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Short communication

Micromixing efficiency of rotating packed bed with premixed liquid distributor

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ABSTRACT

Rotating packed bed (RPB), in which high gravity is simulated by a centrifugal force, plays an important role in process intensification of fluid mixing and mass transfer. However, uneven initial liquid distribution in RPB leads to poor micromixing efficiency in local areas of the packing. Therefore, a premixed liquid distributor is proposed in this work to improve the liquid distribution in RPB. Micromixing efficiency of RPB with the premixed liquid distributor was studied by adopting the iodide–iodate reaction as working system and show better micromixing efficiency compared to that of RPB with non-premixed liquid distributor. Also, the effects of operating conditions (e.g. rotational speed, acid concentration, volumetric flow rate) and geometries using the premixed liquid distributor on micromixing efficiency (characterized by segregation index X_S) were investigated. The results show that segregation index X_S decreases with the increase of rotational speed, and the decrease of acid concentration and volumetric flow rate.

1. Introduction

The process of turbulent mixing is very complex. In this complex process, one can distinguish and describe some simpler stages of mixing, i.e. macromixing, mesomixing and micromixing [1]. The process of mixing on the scale of the whole vessel is called ‘macromixing’ and refers to those large-scale flow processes that cause the realization of large-scale distributions like the residence time distribution (RTD) or distribution of mean concentration in the Eulerian frame. Hence, macromixing determines the environment concentrations for mesomixing and micromixing, and conveys fluids that are undergoing meso- and micro-mixing through environments where the turbulence properties vary. Mesomixing reflects the coarse-scale turbulent exchange between the fresh feed and its surroundings; spatial evolution of the feed plume can be identified with the process of turbulent diffusion. Another aspect of mesomixing is related to the inertial-convective disintegration of large eddies. The structure of large eddies or spots of contaminant of the inertial-convective subrange determine the environment for micromixing. Micromixing, the last of the turbulent mixing stages, concerns with those features of mixing which cause the attainment of homogeneity on the molecular level, i.e. with the reduction of the scale of unmixed blobs of fluid by breakage and deformation, and with final mixing by molecular diffusion [2]. When the micromixing time is comparable to the reaction characteristics, complex chemical dynamics can alter. Therefore, industrial processes, such as polymerization, crystallization and combustion are significantly influenced by micromixing [3,4]. Even if perfect macromixing has been achieved, it does not necessarily imply that the concentrations of the individual molecules are homogeneous on a molecular level (perfect micromixing). Non-homogeneity related to the meso- and macro-mixing have an important indirect impact on the fast reaction since they determine the environment for micromixing.

Rotating packed bed (RPB), also called HIGEE (an acronym for high gravity), is one of the process intensification apparatuses that promote size and weight reduction, enhance inherent safety with lower inventories, improve energy consumption, lower capital cost, and address environmental concerns [5]. RPB, taking advantages of centrifugal force to simulate a high gravity environment, was invented for intensifying gas–liquid mass transfer efficiency by Ramshaw and Mallinson [6] and have been applied to gas–liquid processes successfully such as desorption, absorption, distillation, ozone oxidation, etc. [7–10]. In recent years, there has been an emerging trend towards the application of RPB in liquid–liquid precipitation processes such as the production of drug nanoparticles and inorganic nanoparticles [11,12]. These pioneering efforts show that RPB is a promising novel reactor for fast liquid–liquid reactions because of excellent global micromixing efficiency [13,14]; however, liquid non-uniform distribution truly exists in RPB, leading to poor micromixing efficiency in local areas of the packing [15]. In our previous work, micromixing efficiency of RPB with non-premixed liquid distributor was investigated [14]. There are two

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Nomenclature

A	solution containing $H_2BO_3^-$, I^-, IO_3^-
B	solution of sulfuric acid
V_A	volumetric flow rate of A ($L\,min^{-1}$)
X_S	segregation index
Y	selectivity of iodine
Y_{st}	value of Y in the total segregation case
n_{I_2}	mole number of produced I_2 (mol)
$n_{I_3^-}$	mole number of I_3^- (mol)
$n_{H_0^+}$	mole number of initial H^+ (mol)

disadvantages of using non-premixed liquid distributor. One is that two liquid streams travel in an un-confined space between the distributor and the packing, causing an uneven liquid distribution in the packing of RPB. The other is that it is difficult to scale-up when using non-premixed distributor. So a new premixed liquid distributor was used to improve macromixing and mesomixing in RPB, which led to a better environment for micromixing and enhanced micromixing efficiency. In this work, micromixing efficiency of RPB with premixed liquid distributor was investigated and compared to that of RPB with non-premixed distributor.

2. Experiments

2.1. Experimental setup

RPB unit generally consists of the rotor filled with the packing, casing, liquid inlets, liquid distributor, liquid outlet, gas inlet, gas outlet, motor, etc. [16]. Fig. 1(a) shows a typical RPB unit having a rotor with a vertical axis, and specifications of the RPB are the same as the work of Yang et al. [14]. In order to validate the importance of the inlet region, the vertical-axis RPB was designed to achieve sampling along the packing radius. Fig. 1(b) shows a feeding mode by non-premixed liquid distributor. Fig. 1(c) shows a feeding mode by premixed liquid distributor. Details of the premixed liquid distributor are shown in Fig. 1(d). Diameters of the side pipe and main pipe are 3 mm and 6 mm, respectively. As basic parameters considered, the angle α between side and main pipe is 120° and the length L from the confluence of the pipes to outlet end is 12 mm if not specified. Micromixing efficiency of RPB with these two different type distributors (premixed and non-premixed liquid distributors) was investigated using parallel competing test reactions. The experimental flow chart is shown in Fig. 2.

2.2. Parallel competing reaction system

The parallel competing reaction [17] is described by the following scheme:

$$H_2BO_3^- + H^+ \Leftrightarrow H_3BO_3 \quad \text{(Instantaneous)} \tag{1}$$

$$5I^- + IO_3^- + 6H^+ \Leftrightarrow 3I_2 + 3H_2O \quad \text{(Fast)} \tag{2}$$

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

Reaction (1) is instantaneous while Reaction (2) is fast but on the same order of magnitude as the micromixing process. The rate of Reaction (2) is given by$R_2 = k_2 C_{I^-}^2 C_{IO_3^-} C_{H^+}^2$, where k_2 depends on the ionic strength μ of the medium such as

$$\mu < 0.166\,\text{M}, \quad \log(k_2) = 9.28105 - 3.664\mu^{1/2}$$

$$\mu > 0.166\,\text{M}, \quad \log(k_2) = 8.383 - 1.5112\mu^{1/2} + 0.23689\mu$$

Reaction (3) is an equilibrium reaction and the equilibrium constant can be written as

$$\text{Log}\,K_B = 555T + 7.355 - 2.575\log T,$$

where T is the reaction temperature

In our experiments, solutions A and B were prepared first. The procedure for preparation of solution A was as follows: (1) powders H_3BO_3 (281.017 g) and NaOH (90.909 g) were dissolved in 15 L and 5 L water, respectively. The as-prepared solutions were then mixed to obtain the buffer solution; (2) powders KI (48.42 g) and KIO_3 (12.483 g) were dissolved in 0.5 L and 2.5 L water, respectively. KI and KIO_3 solutions were then added to the buffer solution in sequence; (3) 2 L water was added into the above solution to obtain 25 L of solution A, which was a mixture of iodide (0.01167 mol L^{-1}), iodate (0.00233 mol L^{-1}) and borate ions (0.0909 mol L^{-1}). The solution B was H_2SO_4 solution (0.08–0.13 mol L^{-1}, corresponding H^+ concentration was 0.16–0.26 mol L^{-1}). In the experiments, the test procedure consists of adding a small quantity of solution B (in stoichiometric deficiency) and solution A into the RPB simultaneously.

Under perfect mixing conditions, the injected acid is instantaneously dispersed in the reactive medium and consumed by borates according to Reaction (1), which is infinitely faster than Reaction (2). Otherwise, the aggregation of acid leads to a local overconcentration of acid, which, after complete consumption of local $H_2BO_3^-$, reacts with the surrounding iodide and iodate ions to produce I_2 and I_3^-. The segregation index X_S is defined as Y/Y_{st}, where Y is the ratio of acid mole number consumed by Reaction (2) to the total acid mole number injected, and Y_{st} is the value of Y in the total segregation case when the micromixing process is infinitely slow. Hence

$$Y = \frac{2(n_{I_2} + n_{I_3^-})}{n_{H_0^+}} \quad \text{and} \quad Y_{st} = \frac{6(IO_3^-)_0}{6(IO_3^-)_0 + (H_2BO_3^-)_0}$$

When the two solutions mix in the RPB, chemical species diffuse and reactions take place, and the effluent composition is indicative of the mixing performance. The amount of produced I_3^- in the effluent was detected by a spectrophotometer at 353 nm (UV-2501PC, Shimadzu Corporation, Japan). Since the amount of produced I_3^- can be measured, the only unknown variable is n_{I_2} which can be calculated in terms of Eq. (3). All the experiments were conducted at temperature of 20 ± 1 °C.

3. Results and discussion

3.1. Comparisons of X_S between premixed and non-premixed distributors

There are two feeding modes, i.e. non-premixed and premixed as shown in Fig. 1(b) and (c), respectively were considered. Usually, non-premixed distributor is adopted in the design of RPB. However, non-premixed distributor will cause a lot of problems in chemical processes and reactions, such as non-uniform liquid distribution in RPB [15]. Two liquid streams entering RPB via a non-premixed distributor may not contact fully with each other in the packing, thus leading to an incomplete reaction between them. For overcoming the weakness of the non-premixed distributor, a premixed distributor was proposed. Fig. 3 shows the comparison of micromixing efficiency between premixed distributor and non-premixed distributor. It can be observed that micromixing efficiency is enhanced tremendously by adopting premixed distributor. It is obvious that a premixed liquid distributor will make the two feeding fluids mix macroscopically before entering RPB. Thus an improved micromixing environment is provided for the mixing in the packing area of

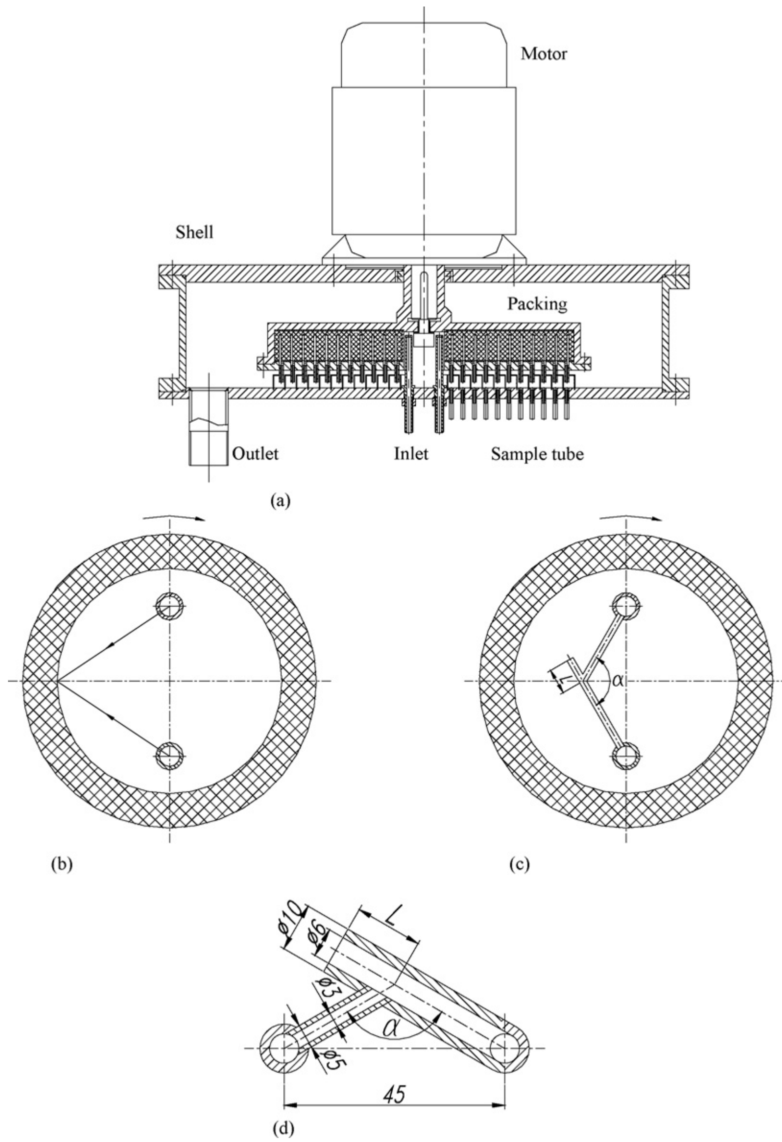

Fig. 1. (a) Schematic diagram of RPB. (b) Feeding mode by non-premixed liquid distributor. (c) Feeding mode by premixed liquid distributor. (d) Details of premixed liquid distributor.

RPB. In the following, micromixing efficiency of RPB with premixed distributor was investigated systematically.

3.2. *Distribution of X_S along the radial position*

It can be seen from Fig. 4 that segregation index X_S decreased sharply at the inlet region and approached a constant value as radial position r increased, which means that micromixing efficiency enhanced with an increasing radial thickness of the packing. The result shows that the inlet region plays an important role in mixing and reaction processes. In the inlet region, the liquid streams make contact with the packing, twisted and broken up by the rotating packing. The interactions are strong as a result of the great relative velocity between radial flowing liquid and the rotating packing. Also, the result shows that X_S values are very small in the whole radial positions and the minimal is near zero, which represent that the injected acid (solution B) was mostly consumed by Reaction (1) and micromixing efficiency of RPB is excellent.

3.3. *Effects of rotational speed on X_S*

In the previous work of Yang et al. [14], the effects of two different rotational speeds (600 rpm and 1200 rpm) on segregation index

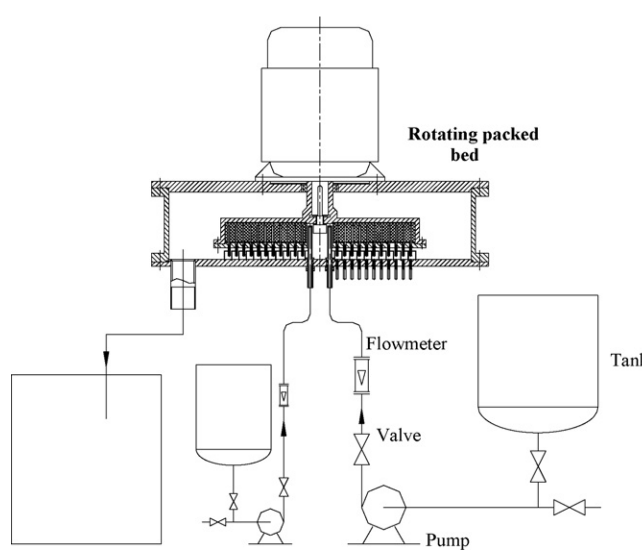

Fig. 2. Experimental setup.

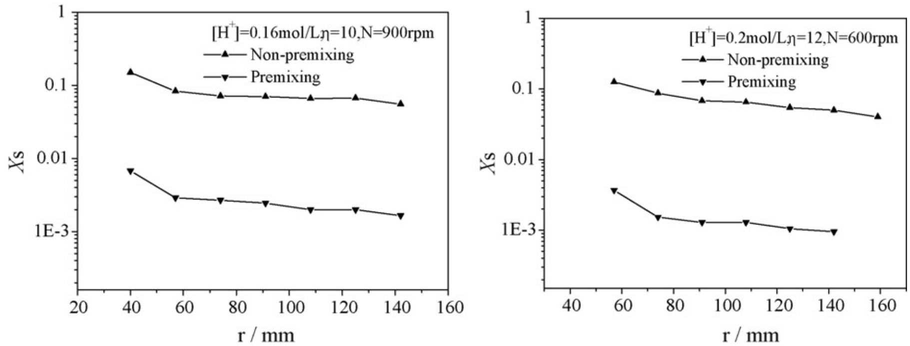

Fig. 3. Comparisons of two different liquid distributors.

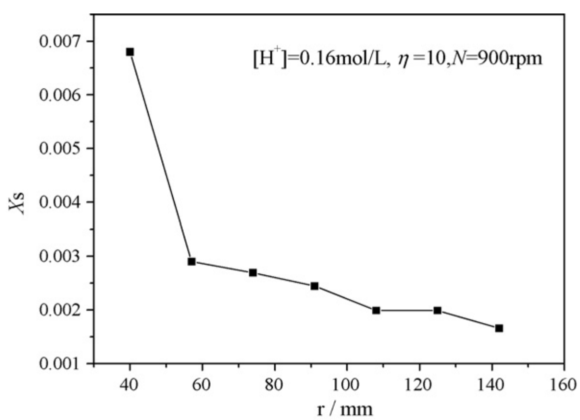

Fig. 4. Distribution of X_S along the radial position.

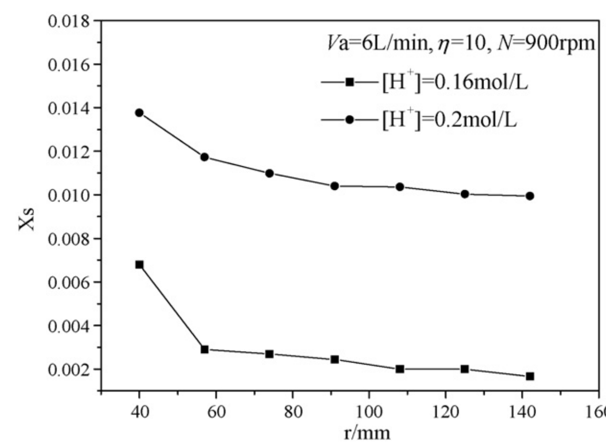

Fig. 6. Effect of reagent concentration on X_S.

X_S were investigated. In order to investigate the effect of rotational speed comprehensively, this work covers a wider range of rotational speeds from 200 rpm to 1200 rpm. The effects of rotational speeds on segregation index X_S were shown in Fig. 5. All of the results showed that a higher rotational speed leads to better micromixing efficiency in RPB, which has similar trends with the results of Yang et al. [14].

3.4. Effects of acid concentration on X_S

The effect of acid concentration on X_S was illustrated in Fig. 6. It can be seen that segregation index X_S increased sharply when acid concentration increased from 0.16 mol/L to 0.2 mol/L. Upon entry into the packing space, the liquid stream is split and dispersed into very smaller entities (i.e., droplets, threads, films). The reactions take place under varying degrees of reactant segregation. A higher acid concentration will lead to a higher segregation of the reagents, causing a local excess of acid concentration, hence, a larger X_S.

3.5. Effects of liquid volumetric rate on X_S

It can be seen from Fig. 7 that X_S decreased as liquid volumetric rate increased, which means that better micromixing efficiency was achieved at higher liquid volumetric rate. This phenomenon may be ascribed to the following two reasons. One reason is that a higher liquid relative velocity between liquid elements and the

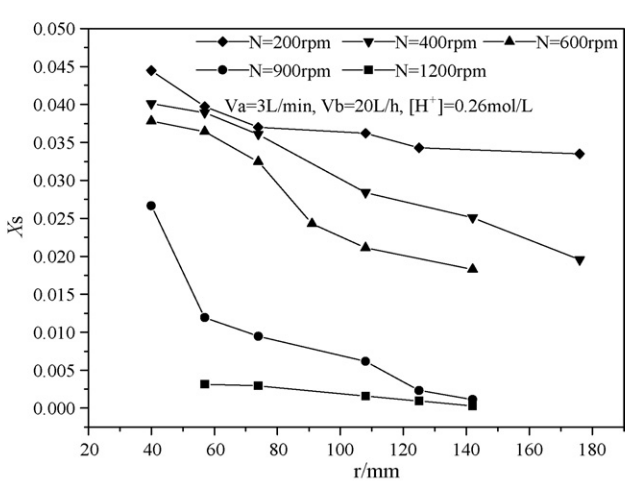

Fig. 5. Effect of rotational speed on X_S.

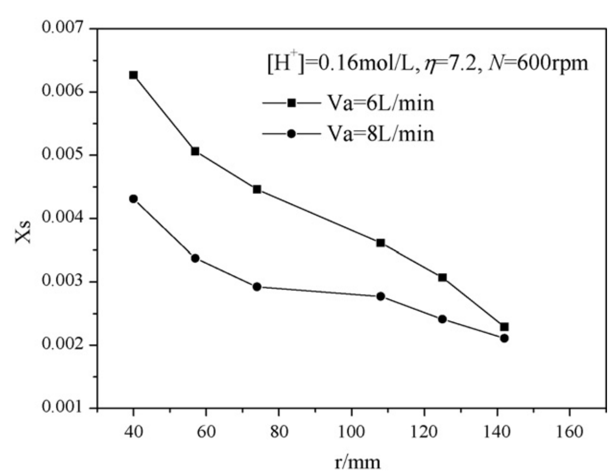

Fig. 7. Effect of volumetric flow rate on X_S.

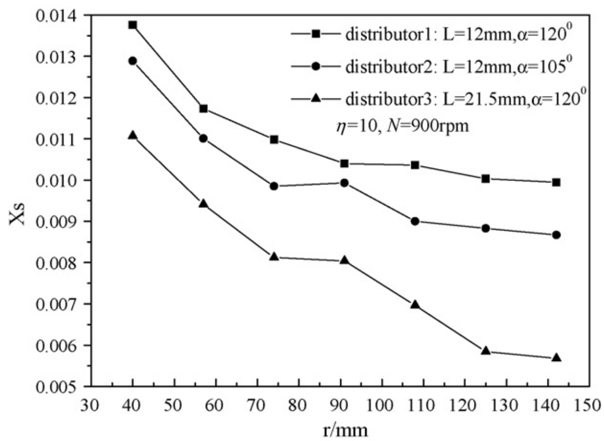

Fig. 8. The effect of geometry on X_S.

rotating packing will be induced with increasing liquid volumetric rate. Consequently, the micromixing efficiency is enhanced as a result of increasing liquid volumetric rate. The other reason is that RTD also decreases with increasing liquid volumetric rate, which may increase the coalescence–redispersion frequency between liquid elements, leading to an intensified mixing.

3.6. The effect of structure and dimensions on X_S

The effect of structure and dimensions on X_S has been investigated using three different premixed distributors (designated as distributor1, distributor2 and distributor3) distinguished by the angle α (between side and main pipe) and the length L (from the confluence of the pipes to outlet end). Other parameters of the three premixed distributors are the same. The results are shown in Fig. 8. Comparison of distributor1 (α and L are 120° and 12 mm, respectively) and distributor2 (α and L are 105° and 12 mm, respectively) shows that micromixing efficiency is better by using distributor2 with a smaller α. While, comparison of distributor1 (α and L are 120° and 12 mm, respectively) and distributor3 (α and L are 120° and 21.5 mm, respectively) shows that micromixing efficiency is also improved by using distributor3 with a larger L. The possible reason is that longer L caused more uniform reactant distribution in macroscopic scale before entering the rotating packed bed. The present investigations only to some extent showed the effects of structure and dimensions on X_S, and systematic investigations will be done in our future research.

4. Conclusions

In this work, a new premixed liquid distributor was used for enhancing micromixing efficiency in RPB. Iodide–iodate test reaction shows that segregation index is very small, which means that micromixing efficiency in RPB is improved enormously by adopting this new distributor. Moreover, at premixed feeding condition, the test reaction results also show that segregation index decreased with an increase of rotational speed and a decrease of H^+ concentration. A slight decrease in segregation index was also observed as the liquid flow rate increased.

More attentions should be paid to the practical industrial applications of RPB based on our experimental results. Also, further work will focus on the optimum design of the premixed distributor and operative parameters in RPB for liquid–liquid mixing.

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