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Diagnosis of Bubble Distribution in a Three-Phase Bubble Column Reactor for Dehydration of Ortho-Boric Acid

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Abstract−For the practical application of a three-phase bubble column as a reactor in the dehydration of ortho-boric acid, we investigated the bubble distribution and its effects on the reaction in a three-phase bubble column reactor (0.102 m ID · 2.0 m in height) operating at relatively low pressure (below the atmospheric pressure). Effects of reaction time, temperature, gas velocity, particle size and gas injection mode (even, wall-side, central and asymmetric distribution) on the fractional conversion of the reaction were determined. The complicated bubble distribution as well as bubbling phenomena in the reactor were diagnosed and interpreted by means of the attractor trajectories and correlation dimension which were obtained from the resultant pressure fluctuations. The fractional conversion was closely related to the attractor shape or correlation dimension of the pressure fluctuations in the reactor. The fractional conversion in the case of even distribution of gas injection exhibited the highest value in all cases studied, at which the attractor of pressure fluctuations was less scattered in the phase space, while their correlation dimension had the lowest value. When the gas was injected by means of wall-side distribution, the conversion level was higher than that in case of central or asymmetric distribution mode. Although a fluid-solid heterogeneous reaction model can be applicable to the reaction, deviations from the model become considerable when the gas injection mode changes from even to wall-side, central or asymmetric mode, orderly.

Key words: Dehydration, Boric Acid, Bubble Distribution, Pressure Fluctuations, Chaos Analysis, Three Phase, Bubble Column Reactor

INTRODUCTION

Bubbling phenomena in a bubble column reactor have been known to be one of the most important factors in determining the heat and mass transfer rates as well as the fractional conversion level, thereby determining the performance of bubble column reactors or contactors. Therefore, several investigators have studied bubbling phenomena in bubble column reactors [Idogawa et al., 1986; Deckwer et al., 1993; Kang et al., 1999a, 2000]. In spite of many investigations on pressurized bubble columns, relatively little attention has been focussed on the hydrodynamics or performance of the bubble column operating at low pressure, which is often encountered in chemical and mechanical processes [Kodo et al., 1975; Kang et al., 1995]. Moreover, most investigations have not considered the effects of distributor or bubble distribution at the distributor on the hydrodynamics and transport phenomena in the column, although the effects should not be ignored [Lanauze and Harris, 1974; Tsuge and Hibino, 1983; Kang et al., 1999a].

For the practical application of a bubble column as a reactor or a contactor, the bubble distribution and its effects on the bubbling phenomena as well as the reaction conversion level have to be diagnosed, especially when operating at low pressure. Specifically, in conducting the dehydration of ortho-boric acid, information on the bubble distribution in the reactor is indispensable, because the removal of water vapor generated during the reaction by means of rising bubbles is one of the important factors in determining the conversion level of the reaction [Kang et al., 1995]. Thus, for the possibility that a three-phase bubble column is applicable as a reactor in conducting the dehydration of ortho-boric acid, the bubble distribution and its effects on the reaction at relatively low pressure (below the atmospheric pressure) have to be analyzed, because those are directly related to the removal of water vapor in the reactor.

However, the bubbling phenomena in the three-phase bubble column reactor are highly complicated, stochastic and non-linear. Fortunately, the complex and irregular multiphase contacting and bubbling behavior in dynamic multiphase flow systems have been successfully described and analyzed by adopting the deterministic chaos theory [Drahos et al., 1992; Kang et al., 1996a, b, 1999b; Kikuchi et al., 1997; Kim and Han, 1999; Lee et al., 2001].

In the present study, therefore, bubble distribution and bubbling phenomena in a three-phase bubble column reactor operating at low pressure have been diagnosed and interpreted by analyzing the resultant pressure fluctuations in the reactor based on the deterministic chaos theory. Also, effects of bubble distribution and bubbling phenomena on the conversion level of the reaction have been discussed.

ANALYSIS

1. Phase Space Portraits

Multidimensional phase-space portraits can be constructed from the pressure fluctuation time series by means of the time delay method [Packard et al., 1980; Roux et al., 1983]. That is, the experi-

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mentally obtained time-series signal, X(t), is digitized with a time step of Δt; the resultant (m+1) values of the signal, X(iΔt), are stored for

$$i=0, 1, 2, \Lambda, m.$$

Thus, the vector time series is defined as

$$\mathbf{Z}_i(t)=[\mathbf{X}(i\cdot\Delta t), \mathbf{X}(i\cdot\Delta t+\tau), \Lambda, \mathbf{X}(i\cdot\Delta t+(p-1)\cdot\tau)],$$
$$i=0, 1, 2, \Lambda, [m-(p-1)k], \qquad (1)$$

where, $\tau=k\cdot\Delta t$, k=1, 2, 3, ...

and p is the dimension of the vector, Z(t). Therefore, moving along with time t, a series of *p*-dimensional vectors representing the *p*-dimensional portrait of the system can be obtained. Occasionally, p is referred to as the embedded phase-space dimension of the reconstructed trajectory or attractor.

2. Correlation Dimension

To estimate the correlation dimension of the time series X(t), their trajectories, reconstructed by resorting to time embedding, have been used. From the trajectories of the vector time series the correlation integral (the space correlation function) of the process, C(r), is defined as [Grassberger and Procaccia, 1983; Abraham et al., 1986]

$$C(r)=\lim_{m\to\infty}\frac{1}{m^2}\begin{bmatrix}\text{number of pairs } (i, j) \\ \text{whose distance } |\mathbf{Z}_i(t)-\mathbf{Z}_j(t)|<r\end{bmatrix} \qquad (2)$$

Formally,

$$C(r)=\lim_{m\to\infty}\frac{1}{m^2}\sum_{i=1}^{m}\sum_{j=1}^{m}H[r-|\mathbf{Z}_i(t)-\mathbf{Z}_j(t)|],\ i\neq j \qquad (3)$$

where m is the number of data points, and H is Heavyside function, which can be expressed as:

$$H[r-|\mathbf{Z}_i(t)-\mathbf{Z}_j(t)|]=\begin{cases}1 \text{ if } r>|\mathbf{Z}_i(t)-\mathbf{Z}_j(t)| \\ 0 \text{ otherwise}\end{cases} \qquad (4)$$

The correlation integral, C(r), has been found to be a power function of r for small r's:

$$C(r)=kr^{D_C} \qquad (5)$$

The slope of the plot of ln C(r) vs. ln r is an estimate of D_C, which is termed as the correlation dimension, for the given embedded space dimension, p.

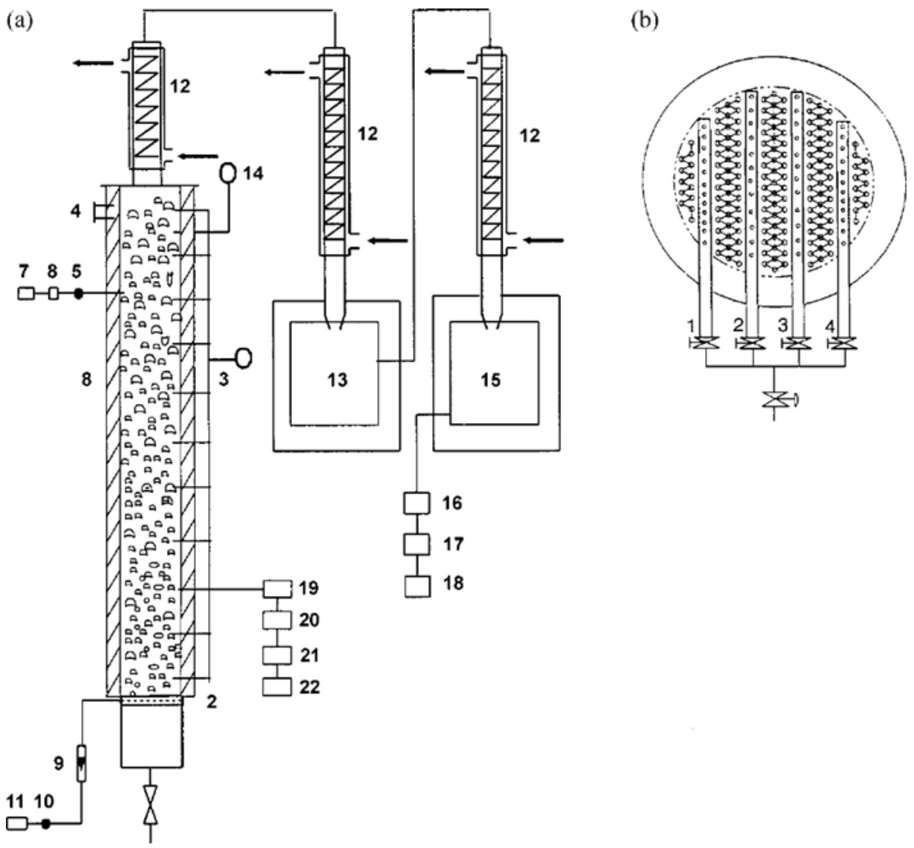

Fig. 1. (a) Experimental apparatus.

1. Reactor
2. Distributor
3. Manometer
4. Feed inlet
5. Thermocouple
6. Digital relay
7. T-Controller
8. Heating mantle
9. Rotameter
10. Filter
11. Compressor
12. Reflux condenser
13. First liquid vessel
14. Pressure gauge
15. Second liquid vessel
16. Filter
17. Vacuum pump
18. Controller
19. Pressure transducer
20. Amplifier
21. A/D Converter
22. Computer

(b) Distributor design.

EXPERIMENT

Experiments were performed in a stainless-steel column of 0.102 m in diameter and 2.0 m high as shown in Fig. 1. The distributor was situated between the main column section and a 0.2 m high stainless-steel distributor box into which liquid was introduced through a 0.025 m pipe from the liquid reservoir. Oil-free compressed air was fed to the column through a pressure regulator, filter and a calibrated rotameter. It was admitted to the column through four 3.0 mm ID perforated pipes drilled horizontally in grid having 12 holes whose diameter was 1.0 mm.

Modes of gas injection at the distributor were either even, wall-side, central or asymmetric distribution, which were adjusted by altering the gas injection line in the distributor. That is, all lines (lines 1-4 in Fig. 1b) in the distributor were opened when the gas injection mode was even, while in the case of wall-side distribution, lines 1 and 4 in the distributor were opened and others (lines 2 and 3) were closed. Similarly, when the gas injection mode was central, only lines 2 and 3 were opened, and either lines 1 and 3 or lines 2 and 4 were opened in the case of the asymmetric gas injection condition. The total amount of gas was equal, although its injection mode was altered in a given operating condition.

The gas holdup was obtained by means of the static pressure drop method [Kang et al., 1996a, 1999a, b, 2000]. Pressure fluctuations were measured by means of pressure sensors (semi-conductor type) at 0.4 m from the distributor. Nine pressure taps were mounted flush with the wall of the column with a 0.2 m interval from the distributor. The output voltage from the pressure transducer was processed by means of a data acquisition system (Data Precision Model, D-6000) and a personal computer. The sampling rate was 5 msec yielding the total sample length of 5000. This combination of sampling rate and time can detect the full spectrum of the signals [Kang et al., 1999a, b, 2000].

A reflux condenser was installed at the top of the column, and it was connected to two condensers in order to obtain the water evaporated. The air in the sealed column was withdrawn by means of a vacuum pump connected to the end of the reflux condenser to maintain the reaction pressure at a constant value.

To adjust the reaction temperature the column was fitted with a heating mantle that was controlled by a temperature control system. The particle size of ortho-boric acid was in the range of $0.152\text{-}0.6\cdot 10^{-3}$ m, and the n-paraffin liquid had a density of 820 kg m^{-3} and a surface tension of $22.01\cdot 10^{3}$ Nm^{-1} was used as a liquid phase. The flow rate of oil-free compressed air was in the range of 0.01-0.11 ms^{-1}, and the range of reaction temperature was 110-150 °C. The reaction pressure was maintained at 92 kPa [Kang et al., 1995]. The solid content in the slurry phase was varied from 10 to 40 wt.% of the liquid phase. The fractional conversion of the reaction was determined by measuring the weights of reactants and products [Kang et al., 1995] at various reaction times.

The product was analyzed by means of DSC (Dupont, Model 990); the temperature elevation rate was 10 °C/min and the nitrogen flow rate was 30 cm^3/min. The crystal structure of the product was analyzed by means of an X-ray diffractometer (DIANO, Model XRD-800) whose scanning speed and threshold intensity were 5 deg/min and 127 cps, respectively. Surface characteristics of the products were analyzed by SEM (Jeol, Model JEM 200CX).

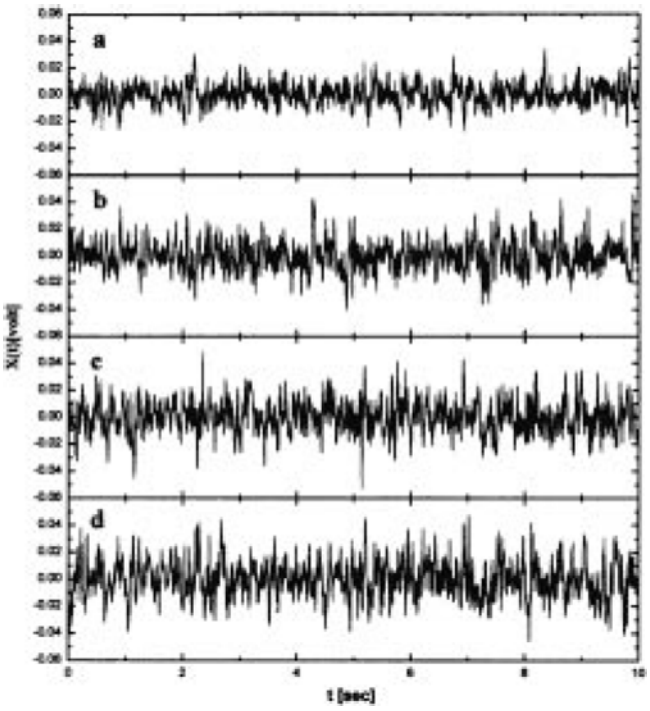

Fig. 2. Typical example of pressure fluctuations in a three-phase bubble column reactor (T=140 °C, t=40 min, P=92 kPa, U_G =0.10 m/s, d_p=0.153 mm, S=10 wt%, bubble distribution mode; a=even distribution; b=wall-side distribution; c= central distribution; d=asymmetric distribution).

RESULTS AND DISCISSION

1. Pressure Fluctuations in the Reactor

The resultant pressure fluctuations can be used in analyzing the hydrodynamics or bubbling phenomena in the multiphase flow reactors or contacting processes, since the fluctuations of pressure can be used as their state variables [Drahos et al., 1992; Kang et al., 1996a; Kikuchi et al., 1997]. To analyze the bubble distribution and obtain the effects of bubble distribution on the dehydration of ortho-boric acid, pressure fluctuations were measured and analyzed. Typical examples of pressure fluctuations can be seen in Fig. 2. It has been understood that the amplitude of pressure fluctuations corresponds to the bubble size, while their frequency is to the bubble frequency. In Fig. 2, the amplitude of signals increases but the frequency decreases in changing the gas distribution mode from even to wall-side, central or asymmetric distribution mode. This can be visualized more conveniently by means of the attractor in the phase space as can be seen in Fig. 3. Since the shape and position of the phase space portrait exhibits the dynamic behavior of the underlying system, the bubbling phenomena and bubble distribution can be explained from this figure. That is, the bubbling phenomena and bubble distribution in the reactor become more complicated and random in changing the gas distribution mode from even to wall-side, central or asymmetric, orderly, since the attractor becomes more scattered and complex with changing the gas distribution mode at the distributor.

The correlation dimension of pressure fluctuations has been calculated to elucidate the bubble distribution in the reactor more easily

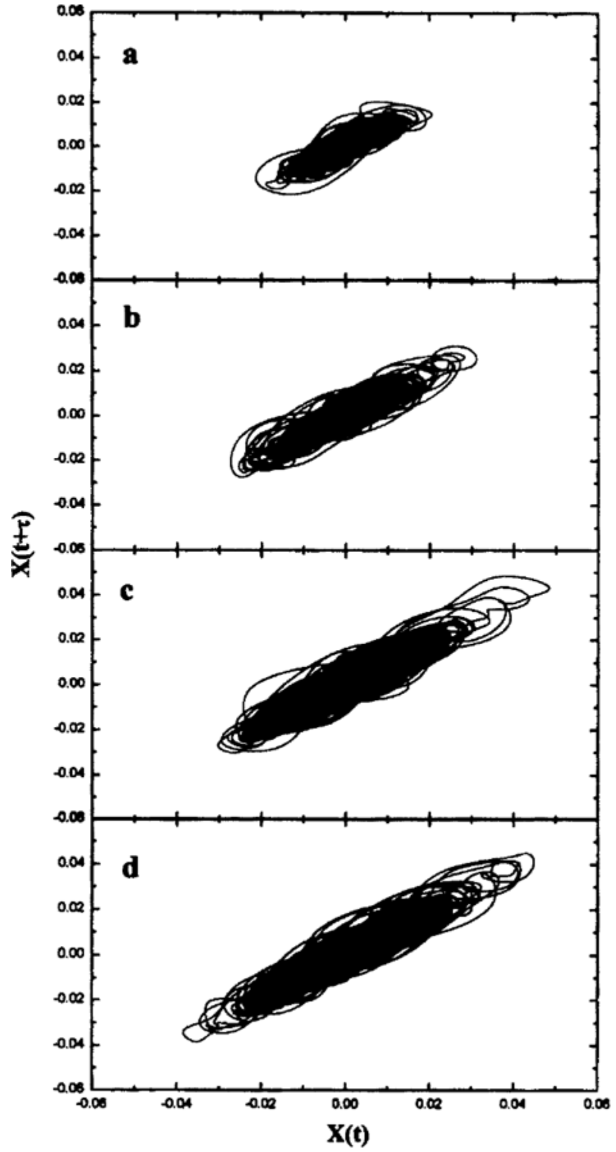

Fig. 3. Typical example of phase space portraits of pressure fluctuations (T=140 °C, t=40 min, U_G=0.10 m/s, P=92 kPa, d_P= 0.153 mm, S=10 wt%, bubble distribution mode; a=even distribution; b=wall-side distribution; c=central distribution; d=asymmetric distribution).

and quantitatively [Drahos et al., 1992; Kikuchi et al., 1997; Kang et al., 1999a, b, 2000]. The correlation dimension, D_C, has been determined by embedding the trajectories in the phase-space dimension. From the repeated calculation with different random samples, the attractor dimension converges when the estimate values are less than 2%. Five or six dimensions of embedding have been required in the space in order to capture the topological features of the attractor.

Effects of gas velocity and reaction temperature on the correlation dimension of pressure fluctuations can be seen in Fig. 4. In this figure, the value of D_C increases with increasing gas velocity or reaction temperature. This can be due to the increase of bubble holdup with increasing gas velocity or reaction temperature [Kang et al., 2000]. Note in this figure that the level of D_C value increases by changing the gas distribution mode from the even distribution. This reveals that the bubbling phenomena in the reactor become more complicated and irregular owing to the uneven gas distribution at the distributor. In other words, the bubble coalescence would increase and thus bubble size distribution would be broader in the reactor when the gas is injected at the distributor by means of wall-side, central or asymmetric distribution mode. This figure also shows that the asymmetric distribution is the worst one and the wall-side mode is better than the central distribution mode, for the better bubble distribution and bubbling phenomena in the bubble column reactor.

Effects of particle size of boric acid, d_P, on the correlation dimension of P-fluctuations can be seen in Fig. 5, at each gas distribution mode. In this figure, the D_C value is not influenced considerably by the particle size, since the d_P is very small (0.15-0.6 mm). Also, it is noted that the D_C value increases gradually in changing the gas distribution mode from even to wall-side, central and asymmetric, orderly.

2. Fractional Conversion of the Reaction

Effects of gas distribution at the distributor on the fractional conversion of the dehydration of ortho-boric acid can be seen in Fig. 6. In this figure, the conversion level is higher than 0.9 when the reaction time is longer than 40 min if gas distribution mode is even or wall-side. However, the conversion level is somewhat lower than 0.9 although the reaction time is longer than 40 min if gas is injected in a manner of central or asymmetric mode, especially when the solid content is increased. This means that the distribution of bubbles contacting with liquid and solid phase during the reaction and thus the fractional conversion of the reaction is closely related to the gas injection mode at the distributor. It is noted that the water vapor generated from the dehydration reaction can be removed effectively by means of rising bubbles when the bubbles are distributed in the reactor evenly. The conversion levels are comparable with those of the results of Kang et al. [1995] although they used a small-scale bubble column reactor (0.05 m ID · 1.0 m in height). Note that the X level decreases gradually by altering the gas distribution mode from even to wall-side, central and asymmetric modes, orderly, in all cases.

Effects of reaction temperature on the fractional conversion of the reaction can be seen in Fig. 7. In this figure, the conversion level decreases noticeably when the reaction temperature increases up to 145-150 °C, since the successive conversion reaction from meta-boric acid to anhydrous occurs at temperatures higher than 145 °C [Kodo et al., 1975]. On the other hand, the increase of temperature leads to the enhancement of vigorous bubbling in the reactor due to the increase of gas holdup as well as more ease of evaporation of water generated from the reaction (Fig. 4). These can result in the increase of conversion level of the reaction. Thus, the optimum reaction temperature would exist for this reaction. As can be seen in this figure, the optimum temperature of this reaction is 130-140 °C, which agrees well with the previous results [Kodo et al., 1975]. It is interesting to note that the conversion level at temperatures of 130-135 °C can be enhanced considerably in the bubble column reactor compared with that in the stirred tank reactor [Kodo et al., 1975]. The reason might be that the water vapor can be removed continuously by means of rising multi-bubbles in the bubble column reactor. Although the effects of temperature on the fractional

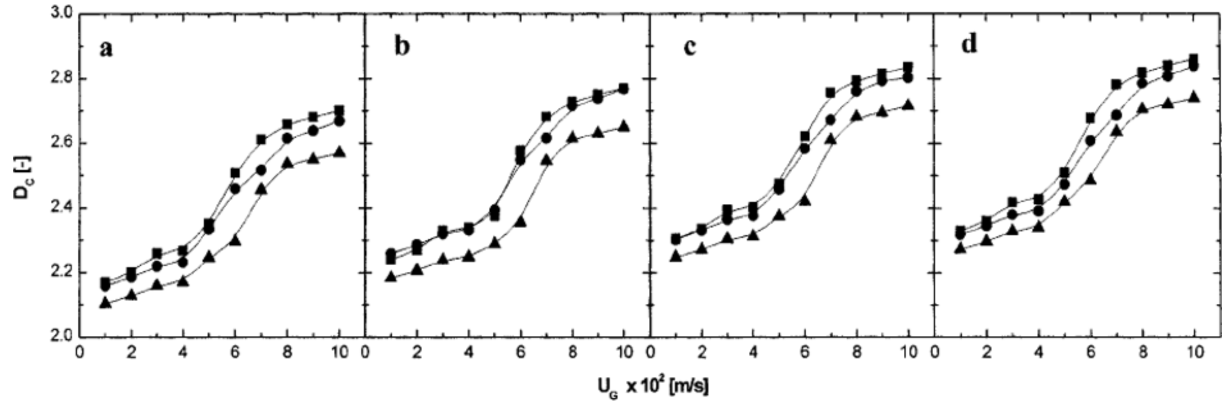

Fig. 4. Effects of U_G on the correlation dimension (D_C) of pressure fluctuations in a three-phase bubble column reactor (P=92 kPa, t=40 min, d_P=0.153 mm, S=10 wt%, bubble distribution mode: a=even distribution; b=wall-side distribution; c=central distribution; d=asymmetric distribution).
T [°C]: ▲ 100 ● 120 ■ 140

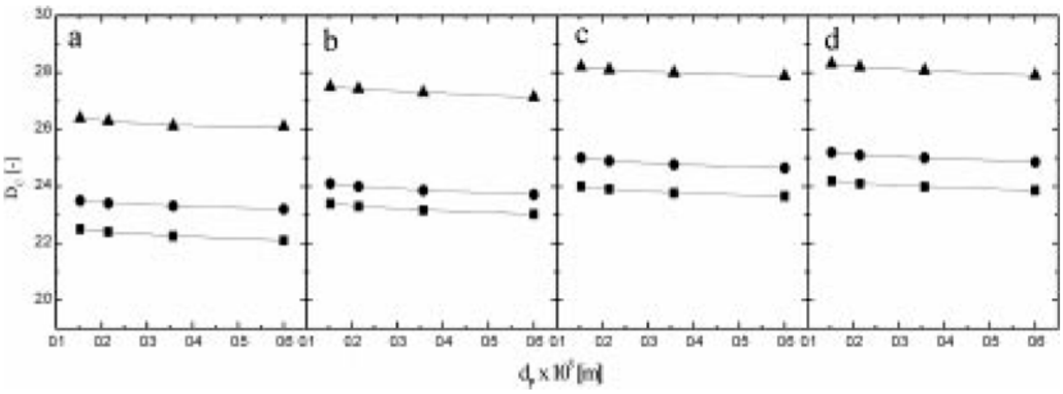

Fig. 5. Effects of d_P on the correlation dimension (D_C) of pressure fluctuations in a three-phase bubble column reactor (T=140 °C, t=40 min, P=92 kPa, S=10 wt%, bubble distribution mode: a=even distribution; b=wall-side distribution; c=central distribution; d=asymmetric distribution).
$U_G \cdot 10^2$ (m/s): ■ 0.05 ● 0.08 ▲ 0.10

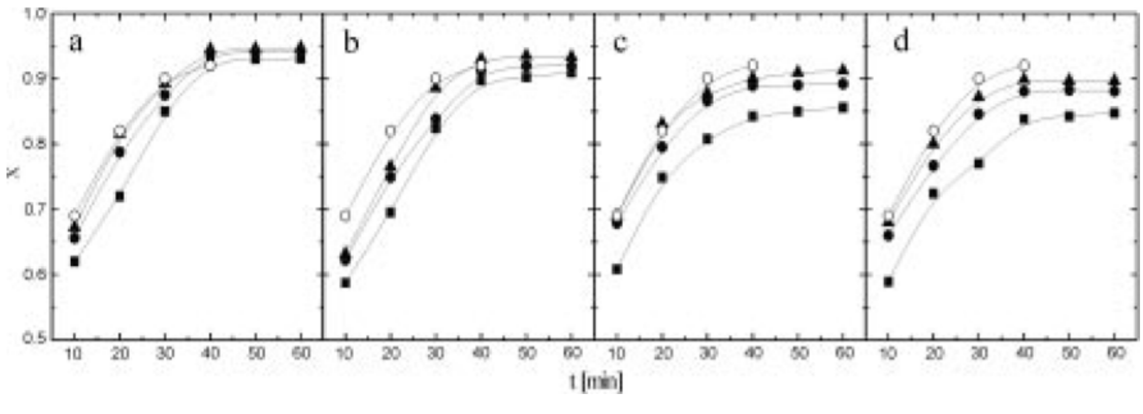

Fig. 6. Effects of reaction time (t) on the fractional conversion (X) of the reaction in a three-phase bubble column reactor (T=140 °C, P=92 kPa, U_G=0.10 m/s, d_P=0.153 mm, bubble distribution mode: a=even distribution; b=wall-side distribution; c=central distribution; d=asymmetric distribution).
S (wt%): ▲ 10 ● 15 ■ 25
Kang et al.: ○ D=0.05 m, T=130 °C, P=92 kPa, S=28 wt%, d_P=0.153 mm, U_G=0.07 m/s

conversion in the reactor are similar, the conversion level in case of gas distribution mode is central or asymmetric is lower than that of the even distribution mode, because the distribution and contacting efficiency of bubbles with other phases are changing in each case in the reactor (Fig. 7). The conversion level tends to decrease with increasing the deviation from the even distribution of bubbles in

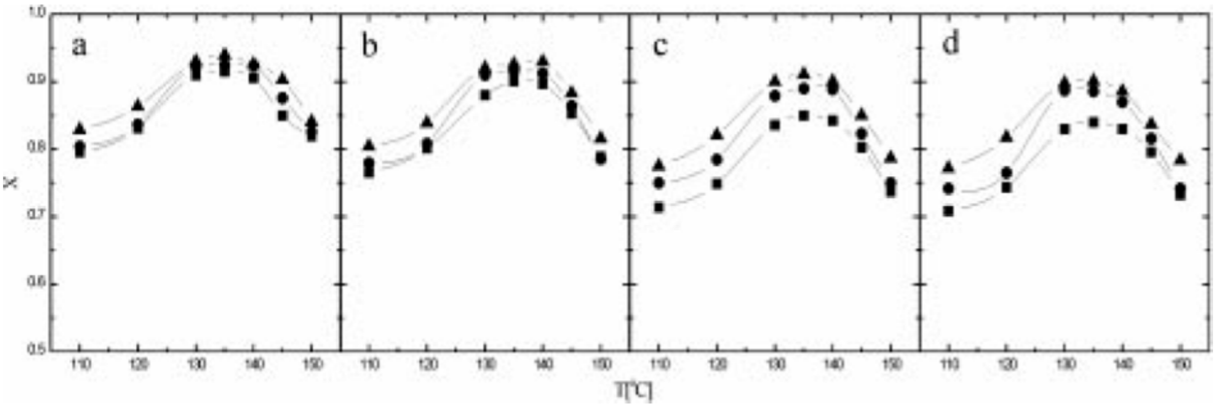

Fig. 7. Effects of temperature (T) on the fractional conversion (X) in a three-phase bubble column reactor (t=40 min, P=92 kPa, U_G= 0.10 m/s, d_P=0.153 mm, bubble distribution mode: a=even distribution: b=wall-side distribution: c=central distribution: d=asymmetric distribution).
S (wt%): ▲ 10 ● 15 ■ 20

the reactor owing to the similar reasons mentioned above.

Effects of gas velocity (U_G) on the fractional conversion of the reaction can be seen in Fig. 8. In this figure, the value of X increases with U_G in all cases, because the mixing and contacting among multiphase (gas-liquid-solid) in the reactor become more effective with increasing U_G due to the increase of gas holdup and vigorous bubbling phenomena. Compared with the results of Kang et al. [1995], the X level is still comparable in the case of even distribution mode

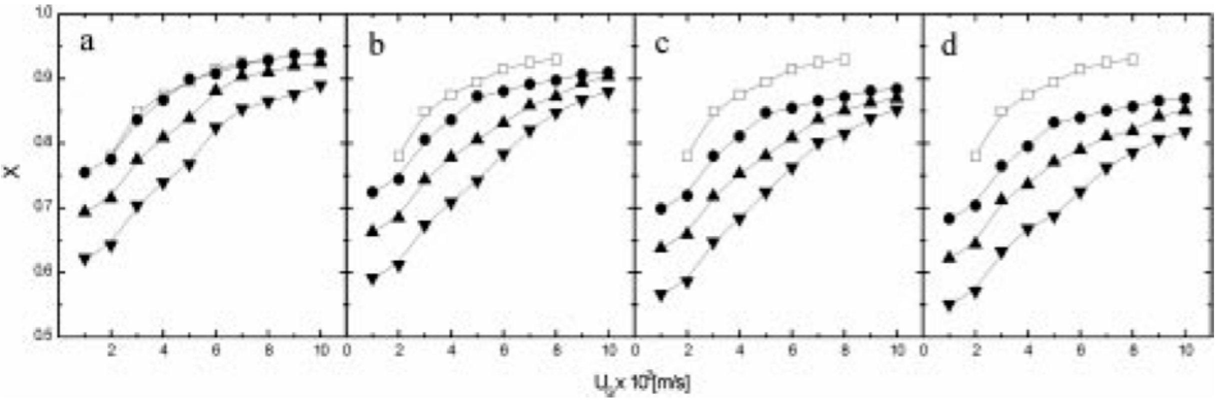

Fig. 8. Effects of U_G on the fraction conversion (X) in a three-phase bubble column reactor (T=140 °C, P=92 kPa, t=40 min, S=10 wt%, bubble distribution mode: a=even distribution: b=wall-side distribution: c=central distribution: d=asymmetric distribution).
d_P (mm): ● 0.153 ▲ 0.357 ▼ 0.600
Kang et al.: D=0.05, T=130 °C, P=92 kPa, t=30 min, S=28 wt%, d_P=0.153 mm

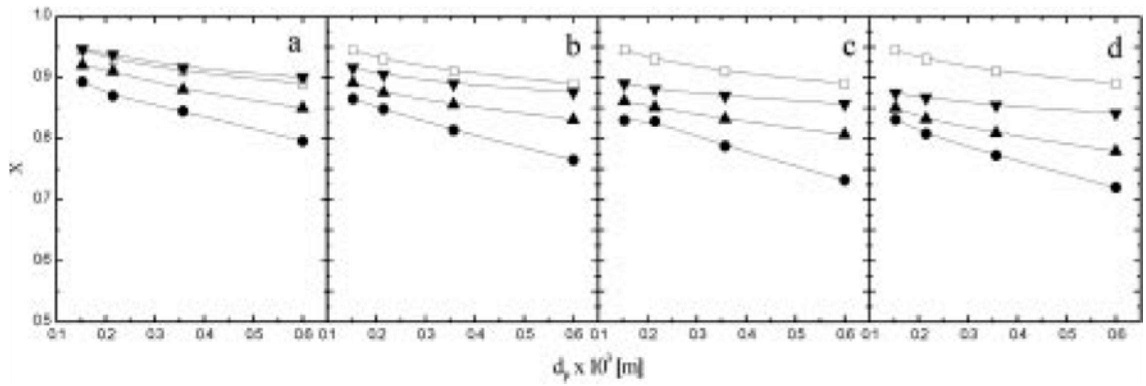

Fig. 9. Effects of d_P on the fractional conversion (X) in a three-phase bubble column reactor (T=140 °C, t=40 min, P=982 Pa, S=10 wt%, bubble distribution mode: a=even distribution: b=wall-side distribution: c=central distribution: d=asymmetric distribution).
$U_G \cdot 10^2$ (m/s): ● 0.153 ▲ 0.357 ▼ 0.600
Kang et al.: D=0.05 m, T=130 °C, P=92 kPa, t=30 min, S=28 wt%, U_G=0.07 m/s

of gas; however, when the gas is injected in a manner of wall-side, central or asymmetric mode, the X levels are somewhat lower than those of Kang et al. (Fig. 8). This figure also shows that the conversion level decreases gradually by altering the gas injection mode from even to wall-side, central and asymmetric modes, orderly, in all cases.

Effects of particle size (d_P) on the value of X can be seen in Fig. 9. Although the correlation dimension of pressure fluctuations does not change considerably (Fig. 5), the X value decreases with increasing d_P. This reveals that this reaction could be controlled by the intraparticle diffusion, since the water molecule inside the Ortho-Boric acid has to be transferred to the surface of the particle to be removed by means of bubbles with existence of n-paraffin around the particle.

Based on the fluid-solid heterogeneous reaction model [Levenspiel, 1972], the relation between the reaction time and the fractional conversion can be written as Eq. (6), for the diffusion inside the particle is rate-determining step.

$$\frac{t}{\tau}=1-3(1-X)^{2/3}+2(1-X) \quad (6)$$

$$\text{where } \tau=\frac{\rho_B R^2}{M_B C_P}\left(\frac{1}{K_S}+\frac{1}{3K_m}+\frac{R}{6D}\right) \quad (7)$$

As can be seen in Fig. 10, the experimental results of this study are well fitted to Eq. (6). It can be noted in this figure that when the gas distribution deviates from the even distribution the fractional conversion also deviates from the fitting of Eq. (6). This enables us to state that other factors such as the mass transfer through the thin film of n-paraffin around the boric acid particle would affect the determination of X level when the bubbles are not distributed evenly during the reaction in the reactor. The deviation of experimental results from the model equation increases when the gas distribution mode at the distributor is changed from even to wall-side, central and asymmetric distribution, orderly.

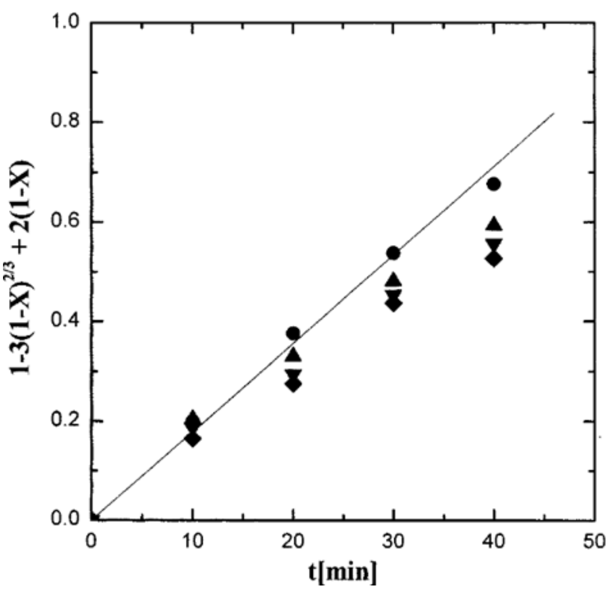

Fig. 10. Plots of the limiting case of diffusion control of the fluid-solid heterogeneous reaction model (T=140 °C, P=92 kPa, S=10 wt%, d_P=0.153 mm, U_G=0.10 m/s). Bubble distribution mode: ● even, ▲ wall-side, ▼ central, ◆ asymmetric.

CONCLUSION

The possibility that a three-phase bubble column can be applicable as a reactor for obtaining meta-Boric acid effectively has been examined by considering the effects of gas injection mode on the bubble distribution and bubbling phenomena and thus on the fractional conversion of the reaction. The effects of gas injection mode on the fractional conversion of the reaction increase with increasing the gas velocity and reaction time. The optimum temperature of this system has been in the range of 130-140 °C to maintain the higher fractional conversion. The complicated bubble distribution and bubbling phenomena in the reactor have been well diagnosed and interpreted by means of the attractor trajectories and correlation dimension from the resultant pressure fluctuations during the reaction. The bubble distribution and bubbling phenomena become more complicated and irregular in altering the gas injection mode from even to wall-side, central and asymmetric distribution, orderly. The level of fractional conversion decreases gradually with increasing the deviation from the even distribution of bubbles in the reactor. The heterogeneous fluid-solid reaction model can be applicable to this reaction; however, the deviation from this model prediction is still considerable when the gas injection mode at the distributor is central or asymmetric.

The results of this study can be utilized for developing the three-phase bubble column reactor to obtain the meta-Boric acid that is widely used in esterification.

NOMENCLATURE

C(r) : correlation integral [-]
C_P : concentration of paraffin at the bulk region [mol/dm^3]
D : column diameter of the reactor [m]
D_C : correlation dimension [-]
d_P : particle diameter [mm]
H : heavyside function [-]
K_m : mass transfer coefficient [m/s]
K_S : reaction rate constant [m/s]
M_B : molecular weight of particle [kg]
P : pressure [kPa]
r : radius of hypersphere [-]
S : solid content [wt. %]
t : time [s]
U_G : gas velocity [m/s]
X : fractional conversion [-]
X(t) : time series signals of pressure fluctuations [V]
Z_i : the vector time series defined as in Eq. (1)

Greek Letters

τ : time delay [s]
ρ : density of liquid [kg/m]
ρ_B : density of particle [kg/m]

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