# Pressure fluctuations and bubble size in viscous three-phase circulation fluidized bed bioreactors

Sung Mo Son, Ik Sang Shin, Suk Hwan Kang, Yong Kang† and Sang Done Kim\*

School of Chemical Engineering, Chungnam National University, Daejeon 305-764, Korea \*Department of Biomolecular & Chemical Engineering, KAIST, Daejeon 305-701, Korea (Received 9 August 2006 • accepted 1 February 2007)

Abstract–Characteristics of pressure fluctuations and bubble size were investigated in the riser of a three-phase cir-<br>ulation fluidized bed bioreactor with viscous liquid medium, whose diameter is 0.102 m (ID) and 3.5 m culation fluidized bed bioreactor with viscous liquid medium, whose diameter is 0.102 m (ID) and 3.5 m in height. Effects of gas (0.01-0.07 m/s) and liquid (0.17-0.23 m/s) velocities and liquid viscosity (0.96-38 mPa·s) on the bubble size in the riser were examined. The bubbling phenomena in the bioreactor with viscous liquid medium were interpreted effectively by measuring and analyzing the pressure fluctuations by adopting chaos theory. The bubble size increased with increasing gas velocity or liquid viscosity, but decreased with increasing liquid velocity. The bubbling phenomena became more complicated and bubble size distribution tended to broad, with increasing gas velocity or liquid viscosity. The bubble size was well correlated in terms of correlation dimension of pressure fluctuations as well as dimensionless groups within these experimental conditions.

Key words: Pressure Fluctuations, Viscous Bioreactor, Bubble Size, Three-phase Circulation, Chaos Analysis

#### INTRODUCTION

For the practical applications of three-phase fluidized beds, a threephase circulation fluidized bed has been proposed, since it can be utilized successfully as a reactor or a contactor when small or light particles and suspensions are fluidized in a viscous liquid medium, which is often encountered in environmental, biochemical and food processing engineering. The three-phase circulation fluidized bed can minimize the dead zone in the column as well as increase the contacting efficiency among gas, liquid and solid phases. These advantages can lead to considerable increases in the fractional conversion as well as production efficiency per unit cross-sectional area of the system [1-6]. Moreover, the deactivated catalyst, bio-media, ion exchange resin, or adsorbent can be regenerated continuously by means of the solid circulation mode between the main reactor or contactor (riser) and the regenerator (downcomer).

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Effects of gas (0.01-0.07)<br>
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with increasing gas the chaos theory. That is, the conglomerating effects of multiple dynamic phenomena such as bubbling behavior in multiphase flow It is understood that the information on the flow and bubbling behaviors of gas phase is known to a play critical role in determining the oxygen transfer in the bioreactors, since the bubbles which exist as a dispersed phase are flowing stochastically and randomly in the viscous liquid medium [7-13]. However, little attention has been focused on the bubbling phenomena in three-phase circulation fluidized bed bioreactors [14-16], especially in reactors with a viscous liquid medium such as wastewater from food processing industries. Recently, the complicated nonlinear multiphase flow and contact phenomena have been successfully described by adopting systems can be analyzed conveniently by means of chaotic parameters such as attractor or dimension [17-21].

In the present study, thus, pressure fluctuations and bubbling phe-

nomena in the riser of three-phase circulation fluidized bed bioreactor with viscous liquid medium were investigated, for the application of this scheme to treat the ammonium component in the wastewater from food processing industries by means of microorganism. The bubble size was measured and dynamic bubbling behaviors in the riser were analyzed by means of the phase space portraits and correlation dimension of the time series of pressure fluctuations.

## ANALYSIS

## 1. Phase Space Portraits

The multidimensional phase space portraits can be constructed from the time series of pressure fluctuations by means of the time delay method [22]. That is, the experimentally obtained time-series signal,  $X(t)$ , is digitized with a time step of Dt; the resultant  $(m+1)$ values of the signal,  $X(i·∆t)$ , are stored for i=0, 1, 2, ..., m. Thus, the vector time series is defined as II<br>Ima<br>Ilan<br>2, the multidimensional phase space portrait<br>(the time series of pressure fluctuations by method [22]. That is, the experimentally<br>al, X(t), is digitized with a time step of D<br>es of the signal, X(i⋅∆t), are stored for i=<br>v ie time series of pressure fluctuations by means of the time<br>nethod [22]. That is, the experimentally obtained time-series<br>X(t), is digitized with a time step of Dt; the resultant (m+1)<br>of the signal, X(i⋅∆t), are store

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

where,  $\tau$ =k· $\Delta t$ , 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 phasespace dimension of the reconstructed trajectory of attractor. Actually, if  $\tau$ =1 and p=3, the state vectors presenting one orbit of attractor are constructed from the experimentally measured time-series signals as  $Z_1 = [X_1, X_2, X_3]'$ ,  $Z_2 = [X_2, X_3, X_4]'$ , etc. Thus, the number of elements, p, of the state vector is equal to the number of coordinates in the reconstructed state space. In this way a two-dimensional reconstructed attractor of a time series can be obtained.

#### 2. Correlation Dimension

To estimate the correlation dimension of the time series  $X(t)$ , the trajectories of them reconstructed by resorting to time embedding

To whom correspondence should be addressed. E-mail: kangyong@cnu.ac.kr

$$
C(r)
$$
\n
$$
= \lim_{m \to \infty} \frac{1}{m^2} \left[ \text{number of pairs (i,j) whose distance } |Z_i(t) - Z_j(t)| < r \right].
$$
\n
$$
(2)
$$
\n
$$
C(r) = \lim_{m \to \infty} \frac{1}{m^2} \sum_{i=1}^{m} \sum_{j=1}^{m} H[r - |Z_i(t) - Z_j(t)|], \ i \neq j
$$
\n
$$
(3)
$$

Formally,

rmally,

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$$
C(r) = \lim_{m \to \infty} \frac{1}{n^2} \sum_{i=1}^{m} \sum_{j=1}^{m} H[r - |Z_i(t) - Z_j(t)|], \ i \neq j
$$
\n(3)

\nhere m is the number of data points, and H is Heavyside function,

\n
$$
H[r - |Z_i(t) - Z_j(t)|] = \begin{cases} 1 & \text{if } r > |Z_i(t) - Z_j(t)| \\ 0 & \text{otherwise} \end{cases} \tag{4}
$$

where m is the number of data points, and H is Heavyside function,

$$
C(r) = \lim_{m \to \infty} \frac{1}{m^2} \sum_{i=1}^{m} \sum_{j=1}^{m} H[r - |Z_i(t) - Z_j(t)|], i \neq j
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$$
(4)  
ne correlation integral, C(r), has been found to be a power func-  
no of r for small r's:  
C(r)=kr<sup>0</sup> (5)

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

$$
C(r) = kr^{Dc} \tag{5}
$$

The slope of the plot of ln C(r) vs. ln r is an estimate of  $D<sub>c</sub>$ , which is termed as the correlation dimension, for the given embedded space dimension, p.



#### Fig. 1. Experimental apparatus.

- 1. Riser 10. Low-pass filter
- 2. Down comer 11. A/D converter
- 
- 
- 5. Butterfly valve 14. Compressor
- 6. Resistivity probe 15. Control valve
- 7. Pressure transducer 16. Flow meter
- 8. Static pressure sensor 17. Pump
- 
- 3. Hopper 12. Computer 4. L/S separator 13. G/L distributor
	-
	-
	-
	-
- 9. Amplifier 18. Liquid reservoir

# EXPERIMENTAL

Experiments were carried out in the riser of a three-phase circulation fluidized bed bioreactor which is composed of three main sections, as can be seen in Fig. 1 [15,16]: the riser column, gas-liquidsolid separator and solid recycle device. The diameter and height of the riser was 0.102 m ID and 3.5 m, respectively. A glass bead whose diameter is  $1.0 \text{ mm } (\rho_s = 2,500 \text{ kg/m}^3)$  was used as a fluidized solid particle, and compressed filtered air was used as a gas phase, respectively. The solid particles were returned to the bottom of the riser through the solid recycle device. The solid circulation rate was  $2.0 \text{ kg/m}^2$  sec, which was determined by measuring the amounts of solid piled up above the butterfly valve in the solid recycle device [4,15,16]. The synthesized wastewater, whose compositions are summarized in Table 1, with aqueous solutions of carboxymethylcellulose (CMC), were used as the continuous liquid medium. Recycling activated sludge, which was prepared from a wastewater treatment facility in Wonchun Dong, Daejeon, Korea, was employed as a source of microorganisms. The density and surface tension of the liquid medium were in the range of 1,000-1,003 kg/m<sup>3</sup> and  $72.9 \times 10^3$ -73.6 $\times 10^3$  N/m, respectively. The characteristics of the pseudoplastic behavior of wastewater were determined by a Brookfield synchrolectric rotational viscometer. In addition, the flow consistency index  $(K')$  and flow behavior index  $(n)$  of CMC solution are summarized in Table 2.

The bubble size was measured by means of dual electrical resistivity probe system [4,15,24]. The probe applied by 1.75 V DC detected the difference in conductivity of bubble and liquid. The dual electrical resistivity probe, which was installed at 0.9 m from the inlet of solid recycle device, consisted of two 7 mm diameter stainless steel pipes coated with epoxy resin. The vertical distance between the tips of the probe was 2 mm. The probe was located at the center between the wall and the center of the column. The tips of the probe, which are made of platinum wire, had a diameter of

Table 1. Composition of the concentrated synthetic wastewater

Concentration $(g/L)$	Component
5.6	Glucose
9.7	CH <sub>3</sub> COONa 3H <sub>2</sub> O
1.32	KH <sub>2</sub> PO <sub>4</sub>
0.006	FeCl, $6H, O$
0.075	CaC <sub>l</sub>
2.8	$(NH_4)$ , $SO_4$
1.0	$MgSO4$ 7H <sub>2</sub> O
0.1098	MnSO <sub>4</sub> ·H <sub>2</sub> O
2.1	NaHCO <sub>2</sub>

## Table 2. Liquid physical properties and fluids flow rate range



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0.2 mm. The analog signals obtained from each probe circuit were processed off-line to produce the digital data. The preselected sampling rate at the personal computer with the DT2805 Lab Card was 500 Hz and the total sampling time was 15 s. The bubble size was calculated from the relationship between the reformed and digitized probe signals and the bubble dwell and lag times [11,15]. To avoid noise as well as small disturbances owing to solids, a lowpass filter was used in taking the signals [15,16].

The static and dynamic pressures were measured by means of pressure sensors. A pressure tap for measuring the dynamic pressure fluctuations was mounted flush with the wall of the column in the test section which is located at 1.0 m above the inlet of solid recycle device. To measure the static pressure drop in the reactor, 10 pressure taps were installed at the wall of the column with the intervals of 0.2 m. The pressure sensors were of semiconductor type (Coppel electronics) that have enough fast response time to measure the dynamic pressure fluctuations in the test section. The output voltage from the pressure transducer, which is proportional to the pressure fluctuations or static pressure, was processed by means of a data acquisition system (Data Precision Model, DT3001) and a personal computer. The voltage-time signals, corresponding to the pressure-time signals, were sampled at a rate of 0.005 sec and stored in the data acquisition system. The total acquisition time was 15

sec having 3,000 data points. This combination of sampling rate and time could detect the full spectrum of hydrodynamic signals (200 Hz) in this three-phase circulation flow system [15,16].

## RESULTS AND DISCUSSIONS

Typical example of pressure fluctuations in the riser with viscous liquid medium can be seen in Fig. 2. Note that the signals become more irregular and complicated, and the amplitude increases with increasing liquid viscosity. The energy of the periodic behavior in the system can be expressed as the amplitude of pressure fluctuations. Since the density of bubbles is quite different from that of liquid or solid phase, the periodic behavior in the three-phase circulation fluidized-bed reactor can be mainly due to the flow behavior of bubbles. These signals, thus, have been analyzed to examine the bubbling behavior in the riser of the reactor. In Fig. 2, the amplitude of signals tends to increase with increasing viscosity of liquid medium, which implies that the bubble size,  $D_b$ , increases with increasing  $\mu_b$ .

The behavior of pressure fluctuations can be visualized by means of an attractor in the phase space. The phase space portraits of pressure fluctuation signals have been constructed with suitable time lag. The optimum value of time lag has been chosen corresponding to the first minimum in the mutual information function [15,22].







Fig. 3. Phase space portrait of pressure fluctuations in three-phase circulation fluidized bed bioreactors with viscous liquid medium ( $U_l$ =0.17 m/s).

Effects of liquid viscosity  $(\mu_l)$  on the strange attractor of pressure fluctuations can be seen in Fig. 3. The attractor becomes more scattered and complex with increasing  $\mu$ . In comparing the traces of pressure fluctuations with bubble size, it is noted that the scattered traces can be caused by large bubbles, while the small or concentrated traces are smaller bubbles, since the size, location and character of the loops in the trajectories are related directly to the underlying systems with their physical events.

For the quantitative measurement of bubbling behavior in the riser, the correlation dimension of pressure fluctuations has been calculated [23]. The attractor dimension, which can be determined by embedding the trajectories in the phase space, essentially con-



Fig. 4. Typical example of correlation analysis of pressure fluctuations (U<sub>G</sub>=0.01 m/s U<sub>L</sub>=0.21 m/s  $\mu$ <sub>L</sub>=0.096 mPa·s).



Fig. 5. Effects of  $\mu_l$  on the correlation dimension of pressure fluctuations in three-phase circulation fluidized bed bioreactors with viscous liquid medium ( $U_L$ =0.21 m/s).

verges with increasing embedding dimension (Fig. 4). From the repeated calculation with different random samples, it has been indicated that the attractor dimension converges when the estimated values are less than 2%. Five or six dimensions of p have been required in the embedding space in order to capture the topological features of the attractor. Effects of liquid viscosity and gas velocity on the correlation dimension of pressure fluctuations can be seen in Fig. 5. In this figure, the  $D<sub>C</sub>$  value increases with increasing  $\mu<sub>L</sub>$  or  $U<sub>G</sub>$ , implying that the resultant bubbling behavior becomes more irregular and chaotic with increasing  $\mu_l$  or  $U_G$  in the reactor. This can be mainly due to the variations of bubble size and distribution in the reactor.

The output signals measured by the resistivity probes have been reformed and digitized to produce the digital data from which the bubble size has been calculated [15,16]. The mean value of bubble size has been determined from the logarithmic normal distribution of bubble size, which can be seen in Fig. 6. In this figure, the distribution of bubble size becomes broad and its mean value tends to increase with increasing  $U_G$  (Fig. 6A) or  $\mu_l$  (Fig. 6B). It can be anticipated that the bubble holdup and its frequency increases, which results in the increase of probability of bubble coalescence; thus, the size distribution becomes broad, with increasing  $U_G$ . It has been understood that the bubble size becomes increased with increasing  $\mu_l$  owing to the bubble coalescence in the beds of viscous liquid medium [1,7,8]. The rising behavior of bubbles would be restricted in beds of viscous liquid medium. This can lead to an increase of



Fig. 6. Probability density of bubble size in three-phase circulation fluidized bed bioreactors with viscous liquid medium (A:  $U_L$ =0.23 m/s,  $\mu_l$ =11 mPa·s, B:  $U_G$ =0.01 m/s,  $U_L$ =0.19 m/s).

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bubble coalescence in a given gas velocity and amount in a reactor.

Effects of  $U_G$  on the bubble size can be seen in Fig. 7. In this figure, the bubble size increases with increasing gas velocity. This is due to the increase of gas holdup and frequency with increasing the throughput of gas phase into the riser. It is interesting to note that the bubble size is smaller in the beds with solid circulation than that in the beds of conventional three-phase fluidization [8,15]. This can be attributed to the fact that the turbulence in the bed would increase with increasing liquid velocity, since the  $U<sub>i</sub>$  value is much higher in the riser of a circulating fluidized bed than that in the conventional three-phase bed. Actually, the bubble size decreases consid-



Fig. 7. Effects of  $U<sub>c</sub>$  on the bubble size in three-phase circulation fluidized bed bioreactors with viscous liquid medium ( $\mu$ = 24 mPa·s).



Fig. 8. Effects of  $U_L$  on the bubble size in three-phase circulation fluidized bed bioreactors with viscous liquid medium ( $U<sub>G</sub>$ = 0.05 m/s).

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erably with increasing liquid velocity in the riser with viscous liquid medium, as can be seen in Fig. 8. It has been understood that the bubble flow regime in the beds of 1.0 mm glass beads belongs to the bubble coalescence regime. In other words, the bubble coalescence in the beds is significant owing to the hindrance effects of fluidized particles on the rising bubbles [1,7,8]. But, the increase of  $U<sub>t</sub>$  results in the increase of rising velocity of bubbles, which leads to a decrease of probability of bubble coalescence in a given amount of bubbles. In addition, the increase of liquid velocity can lead to an increase of turbulence generated in the reactor due to the contacting among gas, liquid and solid phases. Thus, turbulence can act to break down the larger bubbles in the reactor. This consequently results in a decrease of bubble size in the beds.

The bubble size increases gradually with increasing  $\mu$  as can be seen in Fig. 9. It has been known that the increase of  $\mu$ <sub>L</sub> leads to an increase of bubble coalescence [1,8]; thus the bubble size increases with increasing  $\mu$ . However, it can be noted that the increasing trend of  $D_b$  with increasing  $\mu_l$  in the riser of circulation fluidized beds is lower than that in conventional three-phase fluidized beds. This can also be due to the effective turbulence arising from the higher liquid velocity, enough to split the large bubbles in the beds. The bubble size was directly related to the bubbling behavior, which can be represented in terms of pressure fluctuations; thus, it was well correlated with the value of correlation dimension of pressure fluctuations as Eq.  $(6)$ , with a correlation coefficient of 0.940. In addition, the bubble size was well correlated in terms of dimensionless groups as Eq. (7) based on isotropic turbulence theory, with a correlation coefficient of 0.923. essented in terms of pressure fluctuations; thus, it was well corre-<br>ed with the value of correlation dimension of pressure fluctuations<br>Eq. (6), with a correlation coefficient of 0.940. In addition, the<br>bble size was wel iq.<br>ble<br>fic<br><sub>b</sub>=<br><u>D,</u>  $\alpha$ d<sub>e</sub><br>d<sub>e</sub><br>d<sub>e</sub> Eq. (6), with a corre<br>
bble size was well cc<br>
Eq. (7) based on isot<br>
efficient of 0.923.<br>  $D_b = 0.084D_c^{2.462}$ <br>  $\left(\frac{D_b}{d_p}\right) = 36.97 \left(\frac{U_a}{U_L + U_a}\right)$ ted in turb $\frac{\mu_L}{D_{\nu}\rho_L}$ 

$$
_{b}=0.084D_{C}^{2.462}
$$
 (6)

$$
\left(\frac{D_b}{d_p}\right) = 36.97 \left(\frac{U_G}{U_L + U_G}\right)^{0.249} \left(\frac{\mu_L}{D_p \rho_L}\right)^{0.037} \tag{7}
$$

# **CONCLUSION**



Fig. 9. Effects of  $\mu$  on the bubble size in three-phase circulation fluidized bed bioreactors with viscous liquid medium ( $U_L$ = 0.19 m/s).



Fig. 10. Relation between the bubble size and the value of correlation dimension of pressure fluctuations.

The bubbling behavior in three-phase circulation fluidized bed bioreactors with viscous liquid medium was successfully interpreted by analyzing the pressure fluctuations and bubble size. The bubble size increased with increasing  $U_G$  or  $\mu_L$ , but decreased with increasing  $U_l$ . The uniformity and persistency of bubbling behavior in the ing  $U_L$ . The uniformity and persistency of bubbling behavior in the riser was related directly to the bubble size; the correlation dimension of pressure fluctuations increased with increasing  $U_L$ .<br>
The bubble size was w sion of pressure fluctuations increased with increasing  $U_G$  or  $\mu_L$ , but decreased with increasing  $U_L$ .

The bubble size was well correlated with correlation dimension of pressure fluctuations as well as dimensionless groups as

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D<sub>b</sub>=0.084D<sub>c</sub><sup>2,462</sup>  

$$
\left(\frac{D_b}{d_p}\right) = 36.97 \left(\frac{U_G}{U_L+U_G}\right)^{0.249} \left(\frac{\mu_L}{D_F\rho_L}\right)^{0.037}
$$

## **NOMENCLATURE**

- $C(r)$ : correlation integral
- $D_C$  : correlation dimension [-]<br> $D_b$  : bubble size [mm]
- 
- $D_b$  : bubble size [mm]<br>d<sub>n</sub> : particle diameter  $d_p$  : particle diameter [mm]<br> $D_v$  : diffusivity [m<sup>2</sup>/s]
- $D_v$  : diffusivity  $[m^2/s]$
- H : heavyside function
- K : constant
- $K'$  : flow consistency index [Pa·s<sup>n</sup>]
- m : number of data point
- n : flow behavior index
- p : embedded dimension
- r : radius of hypersphere
- t : time [s]
- 
- $U_G$  : gas velocity [m/s]<br> $U_L$  : liquid velocity [m.
- $U_L$  : liquid velocity [m/s]<br> $X(t)$  : time series of pressu  $:$  time series of pressure fluctuations  $[V]$
- $Z_i$  : the vector time series defined as Eq. (1)

#### Greek Letters

- $\rho$  : density [kg/m<sup>3</sup>]
- $\varepsilon$  : holdup [-]<br>  $\tau$  : time delay
- $\tau$  : time delay [s]<br>  $\mu$  : viscosity [Pa·
- : viscosity [Pa·s]

## **Subscripts**

- G : gas phase
- L : liquid phase
- S : solid phase

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