

INVESTIGATION OF HYDRODYNAMIC PARAMETERS IN A NOVEL EXPANDED BED CONFIGURATION: LOCAL AXIAL DISPERSION CHARACTERIZATION AND AN EMPIRICAL CORRELATION STUDY

E. S. Taheri^{1,2}, M. Jahanshahi^{1*}, M. T. Hamed Mosavian³ and M. H. Shahavi¹

¹Nanotechnology Research Institute, Faculty of Chemical Engineering,
Phone/Fax: + 98-111-3220342, Babol University of Technology,
Babol, P.O.Box: 484, Babol, Iran.

*E-mail: mjahan@nit.ac.ir; mmohse@yahoo.com

²Chemical Engineering Department, Quchan Branch, Islamic Azad University, Iran.

³Faculty of Chemical Engineering, Ferdowsi University of Mashhad, Mashhad, Iran.

(Submitted: December 30, 2011 ; Revised: March 27, 2012 ; Accepted: April, 6, 2012)

Abstract - Study of liquid behavior in an expanded bed adsorption (EBA) system is important for understanding, modeling and predicting nanobioproduct/biomolecule adsorption performance in such processes. In this work, in order to analyze the local axial dispersion parameters, simple custom NBG (Nano Biotechnology Group) expanded bed columns with 10 and 26 mm inner diameter were modified by insertion of sampling holes. Based on this configuration, the particles and liquid can be withdrawn directly from various axial positions of the columns. Streamline DEAE particles were used as solid phase in this work. The effects of factors such as liquid velocity, viscosity, settled bed height and column diameter on the hydrodynamic parameters were investigated. Local bed voidages in different axial bed positions were measured by a direct procedure within the column with 26 mm diameter. Increasing trend of voidage with velocity at a certain position of the bed and with bed height at a certain degree of expansion was observed. Residence time distribution (RTD) analysis at various bed points showed approximately uniform hydrodynamic behavior in the column with 10 mm diameter while a decreasing trend of mixing/dispersion along the bed height at a certain degree of expansion was seen in the column with 26 mm diameter. Also lower mixing/dispersion occurred in the smaller diameter column. Finally, a combination of two empirical correlations proposed by Richardson-Zaki and Tong-Sun was successfully employed for identification of the bed voidage at various bed heights (RSSE=99.9%). Among the empirical correlations presented in the literatures for variation of the axial dispersion coefficient, the Yun correlation gave good agreement with our experimental data (RSSE=87%) in this column.

Keywords: EBA chromatography; Local hydrodynamic behavior; Local bed voidage; Local axial liquid dispersion; Empirical correlation.

INTRODUCTION

Expanded bed adsorption (EBA) is an innovative and special chromatography technique for the recovery, separation and purification of target nano/biomolecules directly from complex unclarified

feed stocks without elimination of suspended solid. On the other hand, EBA is a single-unit that combines several unit operation steps such as concentration, centrifugation, micro-filtration and other prior clarification steps. Therefore, this method has advantages over other conventional separation

*To whom correspondence should be addressed
Homepage: <http://nano.nit.ac.ir/IndexEn.asp>

procedures such as decreasing the time of operation and total production costs and increasing product quality and process yield (Pinotti *et al.*, 2009; Ng *et al.*, 2007; da Silva Padilha *et al.*, 2009; Vennapusa *et al.*, 2009; Tong *et al.*, 2002). Analysis of adsorption performance is quite complex because of influential factors, namely fluid hydrodynamics, particle classification, and desired component transfer. It is very important and fundamental to understand the flow hydrodynamic characteristics and interpret the adsorption performance of nano/biomolecules in EBA systems (Moraes *et al.*, 2009; Li *et al.*, 2004). In expanded beds, the adsorbent particles that are employed have a wide size/density distribution. When equilibrium conditions occur, the forces acting on a single particle are in balance and each particle locates at its own axial position without upward and downward movement and particle mixing is weak. Therefore, particles are classified perfectly and variation of the mean particle size and local bed voidage along the bed height are established in an EBA column. This trend lead to inhomogeneous hydrodynamics and adsorption behavior along the bed, while a liquid-solid fluidized bed, particles have a narrow size distribution and each particle can locate at any positions of the bed and cause intensive particle and liquid mixing (Yun *et al.*, 2004). A number of experimental and modeling studies have been performed to determine the variation of the axial distribution characteristics such as mean particle size/density, bed voidage, axial dispersion coefficient and dynamic capacity with bed height within an EBA system. Bruce and Chase (2001) employed in-bed liquid and particle sampling procedures to estimate the variation of liquid hydrodynamic behavior in an expanded bed system within a modified STREAMLINE column for Streamline SP and DEAE adsorbent particles. They found that a classification of the particles occurs and particles with larger size/higher density were located at the bottom regions and smaller size/lower density were near the top regions of the column. In addition, residence time distribution (RTD) analysis was used in different column zones to the determine bed voidage and Bodenstein number along the bed height. Their experimental results showed that the bed voidage and Bodenstein number increase with an increase of column height. On the other hand, axial mixing and dispersion are much lower in the top zones than in the bottom zones (Bruce and Chase, 2001). Yun *et al.* (2004) measured the mean particle size and bed voidage at different positions of a modified column with a direct method (in-bed sampling) for Streamline SP particles. They also

suggested a model to determine the local bed voidage and mean particle size variations along the bed height. They reported an increasing trend of local bed voidage and decreasing mean particle diameter with the increase of bed height (Yun *et al.*, 2004). Yun *et al.* (2005) carried out experimental and modeling studies to estimate the variation of local axial dispersion coefficient with bed height in an EBA system for streamline SP and DEAE. They found that mixing and dispersion are intense near the bottom of column and become calmer near the top zones. Willoughby *et al.* (2000) also studied the variation of mean particle size and voidage along the bed height within a modified STREAMLINE column. Column design, like solid and liquid phase properties, plays an influential role in flow hydrodynamics and adsorption behavior within an EBA system. Therefore, such hydrodynamic studies should be performed in the columns with new design, although most of them were performed in commercial columns. Recently, a simple customized NBG expanded bed column was constructed by the Nanobiotechnology group (Babol University of Technology) (Shahavi *et al.*, 2008; Jahanshahi *et al.*, 2009; Jahanshahi and Ebrahimpour, 2009; Ebrahimpour *et al.*, 2010). Hydrodynamic parameter determination at various position of the customized NBG column is crucial. These key factors describe nano/bioproduction adsorption behavior, breakthrough curve trends, dynamic binding capacity, and bed utilization in each zone of the bed. Gaining the knowledge that flow behaves uniformly with the average/overall hydrodynamic parameters or non-uniformly with the local hydrodynamic parameters in a NBG column should ultimately lead to suitable adsorption model predictions, better simulations and model parametric sensitivity analyses. In this work, the hydrodynamic behavior, local bed voidage, Bodenstein number and local axial dispersion coefficient at different positions of the bed within an EBA system are studied. For this purpose, the customized NBG expanded bed column fabricated previously was modified. Bed voidage was measured at different positions of the column by a direct procedure (sampling method) and RTD analysis carried out. The effect of different operational conditions such as column diameter, settled bed height, flow velocity and flow viscosity on the local bed voidage, mixing and dispersion were investigated and results for the modified columns compared with other studies. Finally, experimental data obtained with the modified NBG column in this work are compared with a series of empirical or semi-empirical correlations.

EXPERIMENTAL

Materials

Streamline DEAE adsorbent was purchased from Amersham Bioscience (Uppsala, Sweden). The particle size distribution was in the range of 100-300 μm and the mean particle size was 200 μm ; the particle density was approximately 1200 kg m^{-3} . Acetone and glycerol were supplied by Merck Company.

Modified Column

Two modified NBG expanded bed columns were used in the experiments. The first column was 10 mm and the second 26 mm in inner diameter and 30 cm in height. An ordinary custom built NBG column was equipped with a simple sintered glass distributor consisting of a 100 μm mesh. Columns were divided

into 9 zones by eight 2- 2.5 mm sampling holes located every 3 cm along the bed height. A net adapter was placed at the top of the columns and lowered to a position 0.5 ± 0.2 cm above the bed surface to minimize the dead volume of the system during the experiments. Sampling holes were sealed with moveable rubber bands before the experiments were carried out. Figure 1 shows a schematic of the modified NBG column and EBA system set-up. Needles that were used in the experiments were of two types. One was a metal needle with 1.5 mm inner diameter connected to a 1 mL graduated syringe. Samples containing liquid and adsorbent particles were simultaneously withdrawn from each sampling hole by this type of needle. Another type was a metal needle with an inner diameter less than 0.1 mm that withdrew only liquid from each sampling position. Selection of the needle depended on the type of experiment.

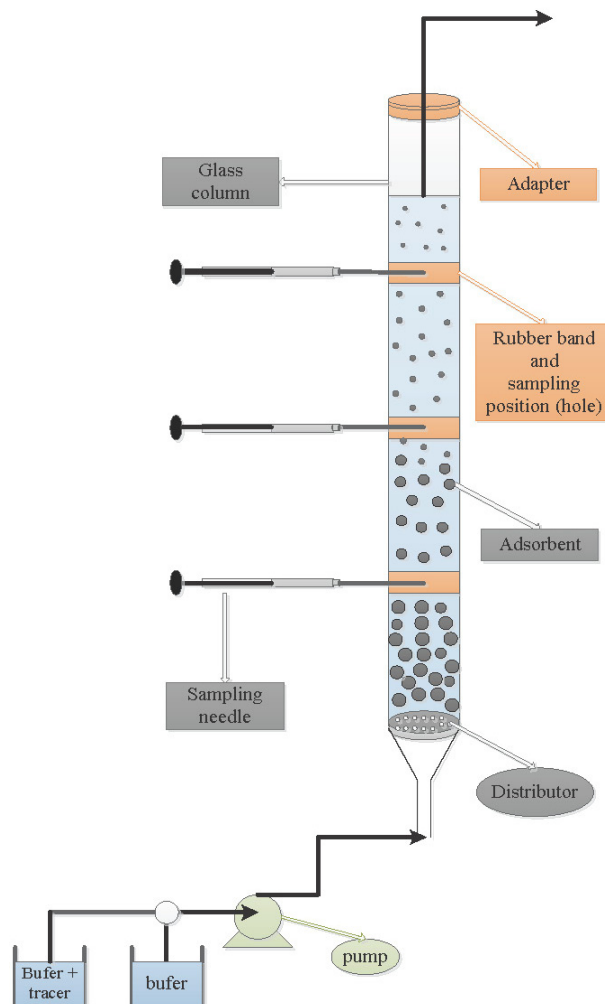


Figure 1: Schematic design of the modified NBG column.

Measurements of RTD

The column was filled with the adsorbent particles. The bed was expanded with buffer until stable expansion was established in the system (about 30 min). RTDs in various positions along the bed height were performed by using a negative step signal method (Amersham Biosciences, 1998; Jahanshahi *et al.*, 2002; Palsson *et al.*, 2001). Dilute acetone (1% v/v) was used as the input to the column in the blank systems expanded with buffer alone and tracer concentrations in the bed effluent were measured spectrophotometrically at 280 nm. This procedure was repeated in each of the holes one by one. Experiments were repeated in triplicate at each point to ensure consistency. The experimental conditions are summarized in Table 1. RTD measurements at each position were used to estimate the Bodenstein numbers and axial dispersion coefficients in expanded bed operation. On the other hand, RTDs were studied to determine liquid mixing and dispersion within the column (Amersham Biosciences, 1998, Jahanshahi *et al.*, 2002; Palsson *et al.*, 2001; Levenspiel, 1972). These parameters were obtained by using equations 1 and 2 (Levenspiel, 1972; Najafpour, 2007; Jahanshahi *et al.*, 2003; Biazus *et al.*, 2006).

$$B_o = 2N = 2 \frac{\overline{t^2}}{\delta^2} \quad (1)$$

$$D_{axl} = \frac{U_1 H}{2 \varepsilon N} \quad (2)$$

where $\overline{t^2}$ and δ^2 are the residence time and variance,

U_1 is the superficial liquid velocity, H is the bed height, ε is the bed voidage, B_o is the Bodenstein number, D_{axl} is the axial dispersion coefficient and N is the number of theoretical plates.

Sampling of Local Bed Voidage

The column was filled with the adsorbent and expanded to a stable height for 45-60 min with the buffer. The sample needle was inserted into the column center through each selective hole for 30 min and then adsorbent particles and liquid (1 mL) were simultaneously withdrawn (in < 3 s) with a needle connected to a syringe. After the, sample was withdrawn and the needle taken out, the hole was sealed with rubber. The syringe was placed vertically (for about 2.5-3 h) until the particles were perfectly settled. The volumes of particles that settled in the syringe and of the liquid above particles were obtained. The voidage was determined from Eq. (3) (Yun *et al.*, 2004).

$$\varepsilon_i = \frac{0.4 V_o + V_l}{V_o + V_l} \quad (3)$$

where ε_i is the local bed voidage in the i th layer of the bed, V_o is the volume of particles in the sampling syringe and V_l is the liquid volume above the particles.

After these steps, samples were returned to the column and the bed was re-expanded for measurement of the voidage in other sampling positions. The experiment was performed at each of the sample ports along the bed one by one and repeated three times at each position. The experimental conditions are illustrated in Table 2.

Table 1: Operating conditions in the RTD experiments

Column diameter (cm)	Adsorbent	Liquid	Settled bed height (cm)	Liquid velocity (m s ⁻¹)	H/H _{sb}
1	Streamline DEAE	Distilled water	6.5	4.6×10 ⁻⁴	1.69
				8.5×10 ⁻⁴	2
				13.8×10 ⁻⁴	2.77
				17.7×10 ⁻⁴	3.23
			10	12.7×10 ⁻⁴	2
		12	14.15×10 ⁻⁴	2	
1	Streamline DEAE	Distilled water + 10% (v/v) glycerol	6.5	4.6×10 ⁻⁴	2.08
		Distilled water + 20% (v/v) glycerol	6.5	4.6×10 ⁻⁴	2.31
2.6	Streamline DEAE	Distilled water	6	4.2×10 ⁻⁴	1.88
				6.3×10 ⁻⁴	2.17
				8.15×10 ⁻⁴	2.5

Table 2: Summary of experimental conditions for the bed voidage experiments

Column diameter (cm)	Adsorbent	Liquid	Settled bed height (cm)	Liquid velocity (m s ⁻¹)	H/H _{sb}
2.6	Streamline DEAE	Distilled water	6	4.2×10 ⁻⁴	1.88
				6.3×10 ⁻⁴	2.17
				8.15×10 ⁻⁴	2.5

THEORY

Adsorbent Particle Size Distribution

Tong and Sun (2002) presented empirical correlation for the variation of the mean particle size along the bed height in an EBA system for streamline particles with a broad size distribution and relatively uniform density. Their correlation is expressed by Eq. (4) (Tong and Sun, 2002):

$$\frac{d_{pi}}{d_p} = 1.21 - 0.46 \frac{h}{H} \quad (4)$$

where (h/H) is the ratio of sampling height to expanded bed height and (d_{pi}/d_p) is the ratio of measured mean diameter at different axial positions to the mean diameter of the streamline particles in the bulk. Kaczmariski and Bellot (2005) used this correlation (Eq. (4)) in their work to explain the mean particle diameter in each layer of bed. Also Tong *et al.* (2003) employed this empirical correlation for variation of mean particle size along the bed in order to model the protein adsorption by taking into account the axial particle size distribution in an EBA system. It was assumed that expanded bed conditions dominated in that system; therefore, equation 4 was employed to determine the mean particle diameter along the bed height.

Local Bed Voidage

Richardson and Zaki (1954) explained the correlation between superficial velocity and bed expansion. This correlation has been widely applied in liquid-solid fluidized beds and EBA systems in numerous empirical and semi-empirical studies of average bed expansion. Al-Dibouni and Garside (1979) and Yun *et al.* (2004) employed this correlation to measure local bed voidage along the bed height in their classified fluidized bed model. Their correlation is expressed as Eq. (5):

$$\frac{U_1}{U_{ti}} = \varepsilon_i^n \quad (5)$$

where U_{ti} is the particle terminal velocity of the *i*th layer and *n* is the bed expansion index (Richardson-Zaki correlation exponent) that can be determined by Eq. (6) (Anspach *et al.*, 1999; Fenneteau *et al.*, 2003; Jahanshahi *et al.*, 2009):

$$\frac{5.1 - n}{n - 2.4} = 0.016 Ga^{0.67} \quad (6)$$

where Ga is the Galileo number obtained from Eq. (7):

$$Ga = \frac{d_{pi}^3 g \rho (\rho_{pi} - \rho)}{\mu^2} \quad (7)$$

where d_{pi} is the particle mean diameter in the *i*th layer, ρ is the liquid density, μ is the liquid viscosity, ρ_{pi} is the particle mean density in the *i*th layer that is relatively uniform along the bed height for streamline particles. Particle terminal velocity can be estimated from Eq. (8):

$$U_{ti} = \frac{d_{pi}^2 g (\rho_{pi} - \rho)}{18\mu} \quad (8)$$

In this work, we used the Richardson-Zaki correlation to investigate local bed voidage at each different position of the bed. The Tong-Sun correlation, Eq. (4), was employed instead of the mean particle size at each position of the bed in these correlations (Eq. (7) and (8)). This method (coupling of two empirical correlations, Richardson-Zaki and Tong-Sun) is discussed for the first time and the results compared with experimental data in the modified NBG column.

Local Axial Dispersion Coefficient

Yun *et al.* (2005) presented an empirical equation for variation of the axial dispersion coefficient with bed height at a certain degree of expansion for streamline particles. They considered the contributions of: (a) local bed voidage variations; (b) effects of interstitial velocity, liquid properties and solid properties; (c) particle movement because of the

particle-particle interactions and the near-wall non-uniform velocity profile to obtain Eq. (9):

$$\frac{D_{\text{axi}} \rho}{\mu} = 13.2 (\text{Re}_{\text{pi}} \varepsilon_i)^{0.65} \beta_{\text{ri}}^{-0.5} \quad (9)$$

where D_{axi} is the axial dispersion coefficient at each position of the bed, ρ and μ are liquid properties, β_{ri} is the relative energy dissipation rate in the i th layer, defined as:

$$\beta_{\text{ri}} = \frac{32 U_1}{\frac{d_c^2 (\rho_{\text{pi}} - \rho)(1 - \varepsilon_i) \varepsilon_i g}{\mu} + 32 U_1} \quad (10)$$

where d_c is the column inner diameter and Re_{pi} is the particle Reynolds number in the i th zone, expressed as:

$$\text{Re}_{\text{pi}} = \frac{d_{\text{pi}} U_1 \rho}{\mu} \quad (11)$$

We used the Yun *et al.* equation (Eq. (9)) here in order to compare the experimental data and correlate the value of the axial dispersion coefficient. Instead of local bed voidage and mean particle size in each layer, the correlations of Richardson-Zaki and Tong-Sun were utilized. Chung and Wen (1968) and Kikuchi *et al.* (1984) presented empirical correlations to explain overall flow hydrodynamic behavior in the liquid-solid fluidized bed. Yun *et al.* (2005) used these correlations for an EBA system to study the flow behavior in each point of bed. We also employed these correlations to determine the variation of the axial dispersion coefficient with bed height in an EBA system and compared experimental data in the customized NBG column with the correlated data.

The Chung-Wen correlation is given by Eq. (12):

$$\frac{D_{\text{axi}} \rho}{\mu} \left(\frac{\text{Re}_{\text{mfi}}}{\text{Re}_{\text{pi}}} \right) = \frac{\text{Re}_{\text{pi}}}{0.2 + (0.11 \text{Re}_{\text{pi}}^{0.48})} \quad (12)$$

where Re_{mfi} is the onset Reynolds number in the i th zone, expressed by:

$$\text{Re}_{\text{mfi}} = \sqrt{33.7^2 + 0.408 \text{Ga}_i} - 33.7 \quad (13)$$

The correlation of Kikuchi *et al.* (1984) is defined by Eq. (14):

$$\frac{D_{\text{axi}} \rho}{\mu} = 500 \beta_{\text{oi}}^{0.43} \exp[-20.5(0.75 - \varepsilon_i)^2] \quad (14)$$

where β_{oi} is the energy dissipation rate per unit mass of liquid in the i th zone.

RESULTS AND DISCUSSIONS

Column with 26 mm Diameter

Local Bed Voidage

Table 3 shows the axial bed voidage at different column heights under various liquid flow velocities. Experimental results indicate that at a certain degree of expansion, the local bed voidage increases along the bed height. An increasing trend of voidage with the increase of flow velocity at a certain position can also be seen from Table 3. The trends in this simple column are similar to those reported in the literature (Yun *et al.*, 2004; Bruce and Chase, 2001; Willoughby *et al.*, 2000). The agreement between the local bed voidage values measured experimentally herein and those predicted by combining the Tong-Sun and Richardson-Zaki correlations is shown in Figures 2, 3 (a)-(c). In addition, the relative sum of squares of errors (RSSE) was calculated and the results shown in Table 4.

Table 3: Experimental voidage variation at different column heights in the bed of streamline DEAE at various liquid superficial velocities (6 cm settled bed height; 2.6 cm column diameter)

	$U_1 = 4.2 \times 10^{-4}$ (m s^{-1})	$U_2 = 6.3 \times 10^{-4}$ (m s^{-1})	$U_3 = 8.15 \times 10^{-4}$ (m s^{-1})
Different Sampling positions	voidage	voidage	voidage
$h_1 = 3$ cm	0.625	0.661	0.705
$h_2 = 6$ cm	0.641	0.673	0.725
$h_3 = 9$ cm	0.678	0.708	0.742
$h_4 = 12$ cm		0.746	0.781

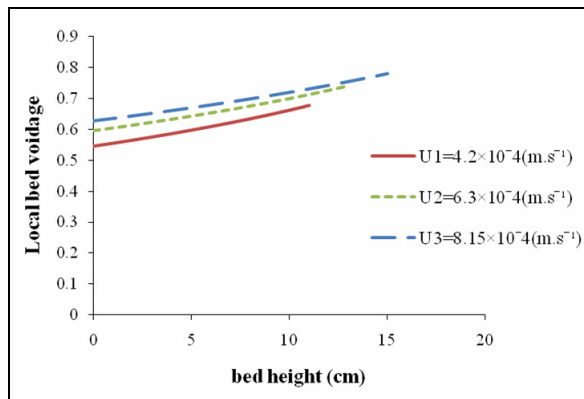


Figure 2: Predicted axial voidage by the Richardson-Zaki and Tong-Sun correlations at different bed heights under various liquid superficial velocities (streamline DEAE; 6 cm settled bed height; 2.6 cm column diameter).

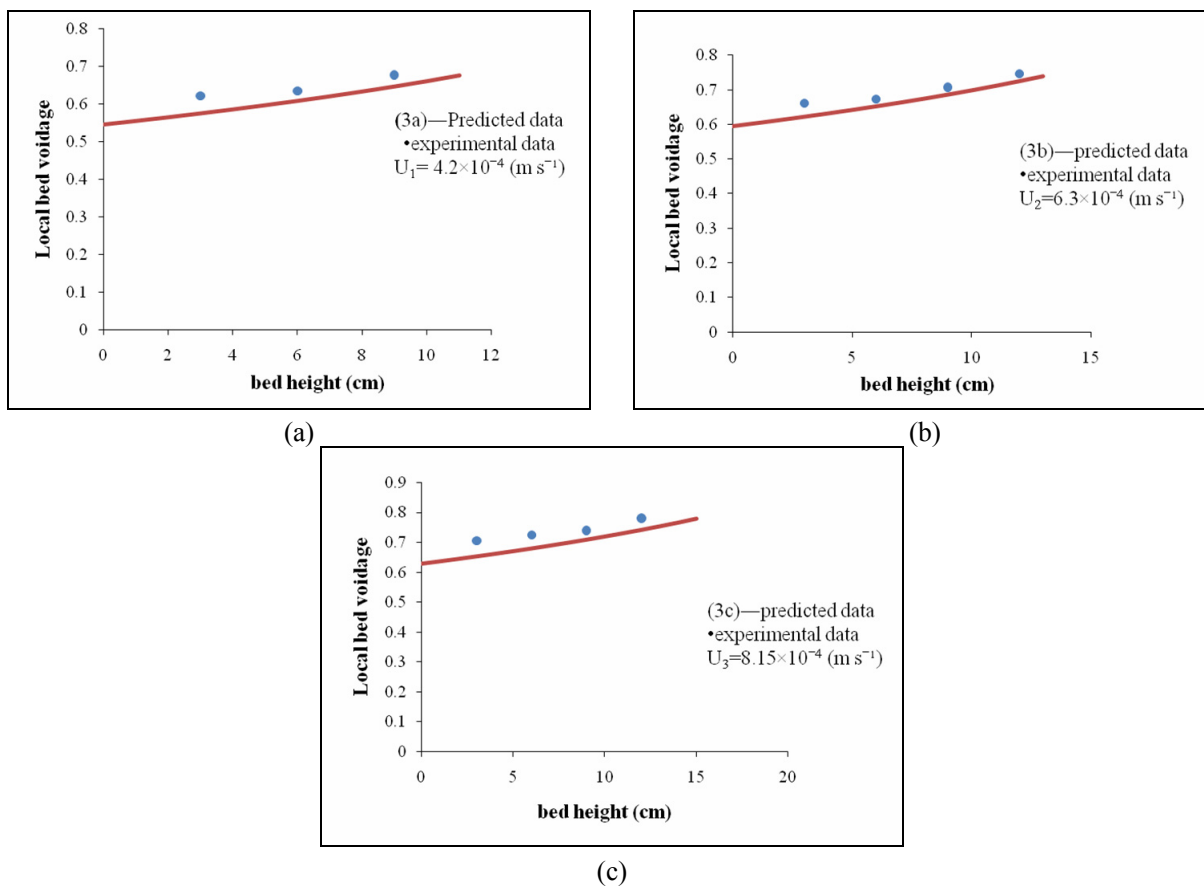


Figure 3 (a)-(c): Comparison of predicted axial voidage with experimental results at different bed heights under various liquid superficial velocities (streamline DEAE 6 cm settled bed height; 2.6 cm column diameter)

Table 4: RSSE for local bed voidage at different flow velocities

Flow velocity	$U_1 = 4.2 \times 10^{-4} \text{ (m s}^{-1}\text{)}$	$U_2 = 6.3 \times 10^{-4} \text{ (m s}^{-1}\text{)}$	$U_3 = 8.15 \times 10^{-4} \text{ (m s}^{-1}\text{)}$
RSSE (%)	99.7	99.9	99.7

The RSSE is described by Eq. (15) (Wright and Glasser, 2001):

$$RSSE = \frac{1}{m} \sum_{i=1}^n \frac{e_i^2 - (e_i - p_i)^2}{e_i^2} \quad (15)$$

where e_i and p_i are the experimental and predicted values and m is the number of data points, a value of 100% for the RSSE corresponds to perfect agreement.

As shown in Figures 2 and 3 and Table 4, the variation of the voidage with bed height is linear. Excellent agreement between predicted and experimental data was observed in most of regions of the bed. Of course there is some deviation between predicted and experimental data in the bottom zones due to the fact that, in this work, we assumed that ideal expanded bed conditions are dominant and particle mixing is low, while the bottom regions of the bed have severe mixing. Therefore, this effect should not be ignored in this region. Another assumption herein was that particles have uniform density. Particle density may only be approximately uniform, which probably lead to slight deviations of the mean particle size from the Tong-Sun correlation. In addition, another reason for these deviations might be the type of sampling procedure used for voidage determination. The present correlations for bed voidage are suitable (Table 4 and Figure 3); therefore, we employed them to determine the correlated local axial dispersion coefficient in the next section.

Local Axial Dispersion Coefficient

Experimental values for the Bodenstein number and axial dispersion coefficient at different positions of the bed are shown at Tables 5 and 6. As can be seen from the tables, for a stable condition, mixing and axial dispersion vary at different positions of the bed and decrease with increasing bed height. These results are similar to data reported in the literature (Bruce and Chase, 2001; Yun *et al.*, 2005). Numerous reasons exist for these non-uniform flow behaviors and some of them are discussed herein. A number of factors such as liquid phase properties (velocity, viscosity, etc), solid phase properties (size/density distribution, porosity, etc), bed voidage, and column design (diameter, distributor, settled bed height, etc) affect on flow behavior in an EBA system. According to our results (Table 3 and Figure 3) and other studies, the local bed voidage changes along the bed height. Since the bed voidage is low in the bottom region and particle concentrations are high, interactions

between particles are intense and unwanted falling and rising of particles occurs, which result in higher liquid back mixing and dispersion in this region. In contrast, voidage in the middle and top zones of the column increases and the interstitial velocity then decreases with the bed height and mixing and dispersion become weaker in these regions. Another important reason for such behavior is column design. The distributor located at the end of column disperses liquid with a non-uniform flow profile. With increasing distance from the column distributor, this effect become lower and uniform velocity profiles are obtained near the column top zones. Therefore, there is a decreasing trend of local mixing and dispersion with bed height.

Table 5: Axial mixing (Bodenstein number) at different column heights in the bed of streamline DEAE under different liquid superficial velocities (6 cm settled bed height; 2.6 cm column diameter)

	$U_1 = 4.2 \times 10^{-4}$ ($m\ s^{-1}$)	$U_2 = 6.3 \times 10^{-4}$ ($m\ s^{-1}$)	$U_3 = 8.15 \times 10^{-4}$ ($m\ s^{-1}$)
Different Sampling positions	B_o	B_o	B_o
$h_1 = 3\ cm$	4.43	3.78	3.71
$h_2 = 6\ cm$	7.2	5.93	4.42
$h_3 = 9\ cm$	9.5	7.31	6.24
$h_4 = 12\ cm$	11.41	9.22	6.7
$h_5 = 15\ cm$			10.87

Table 6: Variation of the local effective axial dispersion coefficient in different column zones at different liquid superficial velocities (streamline DEAE particles; 6 cm settled bed height; 2.6 cm column diameter)

	$U_1 = 4.2 \times 10^{-4}$ ($m\ s^{-1}$)	$U_2 = 6.3 \times 10^{-4}$ ($m\ s^{-1}$)	$U_3 = 8.15 \times 10^{-4}$ ($m\ s^{-1}$)
Different column zones	D_{axl} ($m^2\ s^{-1}$)	D_{axl} ($m^2\ s^{-1}$)	D_{axl} ($m^2\ s^{-1}$)
$h_1 = 0-3\ cm$	4.58×10^{-6}	7.575×10^{-6}	9.35×10^{-6}
$h_2 = 3-6\ cm$	3.44×10^{-6}	5.836×10^{-6}	8.41×10^{-6}
$h_3 = 6-9\ cm$	2.3×10^{-6}	4.13×10^{-6}	6.26×10^{-6}
$h_4 = 9-12\ cm$	1.77×10^{-6}	3.14×10^{-6}	5×10^{-6}
$h_5 = 12-15\ cm$			3.54×10^{-6}

Column diameter is another parameter that also affects flow behavior in an EBA system. Such non-uniform flow behavior obtained within the column with a 26 mm diameter was not observed in the column with a 10 mm diameter in this work. Mixing and dispersion were lower within the column with a 10 mm diameter than a 26 mm diameter. In addition, when the superficial velocity increases, the flow behaves worse and, at a certain position, the local

axial dispersion coefficient increases. However, at very high flow velocities, axial dispersion variation along the bed height may not be observed. This is due to the fact that, at very high flow velocity, convection is severe and particle movements that lead to higher mixing and dispersion increase in all of the bed zones.

In this work, we considered three empirical equations to determine variations of the local axial dispersion coefficient along the bed height (Eqs. (9), (12) and (14)). Then experimental data obtained in the modified NBG column were compared with these correlations (Figures 4, 5 and Table 7). Both the correlations of Chung-Wen and Kikuchi *et al.* show (Figure 4 (b)-(c)) an increasing trend of the local axial dispersion coefficient along the bed height, while our experimental studies show the inverse trend. In addition, there is not agreement between our experimental and correlated data. Therefore, these correlations are not valid for the local axial dispersion coefficient at each position of the bed in the EBA system used in this work. On the other hand, they are suitable for estimation of the

overall dispersion in these systems. Figures 4 (a), 5 and Table 7 make a comparison of the local axial dispersion coefficients measured experimentally and calculated by the Yun equation. The decreasing trend of the axial dispersion coefficient with the increase in bed height at a certain velocity and the decrease of velocity at a certain bed position, similar to our experimental results, can be seen in Figure 4 (a). However, Figure 5 and Table 7, which compare the local axial dispersion coefficients in each zone of bed show a discrepancy between experiment and the correlated values in the zones near of distributor. This irregularity was expected in the bottom bed zones, as described before. With increasing distance from the distributor, this agreement becomes much better and the correlation gives excellent predictions. In addition, an increase of velocity leads to a RSSE decrease. Therefore, this correlation (Eq. (9)) predicts our experimental results for the local axial dispersion coefficient at different bed points better than other correlations (Eqs. (12) and (14)) and can describe local hydrodynamic flow behavior well within the EBA system.

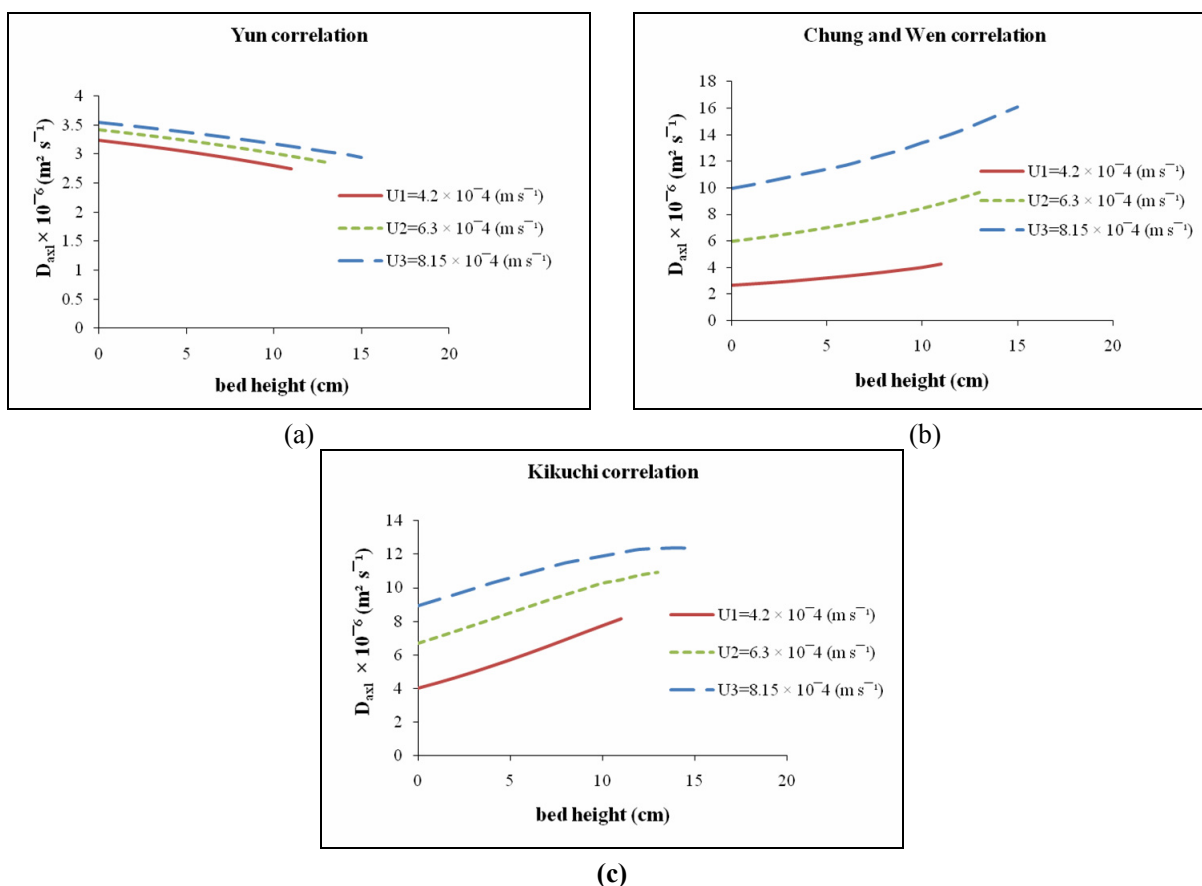


Figure 4 (a)-(c): Predicted axial dispersion coefficients at different bed heights by empirical correlations under various liquid superficial velocities (streamline DEAE; 6 cm settled bed height; 2.6 cm column diameter).

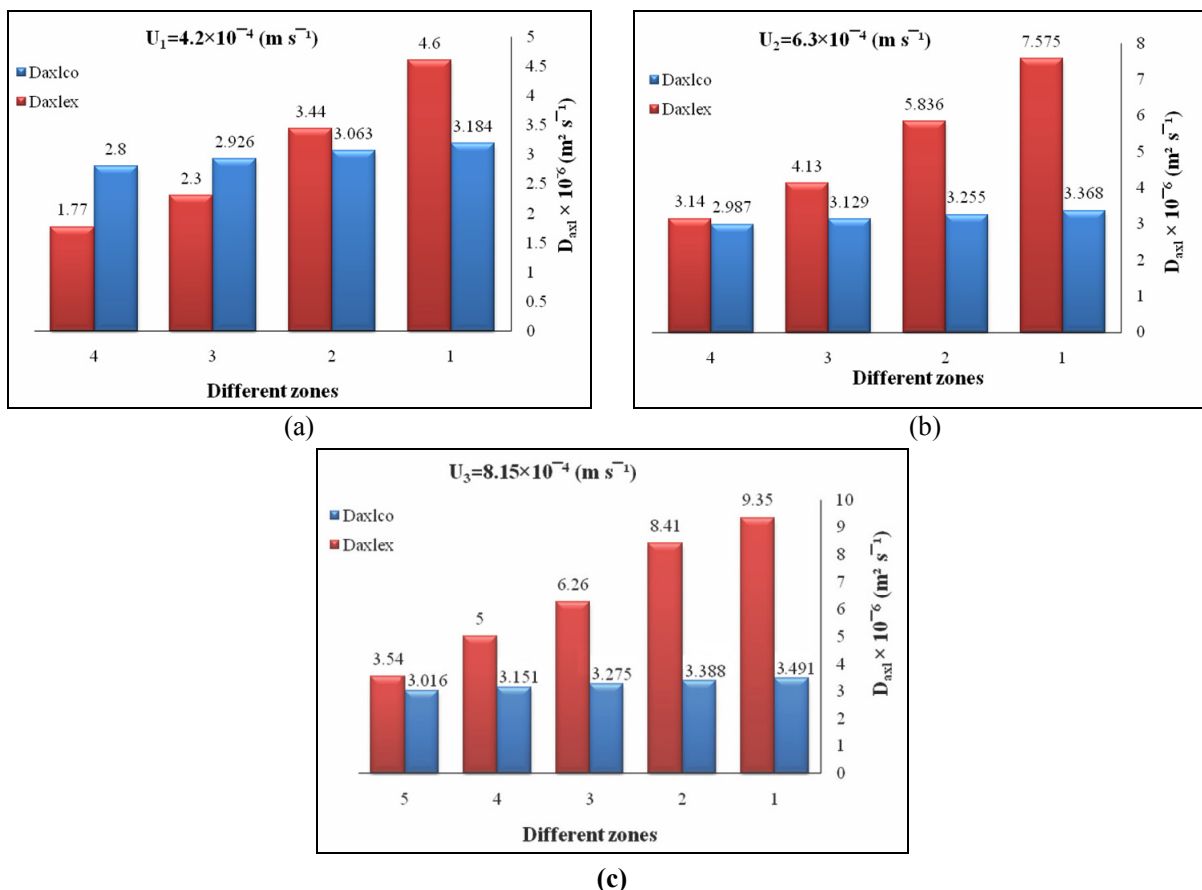


Figure 5 (a)-(c): Comparison of predicted axial dispersion coefficients obtained by the Yun equation with experimental results in different bed zones at various liquid superficial velocities (1: zone between 0-3 cm, 2: zone between 3-6 cm, 3: zone between 6-9 cm, 4: zone between 9-12 cm, 5: zone between 12-15 cm)

Table 7: RSSE for the local axial dispersion coefficient at different flow velocities

Flow velocity	$U_1 = 4.2 \times 10^{-4} \text{ (m s}^{-1}\text{)}$	$U_2 = 6.3 \times 10^{-4} \text{ (m s}^{-1}\text{)}$	$U_3 = 8.15 \times 10^{-4} \text{ (m s}^{-1}\text{)}$
RSSE (%)	87	85.85	77.2

Column with 10 mm Diameter

In order to study the influence of column diameter on fluid local hydrodynamic behavior, another modified NBG column with a smaller diameter (10 mm) was prepared in this research. Therefore, the effect of different parameters such as flow velocity, liquid viscosity, and settled bed height on local hydrodynamic performance within the column with 10 mm in diameter were also examined and discussed.

Influence of Flow Velocity on Fluid Hydrodynamics

Tables 8 and 9 illustrate Bodenstein numbers and local effective axial dispersion coefficients at

different bed positions under various superficial liquid velocities. As can be expected, axial mixing and dispersion increase with increasing superficial liquid velocity at a certain height of the bed. In the bottom of the bed, these hydrodynamic parameters are high and become relatively low near the middle and top regions of the bed. Obvious variations of liquid behavior along the bed height were not observed in the middle and top regions. This is probably due to the fact that the column diameter is small, which result in a uniform flow profile in most of the bed regions (except the distributor region). The other reason is that the variation of the particle concentration along the bed is negligible within this column, resulting in uniform behavior in the middle and top regions of the bed.

Table 8: Axial mixing (Bodenstein number) at different column heights in the bed of streamline DEAE at different liquid superficial velocities (settled bed height = 6.5cm; 1 cm column diameter)

	$U_1 = 4.6 \times 10^{-4}$ (m s ⁻¹)	$U_2 = 8.5 \times 10^{-4}$ (m s ⁻¹)	$U_3 = 13.8 \times 10^{-4}$ (m s ⁻¹)	$U_4 = 17.7 \times 10^{-4}$ (m s ⁻¹)
Different sampling positions	B_o	B_o	B_o	B_o
$h_1 = 3$ cm	4.5	6	5.57	5.3
$h_2 = 6$ cm	12.2	17.28	9.88	8
$h_3 = 9$ cm	11.92	17.1	9.95	8
$h_4 = 12$ cm	12.22	16.79	10.7	7.61
$h_5 = 15$ cm			9.8	7.91
$h_6 = 18$ cm			9.68	7.2
$h_7 = 21$ cm				8

Table 9: Variation of the local effective axial dispersion coefficient in different column zones at different liquid superficial velocities (streamline DEAE particles; 6.5 cm settled bed height; 1cm column diameter)

	$U_1 = 4.6 \times 10^{-4}$ (m s ⁻¹)	$U_2 = 8.5 \times 10^{-4}$ (m s ⁻¹)	$U_3 = 13.8 \times 10^{-4}$ (m s ⁻¹)	$U_4 = 17.7 \times 10^{-4}$ (m s ⁻¹)
Different column zones	D_{axl} (m ² s ⁻¹)	D_{axl} (m ² s ⁻¹)	D_{axl} (m ² s ⁻¹)	D_{axl} (m ² s ⁻¹)
$h_1 = 0-3$ cm	4.72×10^{-6}	5.27×10^{-6}	9.5×10^{-6}	12.31×10^{-6}
$h_2 = 3-6$ cm	2.54×10^{-6}	3.4×10^{-6}	6.87×10^{-6}	9.81×10^{-6}
$h_3 = 6-9$ cm	1.77×10^{-6}	2.11×10^{-6}	5.356×10^{-6}	8.15×10^{-6}
$h_4 = 9-12$ cm	1.77×10^{-6}	2.11×10^{-6}	5.14×10^{-6}	8.35×10^{-6}
$h_5 = 12-15$ cm			5.18×10^{-6}	8.4×10^{-6}
$h_6 = 15-18$ cm			5.45×10^{-6}	8.46×10^{-6}
$h_7 = 18-21$ cm				8.42×10^{-6}

Because of the uniform behavior, the overall bed voidage correlation, Eq. (16) (Li and Chase, 2009; Levenspiel, 1999), was used to calculate the axial dispersion coefficient in this column.

$$\varepsilon = 1 - (1 - \varepsilon_0) \left(\frac{H_{sb}}{H} \right) \quad (16)$$

where H_{sb} is the settled bed height, H is the bed height, ε_0 is the voidage in the fixed bed condition (settled bed voidage), assumed to be 0.4.

Influence of Liquid Viscosity on Fluid Hydrodynamics

The results of studies of the effect of flow viscosity on liquid behavior are displayed in Tables 10

Table 10: Axial mixing (Bodenstein number) at different column heights in the bed of streamline DEAE particles at different liquid viscosities (liquid flow velocity is 4.6×10^{-4} m s⁻¹; 6.5 cm settled bed height; 1 cm column diameter)

	Distilled Water	Distilled Water +10% (v/v) glycerol	Distilled Water
Different sampling positions	B_o	B_o	B_o
$h_1 = 3$ cm	4.5	8.2	5.175
$h_2 = 6$ cm	12.2	12.36	9.34
$h_3 = 9$ cm	11.92	12.6	8.93
$h_4 = 12$ cm	12.22	12.5	9.45
$h_5 = 15$ cm			8.64

and 11. These tables show that mixing and dispersion decrease with increasing viscosity, altered by addition of 10% (v/v) glycerol to the buffer. This is due to the fact that the degree of bed expansion increases with the increase of liquid viscosity under constant velocity. Therefore, at a certain position, decreasing solid particle interaction and increasing bed voidage are expected. However, with an increase of liquid density or viscosity, convection become calmer, resulting in better liquid behavior. In contrast, an increase of viscosity by addition of 20% (v/v) glycerol to the buffer led to worse liquid behavior. Reduction of the discrepancy between the liquid and solid particle density might result in intense mixing and dispersion. In addition, except in the distributor region, the flow hydrodynamics is approximately uniform in this column at various viscosities, as expected.

Table 11: Variation of the local effective axial dispersion coefficient in different column zones at different liquid viscosities (streamline DEAE particles; liquid flow velocity is 4.6×10^{-4} (m s⁻¹); 6.5cm settled bed height; 1 cm column diameter)

	Distilled Water	Distilled Water +10% (v/v) glycerol	Distilled Water +20% (v/v) glycerol
Different column zones	D_{axl} (m ² s ⁻¹)	D_{axl} (m ² s ⁻¹)	D_{axl} (m ² s ⁻¹)
$h_1 = 0-3$ cm	4.72×10^{-6}	3.1×10^{-6}	3.68×10^{-6}
$h_2 = 3-6$ cm	2.54×10^{-6}	2.11×10^{-6}	2.63×10^{-6}
$h_3 = 6-9$ cm	1.77×10^{-6}	1.59×10^{-6}	2.085×10^{-6}
$h_4 = 9-12$ cm	1.77×10^{-6}	1.58×10^{-6}	2.073×10^{-6}
$h_5 = 12-15$ cm			2.13×10^{-6}

Influence of Settled Bed Height on Fluid Hydrodynamics

The results are shown in Tables 12 and 13. As can be seen, except in the bottom zones, the flow behaves approximately uniformly in the middle and top region. Therefore, column diameter is one of the important influential parameters on flow hydrodynamic performance. Our results show that flow behaves nearly uniformly within the smaller diameter column (10 mm) and, with the increase of diameter, this behavior becomes non-uniform and inhomogeneous. Thus, the liquid behavior changes along the bed in column with the larger diameter (26 mm).

Table 12: Axial mixing (Bodenstein number) at different column heights in the bed of streamline DEAE particles at different settled bed heights (streamline DEAE particles; 1 cm column diameter; 0.7 bed voidage)

	$H_{sb} = 10$ cm	$H_{sb} = 12$ cm
Different sampling positions	B_0	B_0
$h_1 = 6$ cm	12.3	14.6
$h_2 = 12$ cm	17.9	18.3
$h_3 = 18$ cm	18.2	19.4
$h_4 = 21$ cm	19.1	18.8
$h_5 = 24$ cm		20.2

Table 13: Variation of the local effective axial dispersion coefficient in different column zones at different settled bed heights (streamline DEAE particles; 1 cm column diameter; 0.7 bed voidage)

	$H_{sb} = 10$ cm	$H_{sb} = 12$ cm
Different column zones	D_{axl} (m ² s ⁻¹)	D_{axl} (m ² s ⁻¹)
$h_1 = 0-6$ cm	8.87×10^{-6}	8.31×10^{-6}
$h_2 = 6-12$ cm	7.23×10^{-6}	7.37×10^{-6}
$h_3 = 12-18$ cm	6.045×10^{-6}	6.43×10^{-6}
$h_4 = 18-21$ cm	5.9×10^{-6}	6.35×10^{-6}
$h_5 = 21-24$ cm		6.21×10^{-6}

Another result here is the fluid hydrodynamic performance enhancement in the column with 10 mm diameter. Mixing and dispersion in the column with 10 mm diameter is lower than in the column with 26 mm diameter.

Of course, these studies have been done on clean feedstock. Therefore, for real nano/biological feedstock, more studies and research should be performed.

CONCLUSION

In order to study fluid local hydrodynamic behavior within an EBA system, a modified simple customized NBG expanded bed column has been fabricated. A direct sampling method was carried out for estimation of the local bed voidage along the bed height and RTD analysis was performed in the modified NBG column to determine the local mixing and axial dispersion at different position of the bed. In addition, the effect of various factors including column diameter, flow velocity, flow viscosity and settled bed height on flow hydrodynamic performance was examined. Results indicated that nearly uniform flow behavior and lower mixing and dispersion occurred in the column with small diameter (10 mm), while the flow behaved non-uniformly within the larger diameter column (26 mm). However, it was shown that mixing and dispersion were intense at the end of bed and with an increase of bed height, these parameters became quite calm. One of the key influential factors on liquid behavior was the superficial velocity. An increasing trend of mixing and dispersion were observed with an increase of velocity at a certain point of the bed. Liquid viscosity was another parameter that affects flow hydrodynamics; increasing liquid viscosity led to decreased mixing and dispersion, but when this increase exceeded a certain value, the inverse trend was observed. The empirical correlations of Richardson-Zaki and Tong-Sun were combined

together to determine the local bed voidage. Calculated data gave quite good agreement with experimental data (RSSE = 99.9%) under the operating conditions of the experiment; hence, the direct procedure is good enough to estimate bed voidage at each point of the bed in the column. Three empirical correlations were extracted from the literature to identify the local axial dispersion coefficient variation. Among them, the Yun equation explained this parameter better than the other correlations and reasonable agreement existed between experimental and correlated values (RSSE = 87%). The local axial dispersion coefficients obtained under our operation condition were between 10^{-6} and 10^{-5} ($\text{m}^2 \text{s}^{-1}$) (most of them are about $10^{-6} \text{m}^2 \text{s}^{-1}$). These results indicated that stable expansion conditions occurred in our modified NBG column and the flow behavior was of the plug nature (Levenspiel, 1972). Therefore, fluid hydrodynamic performance in this column is good enough for recovery and purification of nano/bioproducts. This flow hydrodynamic study was performed for liquid without crude feedstock. Therefore, determination of real feedstock hydrodynamics within an EBA system, development of empirical correlations and mathematical hydrodynamic models for axial dispersion coefficient variation along the bed height with and without nano/biological feedstock, and presentation of mathematical model for nano/bioproducts adsorption performance and purification of nano/bioproducts in a NBG chromatography column will be subjects of our further publications.

ACKNOWLEDGEMENTS

The authors would like to gratefully acknowledge members of the Nanotechnology Research Institute of Babol University of Technology, Babol, Iran.

NOMENCLATURE

B_o	Bodenstein number	
D_{axl}	Axial dispersion coefficient	$\text{m}^2 \text{s}^{-1}$
D_{axli}	Axial dispersion coefficient at ith layer of the bed	$\text{m}^2 \text{s}^{-1}$
d_{pi}	Mean particle diameter at ith layer of the bed	m
d_p	Mean particle diameter	m
d_c	Column inner diameter	m
e_i	Experimental data	
Ga	Galileo number	
H	Total bed height	m

h	Axial bed height	m
H_{sb}	Settled bed height	m
m	Number of data points	
N	Number of theoretical plates	
n	Richardson- Zaki correlation exponent	
p_i	Simulated data	
Re_{pi}	Particle Reynolds number in the ith layer	
Re_{mfi}	Onset Reynolds number in the ith layer	
RSSE	Relative sum of squares of errors	
\bar{t}	Average residence time	s
U_1	Superficial liquid velocity	m s^{-1}
U_{ti}	Terminal velocity at the ith layer of the bed	m s^{-1}
V_o	Volume of settled particles in the sampling syringe	m^3
V_1	Liquid volume above the particles in the syringe	m^3

Greek Letters

β_{ri}	Relative energy dissipation rate in the ith layer	$\text{m}^2 \text{s}^{-3}$
β_{oi}	Energy dissipation rate per unit mass of liquid in the ith layer	$\text{m}^2 \text{s}^{-3}$
ε	Total bed voidage	
ε_i	Local bed voidage in the ith layer of the bed	
ε_o	Settled bed voidage	
μ	Liquid viscosity	Pa s
ρ	Liquid density	kg m^{-3}
ρ_{pi}	Particles mean density in the ith layer	kg m^{-3}
δ^2	Variance	s^2

REFERENCES

- Al-Dibouni, M. R. and Garside, J., Particle mixing and classification in liquid fluidized beds. *Trans. Inst. Chem. Eng.*, 57, 94-103 (1979).
- Amersham Biosciences: Expanded Bed Adsorption – Principles and Methods, Pharma Biotech, Uppsala, Sweden (1998).
- Anspach, F. B., Curbelo, D., Hartman, R., Garke, G. and Deckwer, W. D., Expanded-bed chromatography in primary protein purification. *J. Chromatogr. A*, 865, 129-144 (1999).
- Biazus, J. P. M., Severo Jr. J. B., Santana, J. C. C., Souza, R. R. and Tambourgi, E. B., Study of

- amylases recovery from maize malt by ion-exchange expanded bed chromatography. *Process. Biochem.*, 41, 1786-1791 (2006).
- Bruce, L. J. and Chase, H. A., Hydrodynamics and adsorption behavior within an expanded bed adsorption column studied using in-bed sampling. *Chem. Eng. Sci.*, 56, 3149-3162 (2001).
- Chung, S. F. and Wen, C. Y., Longitudinal dispersion of liquid flowing through fixed and fluidized beds. *AIChE. J.*, 14, 857-866 (1968).
- da Silva Padilha, G., Curvelo-Santana, J. C., Alegre, R. M. and Tambourgi, E. B., Expanded bed adsorption of an alkaline lipase from *Pseudomonas cepacia*. *J. Chromatogr. B*, 877, 521-526 (2009).
- Ebrahimpour, M., Jahanshahi, M. and Hosenian, A. H., Adsorption strategy of plasmid DNA nanoparticulate: Preparative purification by a simple custom expanded bed column. *Chromatographia*, 72, 383-391 (2010).
- Fenneteau, F., Aomari, H., Chahal, P. and Legros, R., Modeling of scale-down effects on the hydrodynamics of expanded bed adsorption columns. *Biotechnol. Bioeng.*, 81, 790-799 (2003).
- Jahanshahi, M. and Ebrahimpour, M., Expanded bed chromatography as a tool for nanoparticulate separation: Kinetic study and adsorption of protein nanoparticles. *Chromatographia*, 70, 1553-1560 (2009).
- Jahanshahi, M., Najafpour, G., Ebrahimpour, M., Hajizadeh, S. and Shahavi, M. H., Evaluation of hydrodynamic parameters of fluidized bed adsorption on purification of nano bioproducts. *Phys. Status. Solidi. C*, 6, (10), 2199-2206 (2009).
- Jahanshahi, M., Patek, A. W., Nienow, A. W. and Lyddiatt, A., Fabrication by three-phase emulsification of pellicular adsorbents customised for liquid fluidised bed adsorption of bioproducts. *J. Chem. Technol. Biotechnol.*, 78, 1111-1120 (2003).
- Jahanshahi, M., Sun, Y., Santos, E., Patek, A., Franco, T. T., Nienow, A. and Lyddiatt, A., Operational intensification by direct product sequestration from cell disruptates: Application of a pellicular adsorbent in a mechanically integrated disruption-fluidized bed adsorption process. *Biotechnol. Bioeng.*, 80, 201-212 (2002).
- Kaczmarzski, K. and Bellot, J. C., Influence of particle diameter distribution on protein recovery in the expanded bed adsorption process. *J. Chromatogr. A*, 1069, 91-97 (2005).
- Kikuchi, K. I., Konno, H., Kakutani, S., Sugawara, T. and Ohashi, H., Axial dispersion of liquid in liquid fluidized beds in the low Reynolds number region. *J. Chem. Eng. Jpn*, 17, 362-367 (1984).
- Levenspiel, O., *Chemical Reaction Engineering*. 3rd Ed. John Wiley & Sons, New York (1999).
- Levenspiel, O., *Chemical Reaction Engineering*. John Wiley & Sons, New York (1972).
- Li, J. and Chase, H. A., Characterization and evaluation of a macroporous adsorbent for possible use in the expanded bed adsorption of flavonoids from *Ginkgo biloba* L. *J. Chromatogr. A.*, 1216, 8730-8740 (2009).
- Li, P., Xiu, G. and Rodrigues, A. E., A3-zone model for protein adsorption kinetics in expanded beds. *Chem. Eng. Sci.*, 59, 3837-3847 (2004).
- Moraes, C. C., Mazutti, M. A., Rodrigues, M. I., Filho, F. M. and Kalil, S. J., Mathematical modeling and simulation of inulinase adsorption in expanded bed column. *J. Chromatogr. A*, 1216, 4395-4401 (2009).
- Najafpour, G. D., *Biochemical Engineering and Biotechnology*. Chapter 17, Elsevier, Amsterdam (2007).
- Ng, M. Y. T., Tan, W. S., Abdullah, N., Ling, T. C. and Tey, B. T., Direct purification of recombinant hepatitis B core antigen from two different pre-conditioned unclarified *Escherichia coli* feedstocks via expanded bed adsorption chromatography. *J. Chromatogr. A*, 1172, 47-56 (2007).
- Palsson, E., Axelsson, A. and Larsson, P. O., Theories of chromatographic efficiency applied to expanded beds. *J. Chromatogr. A*, 912, 235-248 (2001).
- Pinotti, L. M., Fonseca, L. P., Prazeres, D. M. F., Rodrigues, D. S., Nucci, E. R. and Giordano, R. L. C., Recovery and partial purification of penicillin G acylase from *E. coli* homogenate and *B. megaterium* culture medium using an expanded bed adsorption column. *Biochem. Eng. J.*, 44, 111-118 (2009).
- Richardson, J. F. and Zaki, W. N., Sedimentation and fluidization: Part I. *Trans. Inst. Chem. Eng.*, 32, 35-53 (1954).
- Shahavi, M. H., Najafpour, G. D. and Jahanshahi, M., Hydrodynamic behaviour and biochemical characterization of a simple custom expanded bed column for protein purification. *Afr. J. Biotechnol.*, 7, (23), 4336-4344 (2008).
- Tong, X. D. and Sun, Y., Particle size and density distributions of two dense matrices in an expanded bed system. *J. Chromatogr. A*, 977, 173-183 (2002).
- Tong, X. D., Dong, X. Y. and Sun, Y., Lysozyme adsorption and purification by expanded bed chromatography with a small-sized dense adsorbent. *Biochem. Eng. J.*, 12, 117-124 (2002).
- Tong, X. D., Xue, B. and Sun, Y., Modeling of expanded-bed protein adsorption by taking into

- account the axial particle size distribution. *Biochem. Eng. J.*, 16, 265-272 (2003).
- Vennapusa, R. R., Tari, C., Cabrera, R. and Fernandez-Lahore, M., Surface energetics to assess biomass attachment onto hydrophobic interaction adsorbents in expanded beds. *Biochem. Eng. J.*, 43, 16-26 (2009).
- Willoughby, N. A., Hjorth, R. and Titchener-Hooker, N. J., Experimental measurement of particle size distribution and voidage in an expanded bed adsorption system. *Biotechnol. Bioeng.*, 69, 648-653 (2000).
- Wright, P. R. and Glasser, B. J., Modeling mass transfer and hydrodynamics in fluidized-bed adsorption of proteins. *AIChE J.*, 47, 474-488 (2001).
- Yun, J., Yao, S. J. and Lin, D. Q., Variation of the local effective axial dispersion coefficient with bed height in expanded beds. *Chem. Eng. J.*, 109, 123-131 (2005).
- Yun, J., Yao, S. J., Lin, D. Q., Lu, M. H. and Zhao, W. T., Modeling axial distributions of adsorbent particle size and local voidage in expanded bed. *Chem. Eng. Sci.*, 59, 449-457 (2004).