15). The nomenclature for Eq. 2 is as follows: a is the interfacial area (cm²), V is the total volume of the fluid (cm³), k_l is the liquid film mass transfer coefficient of the solute (cm s⁻¹), R is the universal gas constant (cm³ atm mol⁻¹ K⁻¹), T is the absolute temperature, H' is the Henry's law constant with the unit of atm cm³ mol⁻¹, and k_g is the gas film mass transfer coefficient of the solute (cm s⁻¹). The nomenclature for Eq. 3 is as follows: b and m are power function constants, Q is the gas flow rate (liter per second, lps) and H_c is the dimensionless Henry's law constant. Eq. 2 was developed based on the two-resistance theory. It indicates that the stripping rate R_S can be improved by increasing the interfacial area, a, by decreasing the bubble size at a given flow rate. The above theoretical prediction can be supported by the experiment as discussed below. Ezeji et al. have compared two gas bubble delivery systems, the sparger and the impeller, to remove solvents during ABE fermentation. It has been found that the sparger, which delivered smaller bubble size, was preferred in terms of the solvent removal (16). Though Eq. 2 indicates that the bubble size plays an important role for the performance of gas stripping, the effects of flow rates on the performance of gas stripping remain vague. Truong and Blackburn proposed a correlation that related the stripping rate constant to the gas flow rate as shown in Eq. 3 (15). It can be seen that $K_S a$ is linearly proportional to the gas flow rate. However, Eq. 3 is unclear how $K_S a$ changes with respect to the interfacial area, a. Flow rates and interfacial area, a, are two easily managed parameters for gas

* Corresponding author. Tel.: +886 4 2284 0510x509; fax: +886 4 2285 4734.
  E-mail address: syli@dragon.nchu.edu.tw (S.-Y. Li).

stripping. The lack of a mathematical correlation that relates K_Sa with both the flow rate, Q, and the interfacial area, a, makes the scale-up of gas stripping process indefinable.

Since gas stripping is widely used as an *in situ* butanol removal technique to reduce the toxicity of butanol during ABE fermentation, it is necessary to study the effect of the geometry of the fermentor on performance of gas stripping. The agitation system of a fermentor is designed to increase the liquid−gas mass transfer during aerobic fermentation by breaking the air bubbles with high agitation speeds. In this study, the effects of the agitation speed and the flow rate on the performance of gas stripping was systematically investigated, where the separation of 1-butanol was achieved in a fermentor. Furthermore, different types of non-polar gases were used to investigate the performance of 1-butanol stripping.

MATERIALS AND METHODS

Experimental apparatus and conditions for 1-butanol gas stripping   A schematic of the experimental apparatus for 1-butanol gas stripping used in this study is shown in Fig. 1A. An automated fermentor was used as the separation tank where the temperature, gas flow rate, and agitation were controlled by the fermentor console. The mixing was achieved by three six-blade disk turbines in a baffled tank. The geometry of the agitation system is shown in Fig. 1B. 2.5 L of 1-butanol aqueous solution in a 3-L fermentation tank was used during gas stripping and 1-butanol recovery. The temperature in the fermentor was maintained at 37°C under atmospheric pressure. The gas was distributed in the fermentor through a sparger from the bottom side, where the bubble size was roughly estimated between 0.2 and 0.5 cm. The position of the sparger in the fermentor and its dimensions are shown in Fig. 1C. Parallel cold traps were arranged and cooled in liquid nitrogen baths for collecting condensates. Samplings were done from one cold trap to another so that the performance of overall 1-butanol gas stripping process can be recorded.

Analytical method   A quantitative analysis of 1-butanol of both feed solutions and cold trap samples were done by a Hewlett Packard gas chromatography (HP 5890 Series II GC) as described previously [18]. The GC was equipped with a split/splitless injector, flame-ionized detector, DP-FFAP capillary column (30 m, 0.32 mm i.d., 0.25 μm film thickness). The split mode with an injection volume of 1 μL was used, where the split ratio was set to 1:10. The temperature of the injector and detector was maintained at 225°C. The temperature profile of the analysis is described as follows: the oven temperature was initially held at 50°C for 4 min, and raised to 100°C at the ramp of 20°C/min and held for 1 min. Then raised to 170°C at the ramp of 30°C/min and held for 2.5 min. Finally, the oven temperature was raised to 220°C at the ramp of 20°C/min and held for 4 min. Nitrogen was used as the carrier gas.

Mathematical development   The stripping rate constant, K_Sa, can be derived from Eq. 1 as a function of butanol concentrations in the fermentor at a given time. By definite integration of Eq. 1, K_Sa can be expressed as following:

$$K_S a = \frac{\ln\left(\frac{C_{B0}}{C_B}\right)}{t} \qquad (4)$$

where C_{B0} and C_B represent the initial butanol concentration and the butanol concentration in the fermentor, respectively. Therefore, K_Sa can be experimentally measured by plotting the logarithm of C_{B0}/C_B versus time, where the slope is K_Sa.

RESULTS AND DISCUSSION

Concentration dependence of 1-butanol gas stripping   The concentration dependence of 1-butanol gas stripping was investigated where 0.006, 0.012, and 0.025 g/cm^3 were used as initial 1-butanol concentrations. The gas flow rate and the agitation speed were controlled at 66.7 cm^3/s (or 4 L per minute, lpm) and 250 rpm, respectively. Condensates were collected every 1440−3600 s for calculating 1-butanol and water stripping rates as shown in Fig. 2A and B. It can be seen in Fig. 2A that the 1-butanol stripping rate (R_S, g-butanol/cm^3/s) is linearly proportional to the 1-butanol concentration of the feed solution. On the other hand, the water removal rate shows a behavior that is independent of the 1-butanol concentration of the feed solution, see Fig. 2B. A constant water stripping rate can be attributed to the use of dilute 1-butanol solutions for separation, where the water concentration is nearly a constant throughout the stripping process. Fig. 2A and B shows that Eq. 1 is generally valid and useful for describing the process of butanol gas stripping, which has been suggested previously [2,15,16]. In addition to the stripping rate, selectivity is another important parameter used to evaluate the performance of separation processes. Butanol selectivity of gas stripping, α_B, is defined as shown in Eq. 5:

$$\alpha_B = \frac{\left(\frac{w'_B}{1-w'_B}\right)}{\left(\frac{w_B}{1-w_B}\right)} \qquad (5)$$

where w'_B and w_B are weight fractions of butanol in condensates and feed solutions, respectively. Fig. 2C shows that butanol selectivity is in the range of 16−20 with the low butanol concentration of 0.006 g/cm^3 in the feed solution. Butanol selectivity decreased a little to a range of 13−17 with increasing butanol concentrations in the tank. A trade-off between the stripping rate and selectivity during the gas stripping of butanol is reported in this study, which
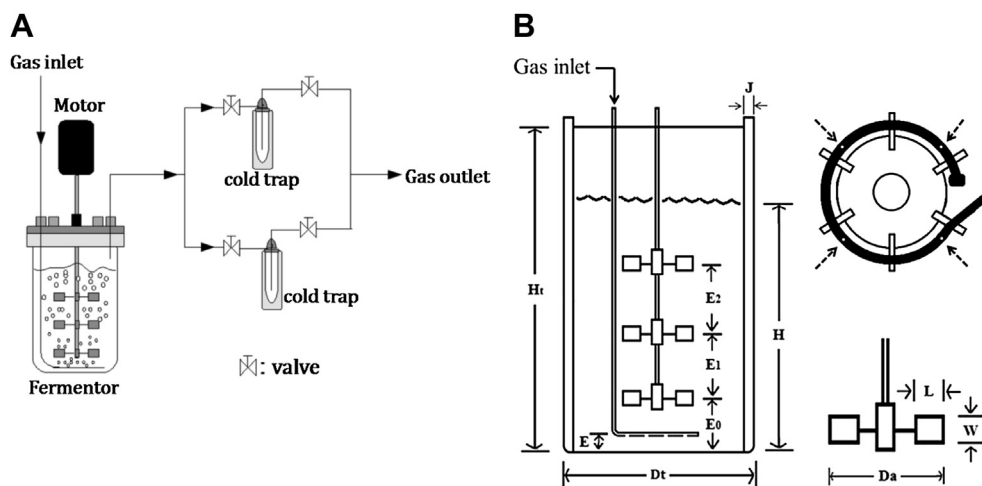

FIG. 1. (A) The schematic of the experimental apparatus for 1-butanol gas stripping used in this study. (B) The geometry of the sparger and the mixing system (expressed in cm): $H_t = 26$, $J = 1.2$ (width of baffles), $H = 17.5$ (with the liquid volume of 2.5 L), $D_t = 13.4$, $D_a = 6.5$, $E_0 = 4.75$, $E_1 = 5$, $E_2 = 4.2$, $E = 1.0$, $L = 1.5$, $W = 1.5$ cm. The figure on the upper right of panel B illustrates the viewing angle from the bottom side of the sparger. Four dashed arrows indicate all four gas outlets (illustrated as white dots) from the sparger where the diameter is 0.1 cm.