Hydrodynamic characters of co-current operation for SO₂ absorption in a laboratory packed column

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Abstract–A co-current operation for SO₂ absorption by water was performed in a laboratory-scale packed column. The effects of L/V (liquid-gas ratio) and F (gas phase loading factor) on the SO₂ absorptivity were both investigated. The absorptivity for co-current increased with the increase of L/V and the percentage of absorptivity increase at higher L/V is larger. At lower F, in regular packing there is fluctuation of absorptivity with F increased, but in random packing there is not. With the increase of F, the absorption curve slowed down. It is proposed that in order to obtain a steady desulfurization efficiency, F factor in both kinds of packings should be higher than $4\,kg^{0.5}/m^{0.5}s$. For absorptivity, which could be reached by both co-current and counter-current, it is suggested that co-current is better because of the higher gas velocity.

Key words: Sulfur Dioxide, Absorption Column, Packed Column, Co-current, Liquid-gas Ratio, Gas Phase Loading Factor

INTRODUCTION

Absorption is an important chemistry unit operation widely used in distillation and gas treatment processes. The absorption column is a dominant equipment, and its cost occupies the most of the investment. Reactor optimization is thus of great importance. Compared with plate columns, packed columns are welcome because of higher absorptivity, little pressure drop and higher production level [1]. However, due to the lack of some hydrodynamic characteristics, like the flow distribution and so on, the research and industry scaling-up of packed columns today is still carried out by a semi-empirical method. Sun et al. [2] built a theoretical model that can be used to predict the two-phase flow distributions in the random packed column. Before that, the existing models could only predict one-phase flow. But similar researches are still less. In the research for packed columns, most investigations are developed on the basis of gas-liquid counter-current flow. Hoffmann et al. [3] summed up a great number of researches, and put forward a standardization method of mass transfer measurements, and this is in the basis of gas-liquid counter-current absorption. Billet et al. [4] developed a model for the advanced calculation of packed columns and made it possible to calculate the mass transfer characters, but it was on the basis of two-phase counter-current model. Ligy et al. [5] investigated the SO₂ absorption, also on the basis of counter-current flow. Many a research described in the literature has been carried out with the mode of counter-current flow [2,6-8]. But this operating mode has a disadvantage in that flooding appears as increasing the gas velocity. However, in the case of reactive absorption with non-equilibrated reaction, co-current flow allows for higher liquid and gas velocities, without flooding. Fewer studies found in the literature are focused on co-current [9-12]. Raynal et al. [9] mentioned the co-current two-phase down-flow mode in the paper, but focused on the liquid hold-up values. Zidar [10] also studied the basis of co-current operating mode, but the equipment was a falling-film column, not a packed column. Sidi-Boumedine et al. [11] proposed a new correlation calculating the hold-up in the packed column under co-current gas-liquid downward flow.

As for the comparisons between co-current and counter-current flow, most of the experiments described in the literature focus on one or some characters of co- or counter-current flow. There are no papers focusing on the comparisons between them in similar conditions.

The present study was carried out on the co- and counter-current flow in two typical packed columns, regular packing and random packing. In this paper, it was carried out with the methods of comparison: (i) Changing the L/V (liquid-gas flow ratio) and F (gas phase loading factor) respectively, and illuminating by SO₂ absorptivity the characters of the two kinds of packed columns. Different liquid-gas flow levels resulted in the change of liquid hold-up, residence time, transfer efficiency, and the two phase's dynamic characters. All of these were considered and discussed. (ii) Comparing the two operating modes (co- and counter-current) in the similar conditions.

EXPERIMENTAL SECTION

1. Materials and Apparatus

The gas phase was composed of SO₂ (99.99%, purchased from TianjinBohaiDongXiang Technology Development CO., LTD.) and air. The liquid phase was pure water. Two kinds of packings were used: one is regular packing, the 250 X Metal corrugated random packing; the other is random packing, the Dg16 Cascade plastic mini-ring.

Inlet and outlet gas concentrations were measured by KANE 940 gas analyzer (KANE Company, UK). A schematic diagram of the experimental apparatus is shown in Fig. 1)

2. Apparatus and Procedure

During the experiment, the inlet concentration of SO₂ was always

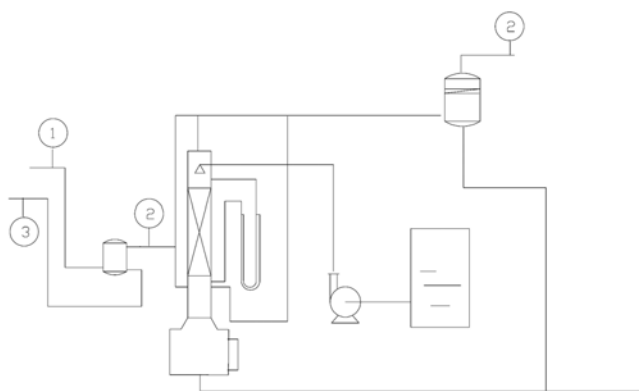[†]To whom correspondence should be addressed.
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Fig. 1. Experiment setup.
  1. A flowmeter for the SO₂ velocity
  2. Gas analyzer for the SO₂ concentration
  3. A flowmeter for air velocity

Fig. 2. Relationship between SO₂ absorptivity and L/V.

controlled below 5,000 ppm to avoid exceeding the measuring range of KANE 940. Experiments were done at a room temperature, using SO₂ and air as the artificial flue gas.

The definition of SO₂ absorptivity is Eq. (1). η_{SO_2} is SO₂ absorptivity. C_{in} and C_{out} are the inlet and outlet concentration, respectively, ppm. All the data could be obtained on the screen.

$$\eta_{SO_2} = \frac{C_{in} - C_{out}}{C_{in}} \tag{1}$$

2-1. SO₂ Absorptivity at Different L/V

First, the gas flow rate (V) was kept constant at 20 m³/h and the liquid flow rate (L) was varied from 40 to 400 m³/h. Both the gas and liquid were fed into the top of the column, and then left from the bottom of column. Second, V was increased to 40 m³/h, and the above procedures were repeated. Once the L/V value was changed, the plateau value was written down when no significant changes were observed. Meanwhile, the phenomenon inside the column was also observed.

2-2. SO₂ Absorptivity at Different F Factor

F factor is gas phase loading factor and expressed in kg⁰·⁵/m⁰·⁵s. The definition of F is Eq. (2). V is gas velocity, m/s. ρ is the gas density, kg/m³.

$$F = V \cdot \sqrt{\rho} \tag{2}$$

The operating conditions were controlled around room temperature and atmospheric pressure. F factor is a function of gas velocity and gas density. Different densities of gases could produce different resistances to liquid trickles, which results in different residence time for liquid trickles.

Because of the limited laboratory condition, only the gas velocity could be changed, but the gas density, connected with the pressure and temperature, couldn't. At atmospheric pressure, gas could be treated as ideal gas. According to the van der Waals equation, the gas density could be treated as a constant, and thus during the whole experiment there were no changes coming from gas density. Besides, in the industry design, it is customary for us to use the F factor. So in order to make the industrial scaling-up easy in the future, in this paper we use the F factor instead of V. Billet and Schultes [4] in their paper put forward an equation including the gas phase loading
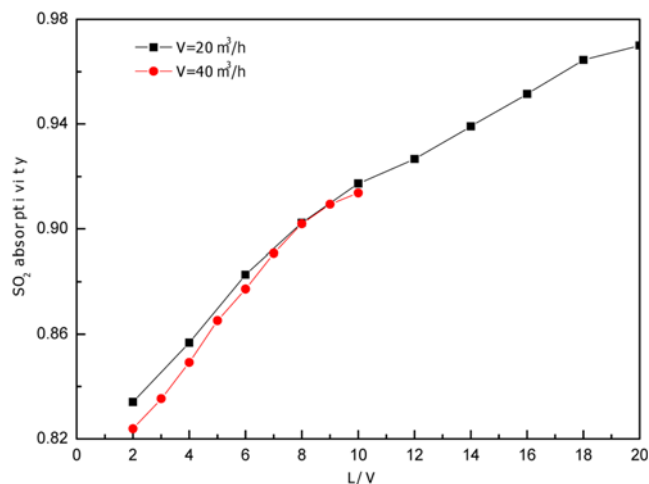
to precalculate the pressure drop. But the use of gas phase loading factor is less investigated in the literature.

In this part, V was adjusted at constant L to control the range of F factor from 1 to 8 kg⁰·⁵/m⁰·⁵s, where SO₂ absorptivity was investigated at three different L/V levels.
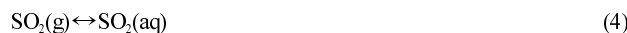
RESULTS AND DISCUSSIONS

1. The Influence of L/V in Co-current Flow

Fig. 2 shows that the SO₂ absorptivity at V=20 m³/h is globally higher than that at V=40 m³/h. The two lines increased with increase of L/V value. L/V=10 is an important point where the gradient turned to decreasing. The SO₂ absorptivity increases by 10% during L/V= 2-10, but less than 5% during L/V=10-20 (Fig. 2).

The gas velocity was constant at V=20 m³/h and V=40 m³/h, respectively. With the increase of L/V, SO₂ absorptivity increased. Thus, the gas velocity has no effect on the present results, and the effect of liquid velocity only was being looked at. As underlined by Sidi-Boumedine et al. [11], the liquid hold-up is an important hydrodynamic parameter for gas/liquid flow in packed columns, and the hold-up indirectly enables the determination of the transfer parameters. With the increase of liquid flow rates, the liquid hold-up tended to saturation. Micro-turbulent motion became more intense because of the increase of L, which can accelerate the diffusion of SO₂ in the water. Effective interfacial area ("a", m²/m³) enlarges with the L increased [11]. As the following Eq. (3) shows, when the effective area is increased, one observes an increase of the molar mass transfer flux (N_A), so the SO₂ absorptivity increases with it [13].

$$N_A = K_y \cdot a \cdot \Delta Y \tag{3}$$

$$SO_2(g) \leftrightarrow SO_2(aq) \tag{4}$$

$$SO_2(aq) + H_2O \leftrightarrow H^+ + HSO_3^- \tag{5}$$

Another reason for the enhancement of SO₂ absorptivity is pH value. More liquid hold-up in the packing resulted in decreasing slowly for solution pH. The Eq. (5) shows the first hydrolyzation of SO₂ in liquid. As we know, dilution is good for hydrolyzation. From Eq. (5), dilution makes more SO₂ (aq) hydrolyze, which decreases the amount of SO₂ (aq). Because of the gas-liquid equilibrium, more
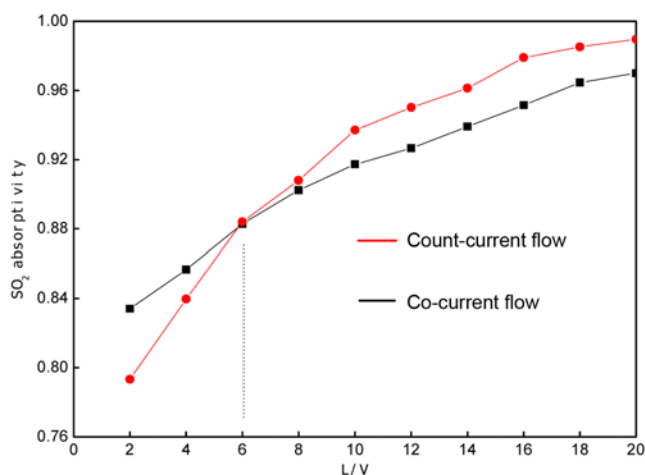
Fig. 3. The comparison between co- and counter-current flow in the condition of regular packing. V = 20 m³/h, L/V = 2-20.

SO_2 (g) would transfer into the liquid, and turned to be SO_2 (aq). Of course, with the increase of liquid hold-up, the SO_2 absorptivity couldn't be increased all the time.

2. Comparison Between Co-current and Counter-current Flow

Fig. 3 represents that when L/V is lower than 6, the SO_2 absorptivity with co-current flow is higher than that with counter-current, while when L/V is higher than 6, the SO_2 absorptivity with counter-current is higher. As we know, there is stronger driving force for mass transfer in the counter-current flow and the absorptivity for counter-current flow should be higher than that for co-current flow. But it is found experimentally that there is a point of intersection in Fig. 3. Lower than L/V=6, the liquid and gas velocities are both slow, so the residence time is relatively long, and thus the contact time for gas and liquid is relatively long. But when L/V is above 6, this advantage weakens gradually (Fig. 3).

It is worth to note that the absorption efficiency would be enhanced as the L/V was increased, but the equipment investment and energy and absorbent consumption are also increased with it. So how to optimize the L/V is very important.

3. The Influence of F Factor in Co-current Flow

The two figures are performed at three different L/V levels, respectively, and with the F factor ranging from 1.5 kg$^{0.5}$/m$^{0.5}$s to 8 kg$^{0.5}$/m$^{0.5}$s. The only difference is the packing, and one is the regular packing but the other is the random packing. Fig. 4(a) shows that lower than F=3 kg$^{0.5}$/m$^{0.5}$s, the three lines all fluctuated, but higher than F=3 kg$^{0.5}$/m$^{0.5}$s, the lines were smooth. Compared with Fig. 4(a), Fig. 4(b) represents three relatively smoothed curves; lower than F=3 kg$^{0.5}$/m$^{0.5}$s, the curves increases, and higher than F=3 kg$^{0.5}$/m$^{0.5}$s, they are the same as the ones in the Fig. 4(a). It is proposed that in order to obtain steady desulfurization efficiency, F factor higher than 3 kg$^{0.5}$/m$^{0.5}$s is better (Fig. 4(a), Fig. 4(b)).

In the above experiment, each curve was obtained at one L/V level. The changed parameter is the gas or liquid velocity. Now consider the following three conditions. (i) Gas velocity was constant, and L was increased, so the liquid hold-up was increased on the surface of packing [4]. As mentioned before, this is beneficial for SO_2 absorption. (ii) Increasing the two factors, gas and liquid flow rates, which made the residence time of two phases shorter. This
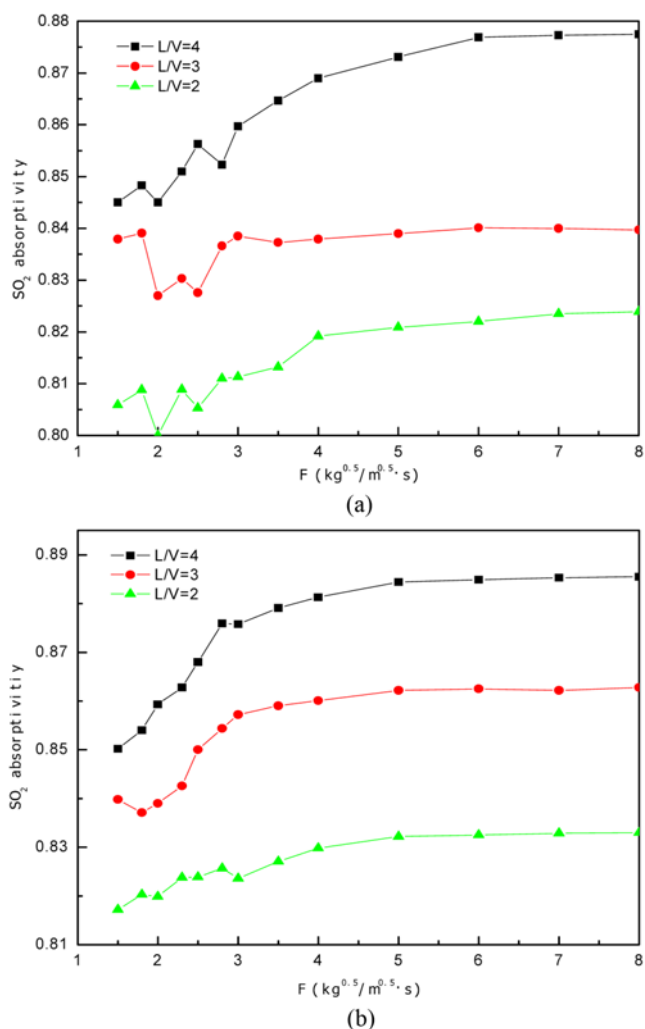

(a)

(b)

Fig. 4. (a) Comparison diagram for desulfurization efficiency of regular packing in the case of co-current operation. (b) Comparison diagram for desulfurization efficiency of random packing in the case of co-current operation.

operation is bad for the gas transmission. (iii) As the following Eq. (8) represents, even though the total transmission was increased with the increase of the gas and liquid flow rates, because of the little transmission driving force (10^{-3}), the last total transmission mass flux is still small. So in the above pictures, the lines don't increase.

$$k_G a = 9.81 \times 10^{-4} G^{0.7} W^{0.25} \quad k_L a = aW^{0.82} \tag{6}$$

Formula conditions:

G(gas velocity)=320-4,150 kg/(m²·h):

W(liquid velocity in the empty column)=4,400-58,500 kg/(m²·h):

Φ=25 mm, ring packing

$$\frac{1}{K_G} = \frac{1}{k_G} + \frac{1}{k_L \times H} \tag{7}$$

$$N_A = K_G(P_A - P_A^*) = K_\gamma (Y - Y^*) \tag{8}$$

4. Comparison between Co-current Flow and Counter-current Flow in the Same Condition

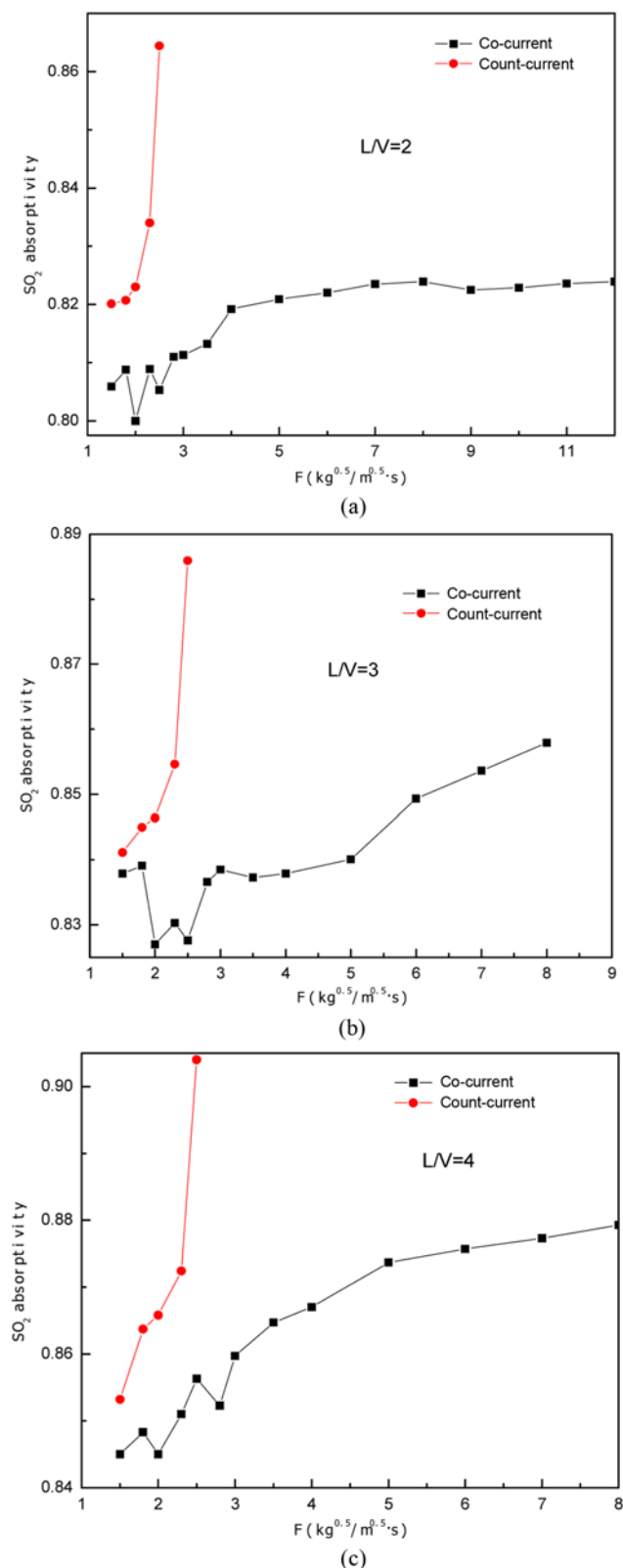All the data in Fig. 5(a), 5(b), 5(c) were obtained in the regular

Fig. 5. (a) At L/V=2, the comparison diagram for desulfurization efficiency of regular packing in two kinds of operating modes. (b) At L/V=3, the comparison diagram for desulfurization efficiency of regular packing in two kinds of operating modes. (c) At L/V=4, the comparison diagram for desulfurization efficiency of regular packing in two kinds of operating modes.

packing, metal corrugated plate packing. The three figures show that at the same F factor, desulfurization efficiency in the co-current flow is lower than that in the counter-current. During the counter-current flow, desulfurization efficiency was enhanced with the increase of F factor. As displayed in the figures, at three different L/V levels and in the condition of counter-current flow, the highest desulfurization efficiency increased from 86% via 89% to more than 90%. But after the F was increased higher than 2.5 kg$^{0.5}$/m$^{0.5}$·s, big bubbles happened somewhere in columns, and with it, the SO$_2$ absorptivity dropped (The test was stopped for the flooding, and the outlet SO$_2$ concentration increased rapidly, which displayed in the screen of gas analyzer). Liquid turned to be continuous phase and gas turned to be dispersed phase [14]. Liquid was carried out of the column with the slightest increase in gas velocity [1], and the absorptivity decreased. Here the gas velocity rose above the loading point, so the liquid flow was totally impeded and nearly no liquid was used for SO$_2$ absorption [4]. During the co-current flow, the desulfurization efficiency was enhanced with the increase of F factor, but the total enhancement was not obvious; meanwhile, the beginning of the curve is fluctuating. The advantage for co-current flow is that with the increase of F, flooding phenomena do not exist. But higher than some F factor, pulse flow appears [15], which is different from the counter-current flow (Fig. 5(a), Fig. 5(b), Fig. 5(c)).

The figures represent that higher than the F=3 kg$^{0.5}$/m$^{0.5}$·s, the desulfurization efficiency for counter-current flow is close to that for co-current flow within the F range from 1.5 kg$^{0.5}$/m$^{0.5}$·s ~2.5 kg$^{0.5}$/m$^{0.5}$·s. For example, at L/V=4, both current flows could reach the SO$_2$ absorptivity ranged from 85-88%.

It is suggested that gas-liquid co-current flow could be considered, if the concentration of flue gas is not rigorous. In the co-current flow, there is no flooding. The cost will decrease while the production will increase.

All the data in Fig. 6(a), 6(b), 6(c) were obtained in the Cascade plastic mini-ring packing. The changing trend in the random packing is nearly the same as in regular packing. The distinction existed with the low F factor. The figures show less fluctuations happened in the random packing than in the regular packing (Fig. 6(a), Fig. 6(b), Fig. 6(c)).

In the progress of the experiment, because of the scrambling of packing, pulsing phenomenon was stronger than that in the regular packing. The phenomenon was good for the liquid refreshment on the surface of packing, which is also helpful for gas transmission [15].

The random packing column is not available for reaction within high pressure. So in the warm condition for absorption, the co-current flow could be considered.

5. An Example for the Advantage of Co-current Flow

The data were calculated according to SO$_2$ absorber. It is supposed that the column inner diameter was 16 m, and the height of packing was 5 m. The price for packing was 8,000/m^3. So the total cost is ¥8.04 million. Increasing the F factor from 4 kg$^{0.5}$/m$^{0.5}$·s to 20 kg$^{0.5}$/m$^{0.5}$·s by decreasing the column diameter and the results are listed in Table 1. It shows that with the increase of F factor, the packing cost decreased largely. Meanwhile, considering the figures about relations of SO$_2$ absorptivity and F factor shown in this paper, SO$_2$ absorptivities tend to steadiness with F>4 kg$^{0.5}$/m$^{0.5}$·s. When some absorptivity could be reached by both co- and counter-current
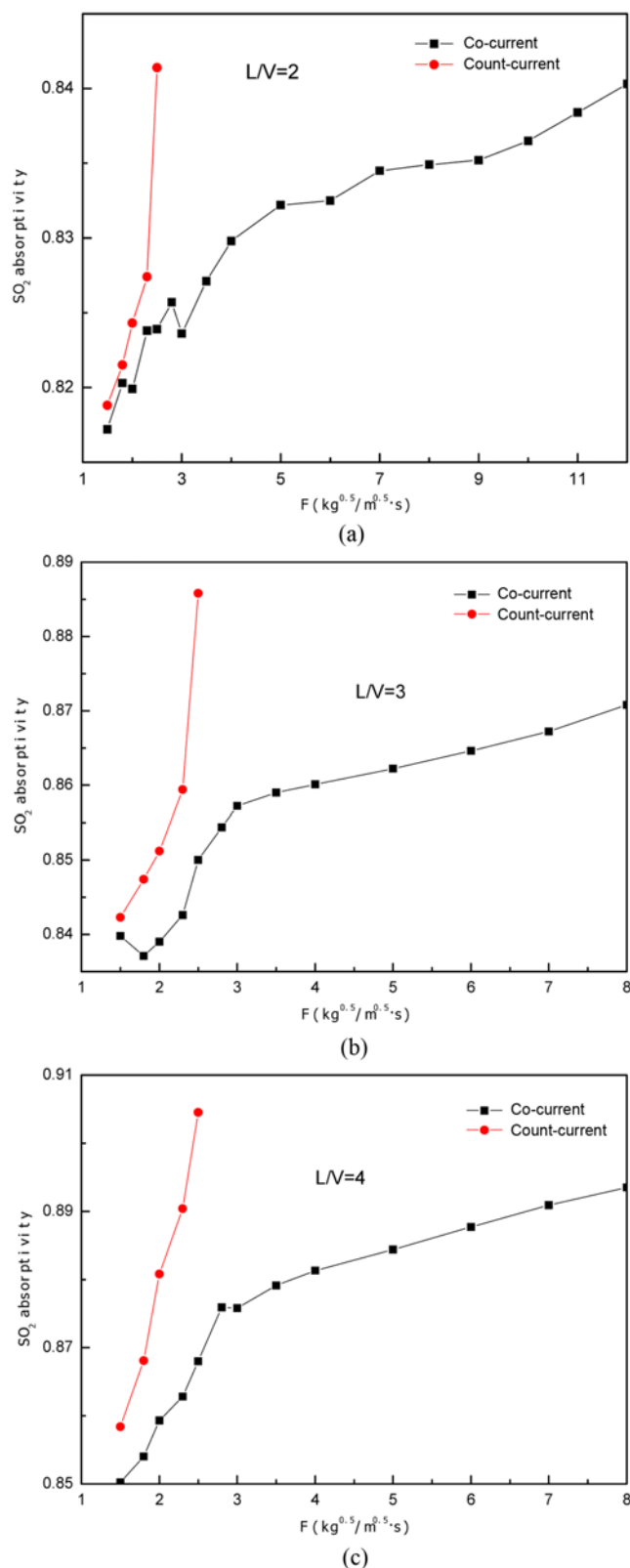
Fig. 6. (a) At L/V=2, the comparison diagram for desulfurization efficiency of random packing in two kinds of operating modes. (b) At L/V=3, the comparison diagram for desulfurization efficiency of random packing in two kinds of operating modes. (c) At L/V=4, the comparison diagram for desulfurization efficiency of random packing in two kinds of operating modes.

Table 1. Data for comparison of packing cost in different design F factors (Not contain the cost of internal parts and column equipment)

F factor ($kg^{0.5}/m^{0.5}\cdot s$)	Diameter/ mm	Packing price/ million	Save percent
4	12650	¥6.03	25%
6	10330	¥4.02	50%
8	8945	¥3.02	62.5%
10	8000	¥2.42	70%
12	7303	¥2.01	75%
14	6760	¥1.73	78.6%
16	6325	¥151	81.3%
18	5963	¥§134	83.3%
20	5657	¥121	85%

operating condition, the former is better, because higher gas velocity could be realized by smaller diameter, and which may result in lower packing price. So in the industry, it is good to consider all factors to gain the biggest benefit. It is suggested that in the condition of co-current flow, F=4-8 $kg^{0.5}/m^{0.5}\cdot s$ is better (Table 1).

CONCLUSION

Experimental study was conducted to investigate the advantages of co-current flow and how to select the operating mode in an absorption process. By changing the gas and liquid velocities, it was found that the co-current system has the following advantages: (i) co-current can be operated steadily under high gas or liquid velocity, and the SO_2 absorptivity also increases with the increase of gas or liquid velocity, but the percentage of desulfurization efficiency increase turned down at some velocity, as appeared in the present paper, (ii) high velocity can be obtained by decreasing the column diameter, which reduces the cost, and (iii) high velocity enhances the productivity and throughput. Based on the figures, from Fig. 5 to Fig. 6, the higher L/V, and saving on packing price, for the system in this paper, it is suggested that in the condition of co-current flow, F=3-8 $kg^{0.5}/m^{0.5}\cdot s$ is better.

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