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Characteristics of Micromixing in a Rotating Packed Bed

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Abstract—A parallel-competing chemical test reaction was performed in a rotating packed bed (RPB) with different radii to evaluate the micromixing efficiency. Experimental results indicated that increasing the rotational speed and the inner radius of the packed bed could intensify micromixing effectively. Increasing the outer radius of the bed had no significant effect on micromixing. In addition, experiments on the synthesis of magnesium hydroxide particles in an RPB and in a spinning disk reactor (SDR) were performed. Although higher micromixing efficiency was achieved and smaller particles were produced at a low liquid flow rate using an SDR, an RPB was a more effective device when the flow rate increased. Comparing with the reported segregation index of other mixing devices, a Higee system is a promising alternative as far as micromixing efficiency is concerned.

Key Words : Higee, Rotating packed bed, Micromixing, Segregation index

INTRODUCTION

When the time scale of a chemical reaction is smaller than or of the same magnitude as the time scale of a mixing process, micromixing (*i.e.*, mixing at molecular scale) may have an influence on the quality of products produced through combustion and precipitation. As a result, experimental methods for characterizing micromixing efficiency are needed.

To quantitatively estimate micromixing efficiency, two types of chemical test reactions are usually used, *i.e.*, consecutive-competing reactions (Bourne *et al.*, 1985) and parallel-competing reactions (Bourne and Yu, 1994; Fournier *et al.*, 1996). These reactions involve an instantaneous reaction coupled with a slower reaction, and the final product distribution depends on the micromixing efficiency. Fournier *et al.* (1996) proposed a parallel-competing reaction strategy which includes acid-base neutralization and the Dushman reaction:

$$H_2BO_3^- + H^+ \rightleftharpoons H_3BO_3 \text{ (quasi-instantaneous)}, \tag{1}$$

$$5I^- + IO_3^- + 6H^+ \rightleftharpoons 3I_2 + 3H_2O \text{ (fast)}. \tag{2}$$

Since the first reaction can be taken as being instantaneous, all hydrogen ions can be assumed to react with excess $H_2BO_3^-$ if mixing is perfect. Otherwise, iodine will be generated and will further react with I^- to form I_3^-, as shown in reaction (3):

$$I_2 + I^- \rightleftharpoons I_3^- . \tag{3}$$

The equilibrium constant of reaction (3) is calculated as follows (Guichardon and Falk, 2000):

$$K_B = \frac{555}{T} + 7.355 - 2.575 \log T . \tag{4}$$

The yield of reaction (2), Y, can be defined as the ratio of the moles of acid consumed by the second reaction to the total moles of acid introduced into the system:

$$Y = \frac{2(n_{I_2} + n_{I_3^-})}{n_{H^+,0}} = \frac{2(C_{I_2} + C_{I_3^-})Q}{C_{H^+,0} Q_H} . \tag{5}$$

In Eq. (5), Q ($=Q_I+Q_H$) represents the total liquid flow rates of the two solutions introduced into the system, and the subscript 0 denotes the inlet conditions. In the case of total segregation, the yield of reaction (2) is controlled by the concentration of $H_2BO_3^-$ and IO_3^-, because both reactions (1) and (2) are quasi-instantaneous, compared with the mixing process. As a result, the quantity of iodine formed is only determined by the stoichiometric ratio of the reactions, and the yield of reaction (2) under total segregation (Y_{ST}) can be expressed as

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$$Y_{ST} = \frac{6C_{\mathrm{IO_3^-},0}}{6C_{\mathrm{IO_3^-},0} + C_{\mathrm{H_2BO_3^-},0}} . \tag{6}$$

According to Eqs. (5) and (6), the segregation index (*Xs*) can be written as follows:

$$Xs = \frac{Y}{Y_{ST}} = \frac{(C_{\mathrm{I_2}} + C_{\mathrm{I_3^-}})\,Q}{C_{\mathrm{H^+},0}\,Q_{\mathrm{H}}}\left(\frac{6C_{\mathrm{IO_3^-},0} + C_{\mathrm{H_2BO_3^-},0}}{3C_{\mathrm{IO_3^-},0}}\right). \tag{7}$$

By definition, $Xs = 0$ denotes perfectly micromixing conditions, and $Xs = 1$ represents complete segregation. A detailed discussion and calculation of this segregation index can be found in work of Fournier *et al.* (1996).

A rotating packed bed (*i.e.*, a Higee system) can play an important role in process intensification by significantly enhancing the mass transfer process. High centrifugal fields are generated by rotation; therefore, the system can be operated at a higher gas/liquid ratio due to the decreased flooding tendency. Moreover, thinner liquid films and smaller droplets can be obtained in the bed because of the high shear force. The enhancement of mass transfer in gas/liquid, liquid/solid and liquid/liquid extraction systems has been demonstrated (Ramshaw and Mallinson, 1981; Ramshaw, 1993; Lin and Liu, 2000, 2003; Chen and Liu, 2002; Lin *et al.*, 2002, 2004; Chen *et al.*, 2005). In addtion, for a liquid/liquid mixing system, our previous work showed that the micromixing efficiency can be improved in a Higee system in the aid of centrifugal force (Chen *et al.*, 2004). In addition, a spinning disk reactor (SDR) was recently proved to be useful in the pharmaceutical and fine chemical industries due to its characteristics of high heat and mass transfer and high mixing efficiency (Oxley *et al.*, 2000; Brechtelsbauer *et al.*, 2001). In this study, the effect of the radius of the rotor on micromixing in an RPB was investigated. The micromixing efficiency of an SDR was also measured in this work, so that a comparison between an RPB and an SDR could be made.

Precipitation processes are of great importance in the chemical industry. They have been found to be greatly affected by micromixing, because the reaction time of precipitation is on the same order of magnitude as the characteristic time for the micromixing process. Marcant and David (1991) demonstrated a simplified mixing model for qualitatively predicting the influence of micromixing on precipitation. They found that increasing the mixing intensity could increase the crystallization rate and reduce the particle diameter in primary nucleation. In this study, magnesium hydroxide was precipitated in a liquid-liquid reaction system in both an RPB and an SDR. The mean particle size of the magnesium hydroxide prepared in these two devices was compared.

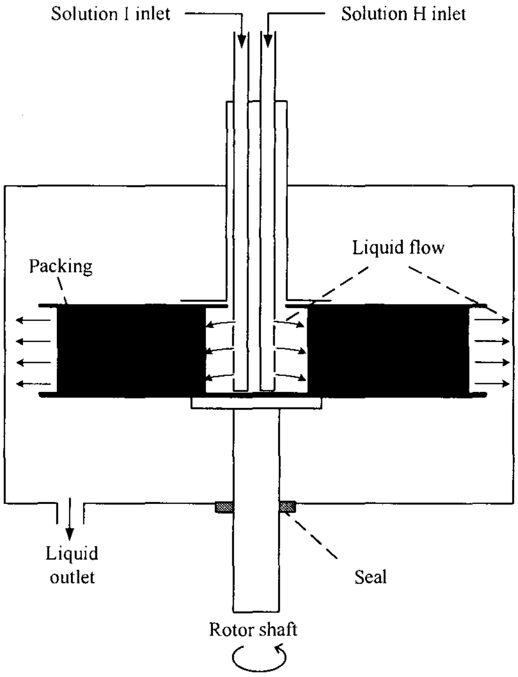

Fig. 1. Main structure of a rotating packed bed.

EXPERIMENTAL SECTION

The main structure of a rotating packed bed is shown in Fig. 1. Solution I and solution H enter the rotating packed bed from two different liquid distributors, respectively. There are three 0.5-mm diameter holes in each liquid distributor, located on opposite sides. Inside the rotor, liquid is pulled through the bed and is split into thin films and small droplets as a result of the centrifugal force. As the liquid leaves the bed, it splashes on the stationary housing and is collected at the bottom. Two rotating packed beds, RPB-1 and RPB-2, were used in this study. The inner and outer radii of the packed bed could be varied from 1 cm to 6 cm in RPB-1. In RPB-2, the inner and outer radii were set at 2 cm and 4 cm, respectively. The heights of both rotating packed beds are 2 cm.

The main structure of the SDR used in this study is shown in Fig. 2. Two liquid solutions flowed onto the disk surface from two glass tubes that were vertical to the disk surface. Both of the tubes ended 0.5-cm above the disk surface. The radial distance between the tube and the axle center of the disk was 1 cm. The disk made of PET film had a radius of 4 cm. The specifications for RPB-1, RPB-2, and the rotating disk, the types of packing used, and the operating conditions are listed in Table 1.

Two solutions containing reactants were prepared for the chemical test reaction. For solution I, boric acid and sodium hydroxide were first added to water, and then potassium iodate and potassium iodide were introduced into the solution. This sequence

Table 1. The specifications of RPB-1, RPB-2, and SDR, and the operating conditions.

Specifications and Operating Conditions	RPB-1	RPB-2	SDR
r_i (cm)	1~5	2	–
r_o (cm)	2~6	4	4
Packing used	wire mesh	wire mesh	–
Rotational speed (rpm)	600~1500	600~2300	600~2300
Q (mL/min)	572	260~900	260~900

Table 2. The initial concentrations and the flow rate ratios of the reactants.

Equipment	Solution I			Solution H	Q_I / Q_H
	$C_{IO_3^-,0}$ (mol/L)	$C_{I^-,0}$ (mol/L)	$C_{H_2BO_3^-,0}$ (mol/L)	$C_{H^+,0}$ (mol/L)	
RPB-1	2.3×10^{-3} [a]	1.2×10^{-2} [a]	0.09 [a]	0.1 [a]	7 [a]
RPB-2	4.0×10^{-4}	2.0×10^{-3}	0.05	0.4	11
SDR	4.0×10^{-4}	2.0×10^{-3}	0.05	0.4	11

[a] same as Monnier *et al.* (2000)

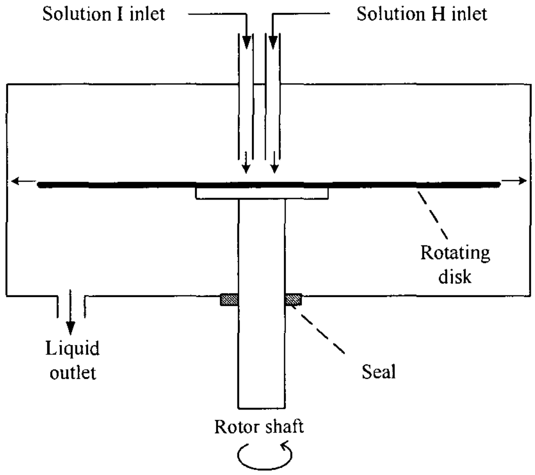

Fig. 2. Main structure of a spinning disk reactor.

ensured that iodide and iodate ions would coexist in the basic solution and prevent iodine formation. The pH of solution I was maintained at 10.9 throughout this study. The other solution, H, was dilute sulfuric acid. The initial concentrations of the reactants and the flow rate ratios are listed in Table 2. Both solutions were pumped into the rotating packed bed and mixed with each other in the bed at 25°C. The concentration of I_3^- in the outlet stream was measured with a spectrophotometer (Spectronic Genesys 5) at 353 nm. The measurement was carried out within 3 min after a sample was taken.

In addition, the synthesis of nanosized magnesium hydroxide particles was performed in both an RPB and an SDR in this study. A liquid-liquid reaction system was to mix the magnesium chloride solution and sodium hydroxide solution. The concentrations of the magnesium chloride solution and sodium hydroxide solution were 0.83 mol/L and 1.66 mol/L, respectively. Liquid flow rates ranging from 320 mL/min to 750 mL/min were investigated, and the rotational speed was set at 2000 rpm. The particle size of the magnesium hydroxide was measured with a zeta sizer (Malvern, 3000 HSA).

RESULTS AND DISCUSSION

The dependence of the segregation index on the rotational speed is shown in Figs. 3 and 4. It is not surprising that *Xs* decreased as the rotational speed increased in all cases, indicating that the centrifugal force could effectively enhance micromixing. This is probably due to the high relative velocity between the liquid droplets in the packing, based on our previous findings (Chen *et al.*, 2004) and visual observations of liquid flow in an RPB made by Guo *et al.* (2000). The liquid was vigorously fragmentized by the fast rotating packing into small droplets and thin films in the entrance region. Some of them were captured by the packing and some kept traveling through the void space. Then, vigorous impingement of the liquid occurred, resulting in good mixing efficiency. As the rotational speed increased, the azimuthal velocity of the liquid on the packing increased, and the impingement among liquid elements became more vigorous. Thus, better micromixing efficiency was achieved.

The effect of the radial position of the packing on the micromixing efficiency was investigated in this study. Figure 3 shows the dependence of *Xs* on the rotational speed for different inner radii of the packed bed in RPB-1. The inner radii were 1, 3, 4, and 5 cm, respectively, while the outer radius was set at 6 cm. It is obvious that *Xs* decreased as the inner radius of the bed increased. When the inner radius of the bed increased, the impingement of the liquids became more vigorous due to the increasing azimuthal velocity of the liquid on the packing; therefore, the micromixing efficiency increased.

Figure 4 shows the dependence of *Xs* on the rotational speed for different outer radii of the packed

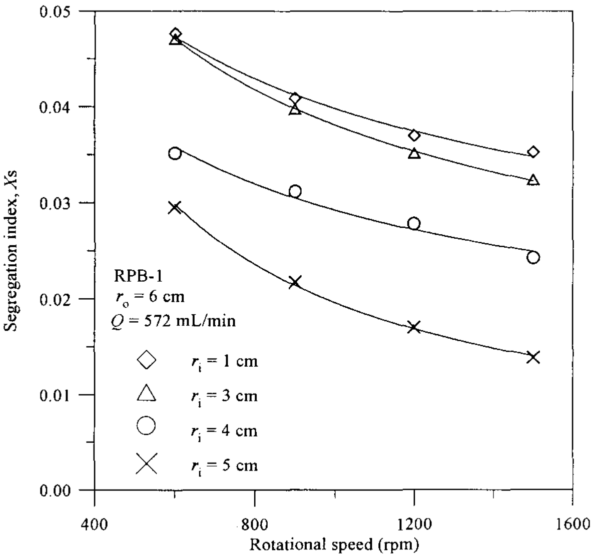

Fig. 3. Dependence of the segregation index (Xs) on the rotational speed under various inner radii of the packing.

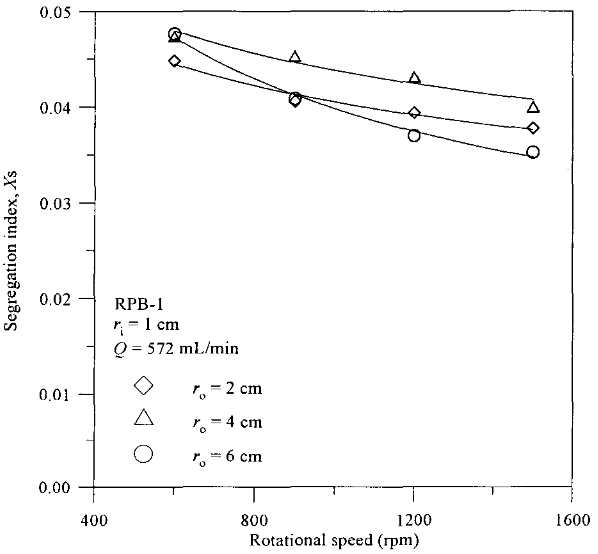

Fig. 4. Dependence of the segregation index (Xs) on the rotational speed under various outer radii of the packing.

bed in RPB-1. The outer radii were 2, 4, and 6 cm, respectively, while the inner radius was set at 1 cm. From Fig. 4, it can be seen that the variation of Xs for the three outer radii was not significant. This result implies that the micromixing efficiency may already be determined in the bed near the inner radius. Increasing the radial thickness of the packing beyond this region did not significantly improve the micromixing efficiency, even though the centrifugal force increased with the radial position. The experimental results shown in Figs. 3 and 4 indicate that intense mixing occurred within the region of initial 1 cm from the inner radius of the packed bed. In addition, increasing the rotational speed and inner radius of the bed could effectively intensify micromixing. The enhancement of the micromixing efficiency could be mainly attributed to the increasing relative velocity as the liquid collapsed into small elements, such as films and droplets.

The micromixing efficiency of the SDR was also investigated in this study. Figure 5 shows the dependence of Xs on the rotational speed for the SDR and RPB-2 under four different liquid flow rates. As expected, the micromixing efficiency of the SDR increased with the rotational speed. In addition, it is clear that the SDR could provide better micromixing than the RPB at a low liquid flow rate. From Fig. 5(a), it is found that the segregation indices of RPB-2 were about 10 times higher than those of the SDR at a liquid rate of 260 mL/min. This is probably due to maldistribution of the liquid in the RPB under a low liquid flow rate (Burns and Ramshaw, 1996). The packing surface in the RPB was not completely wet, resulting in less mass transfer and lower mixing efficiency. However, in the SDR, the liquid was evenly distributed on the disk surface under a low liquid flow rate. Thinner liquid films were obtained in the SDR at a low liquid flow rate; therefore, better micromixing was achieved. In addition, it is noteworthy that the micromixing efficiency in the SDR clearly decreased as the liquid flow rate increased, while the efficiency of the RPB increased. At a liquid rate of 900 mL/min and a rotational speed of 600 rpm, the Xs for the SDR was higher than that for RPB-2, as shown in Fig. 5(d). As the liquid flow rate increased, the thickness of the liquid film in the SDR increased, which caused the micromixing efficiency to decrease. In the RPB, the variation of the thickness of the liquid film may be relatively less as the liquid flow rate increased due to increased wetting of the surface area of the packing. As a result, the SDR is suitable for liquid mixing at low liquid rates, while the RPB provides higher micromixing efficiency at high liquid flow rates.

Nanosized magnesium hydroxide particles were also synthesized in the RPB and in the SDR in this study. A liquid-liquid reaction system was used to mix magnesium chloride solution and sodium hydroxide solution. The particle size was expected to reflect the micromixing efficiency of the devices (RPB and SDR) due to the fast reaction rate. The particle size distribution of the magnesium hydroxide remained about the same under different liquid flow rates in the RPB and SDR, although some larger particles appeared in the SDR at a liquid rate of 750 mL/min. Figure 6 shows the dependence of the mean particle size on the liquid flow rate in the RPB and SDR. It is found from the figure that as the liquid flow rate increased, the particle size decreased in the RPB but clearly increased in the SDR. In addition, at

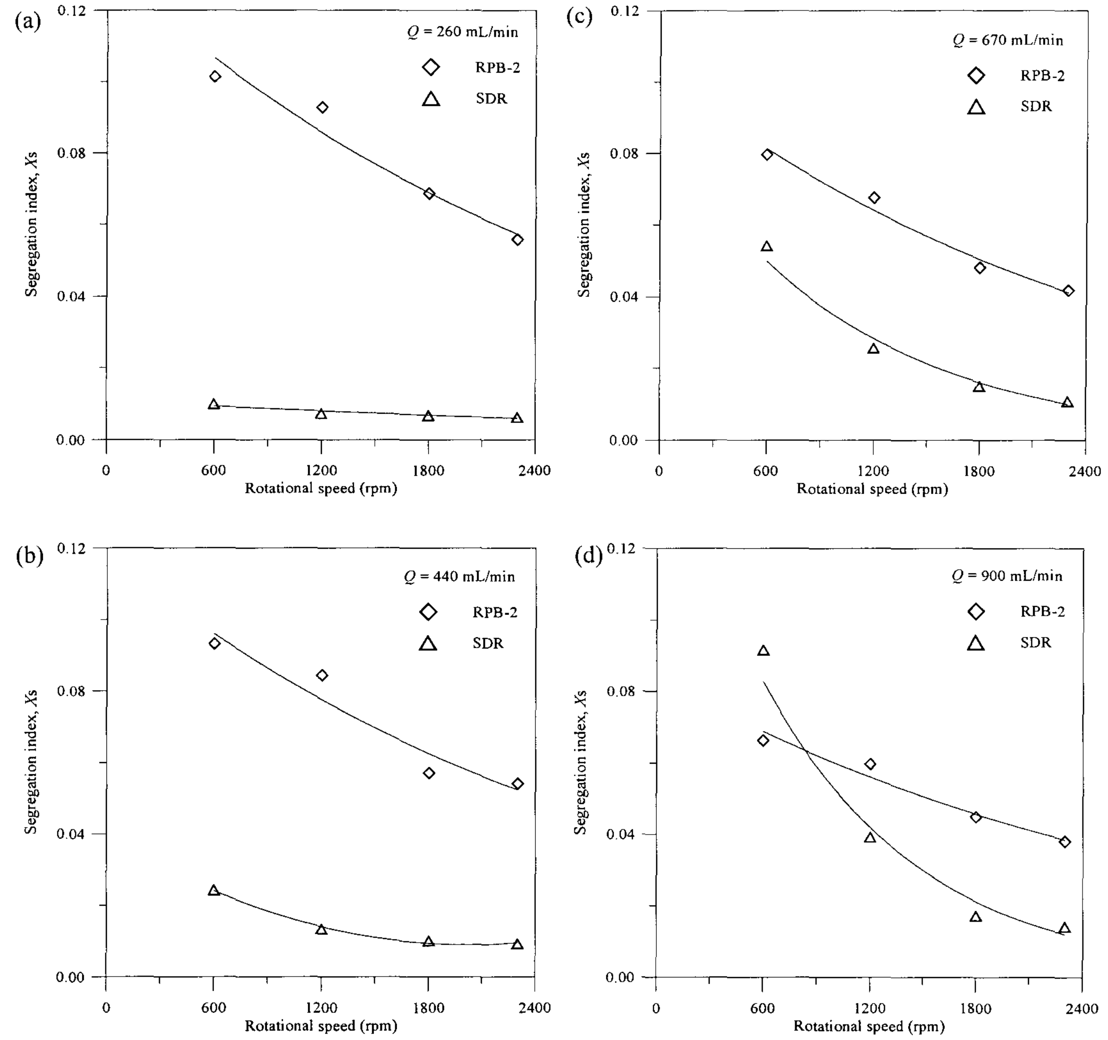

Fig. 5. Dependence of the segregation index (Xs) on the rotational speed in RPB-2 and the SDR. (Liquid flow rate: (a) 260 mL/min; (b) 440 mL/min; (c) 670 mL/min; (d) 900 mL/min).

a low liquid flow rate, smaller particles were obtained in the SDR than in the RPB. However, as the liquid flow rate increased, the particles prepared in the RPB clearly became smaller than those in the SDR. These findings are similar to the experimental results for micromixing obtained in the RPB and SDR, as shown in Fig. 5. The results indicate that increasing the micromixing intensity could reduce the particle size, which conforms to the finding of Marcant and David (1991).

The micromixing efficiency of several kinds of mixing devices has been investigated on the basis of the same chemical test reaction used in this study. In 1999, Liu and Lee (1999) measured the micromixing efficiency of a Couette flow reactor. They found that Xs decreased from 0.95 to 0.14 as the rotational speed increased. They attributed the enhancement to the transition from laminar Couette flow to turbulence. Fang and Lee (2000) investigated the segregation index in a continuous stirred tank reactor (CSTR). Their results showed that both the mechanical agitation and the position of the feed had a strong influence on the micromixing efficiency and that the segregation indices ranged from 0.15 to 0.7. Fang and Lee (2001) studied micromixing in a static mixer. They found that, though the pressure drop in a static mixer was higher, better micromixing was achieved. The segregation indices ranged from 0.03 to 0.7. In 2000, Monnier *et al.* (2000) studied micromixing in a continuous flow cell, enhanced by ultrasound. The reactant concentrations and the liquid flow rate ratio in their investigation were exactly the same as those in RPB-1, as shown in Table 2. They found that increasing the ultrasound power could improve the micromixing efficiency, as Xs varied from 0.03 to 0.07 with Q = 572 mL/min. Figure 7 shows compares the segregation indices in various mixing devices. It is clearly found that the segregation index in the Higee system was lower than that in the other mixing devices. This indicates that, with the aid of centrifugal force, better micromixing efficiency can be achieved in a Higee system.

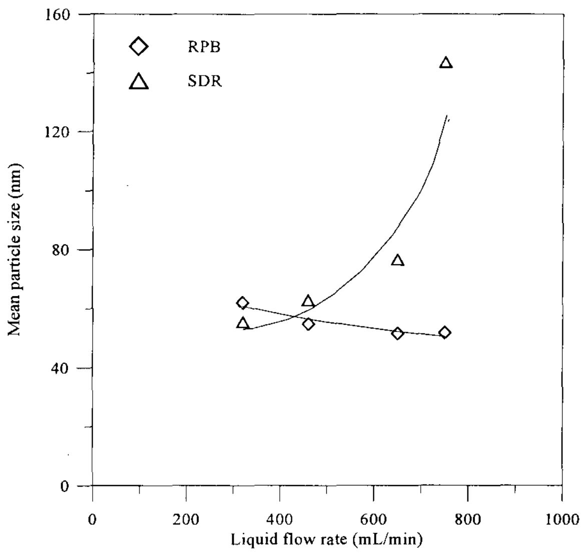

Fig. 6. Dependence of the particle size on the liquid flow rate in the RPB and SDR.

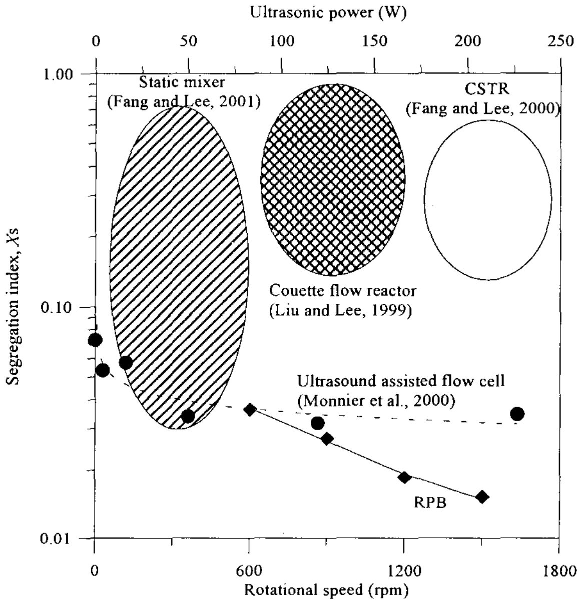

Fig. 7. Comparison of the segregation indices of various mixing devices.

CONCLUSION

This work investigated the micromixing efficiency in a rotating packed bed, based on a parallel competitive reaction scheme, as a function of the rotational speed and inner and outer radius of the packed bed. Experimental results showed that the segregation index decreased as the rotational speed and inner radius of the bed increased. The segregation index did not change significantly under various outer radii of the bed. These findings indicate that the enhancement of the micromixing efficiency was due to the increased relative velocity of impingement among the liquid elements, especially in the entry region, located roughly the initial 1 cm from the inner radius of the bed. Increasing the thickness of the packing beyond this region did not influence the micromixing efficiency. It is also noteworthy that higher micromixing efficiency could be achieved in the SDR than in the RPB at a low liquid flow rate. However, as the liquid rate increased, the RPB achieved higher efficiency than the SDR did. In addition, the experimental results for the synthesis of magnesium hydroxide particles in the RPB and SDR show that smaller particles were obtained in the RPB than in the SDR at a high liquid flow rate. Considering the reported segregation indices obtained with other mixing devices, the Higee system is a promising alternative approach to increasing the micromixing efficiency.

ACKNOWLEDGEMENT

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NOMENCLATURE

C_i	concentration of product *i*, mol/L
$C_{i,0}$	initial concentration of reactant *i* before mixing, mol/L
K_B	equilibrium constant, L/mol
n_i	molar flow rate of product *i*, mol/s
$n_{i,0}$	molar flow rate of reactant *i* before mixing, mol/s
Q	total volumetric flow rate of the two solutions = $Q_{\mathrm{H}} + Q_{\mathrm{I}}$, mL/min
Q_{H}	volumetric flow rate of solution H, mL/min
Q_{I}	volumetric flow rate of solution I, mL/min
r_i	inner radius of packing support, cm
r_o	outer radius of packing support, cm
T	temperature, K
X_S	segregation index
Y	yield of reaction (2)
Y_{ST}	yield of reaction (2) in the case of total segregation

Subscripts

H	solution H
I	solution I

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旋轉填充床中微觀混合特性之探討

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摘　要

本研究利用一平行競爭化學反應量測不同半徑旋轉填充床之微觀混合效率，實驗結果顯示，增加填充床轉速與內半徑能夠提升混合效率，而外半徑的變化則對微觀混合無明顯影響。此外，本研究分別以旋轉填充床和旋轉盤為反應器製備氫氧化鎂顆粒，結果顯示在低液體流量下於旋轉盤中可以製備較小的氫氧化鎂顆粒，當液體流量提升時，旋轉填充床中能夠得到較小的顆粒。將旋轉填充床之分離指標與其他混合系統比較，可以發現超重力系統確實具有較佳的微觀混合效率。