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# Mass transfer of acrylonitrile wastewater treatment by high gravity air stripping technology

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## ABSTRACT

Air stripping is the one of effective technologies for removing volatile organic compounds from wastewater. However, the low removal rate of acrylonitrile in wastewater by air stripping at ambient temperature and the huge stripping column remain a technical problem. Hence, high gravity is adopted to intensify air stripping process which is strongly affected by gas–liquid mass transfer. The effects of high gravity factor, gas–liquid ratio, liquid spray density and initial concentration of acrylonitrile wastewater on the liquid overall mass transfer coefficient and the removal rate of acrylonitrile were investigated separately. Under the suitable conditions, the liquid overall mass transfer coefficient and acrylonitrile removal efficiency could reach 0.906 kmol $m^{-3}$ $s^{-1}$ and 69.1%, respectively. Furthermore, correlations of liquid overall mass transfer coefficient and removal rate of acrylonitrile were established. Comparing experimental data with fitting data, the average relative errors are both below 4%, revealed that the accuracy of correlations was reasonable. In addition, high gravity air stripping and conventional technologies were compared. The obtained results imply great potential and good economic benefits of high gravity air stripping technology in the treatment of acrylonitrile wastewater.

*Keywords:* High gravity; Stripping; Mass transfer; Wastewater; Purification

## 1. Introduction

As an important chemical industrial raw material, acrylonitrile is widely used in the manufacturing of acrylic fiber, nitrile rubber, acrylonitrile–butadiene–styrene (ABS) plastic and synthetic resins. However, much toxic acrylonitrile wastewater of high concentration is released in the production [1]. It increases the environment pollution and human health risks since acrylonitrile is the one of volatile organic compounds. In spite of high treatment cost and secondary pollution, incineration is most frequently used in the industrial treatment process with its completeness. Pressured hydrolysis and wet oxidation process have not achieved good industrial application due to high energy consumption and operation cost. So, the available and practical treatment method for this wastewater has represented a technological and critical problem to satisfy the effluent discharge standards [2].

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Traditionally, acrylonitrile wastewater can be treated by air stripping [3], adsorption [4], advanced oxidation [5], distillation [6], biological treatment [7], or a combination [8] of these methods, etc. [9,10]. For the removal and recovery of volatile organic compounds in water, air stripping is assumed to be the best known method technically and economically [11,12]. In general, air stripping is a process which involves the mass transfer of acrylonitrile from the liquid phase to the gas phase [13]. Recent research demonstrates that the removal rate of $BOD_5$ from ABS manufacturing wastewater by air stripping was in the range of 2–10% [14]. Freeman determined the relative rates of air stripping and biological removal of acrylonitrile from activated sludge systems [15]. In order to get a high removal rate, air stripping is usually operated in a packed tower because it can provide a larger mass transfer area. However, in practice, air stripping column is as high as 20 m and the installed column cost is very high. The main reason is that acrylonitrile is dissolved in the water easily and its solubility can be reached 73,000 mg $L^{-1}$ at ambient temperature. Mass transfer only depends on gravity in the ordinary equipment such as packed tower or aeration tank which results in lower removal rate and mass transfer performance. Consequently, mass transfer process between liquid and gas phase should be intensified.

As a novel process intensification technology, high gravity technology has been utilized to save energy, reduce cost, and increase benefit and made an outstanding contribution to the chemical industry [16]. On the basis of this technology, rotating packed bed (RPB) is developed and its applications have been involved in many unit operations such as stripping [17–19], adsorption [20,21], distillation [22], preparation of nanoparticles [23], etc. In a RPB, the fluids are split or spread into very fine droplets, threads, and thin films [24,25] by the strong high gravity, thus intensifying micromixing and mass transfer process significantly transfer coefficient in a RPB is 1–3 orders of magnitude higher. Therefore, an equipment size may be smaller, thereby benefiting a reduction in capital and operation costs.

In this study, high gravity technology is first introduced to enhance the gas–liquid mass transfer between air and acrylonitrile wastewater. The main focus of this study is to systematically study the effects of high gravity factor, gas–liquid ratio, liquid spray density, initial concentration of acrylonitrile wastewater on liquid overall mass transfer coefficient and removing rate of acrylonitrile. The mass transfer coefficient and removal rate could be estimated through the established correlations to provide guidance for industrial application.

## 2. Experimental

### 2.1. Experimental setup

High gravity air stripping experiments were conducted in a RPB. The specification of rotor is given in Table 1. The packing used in this study is Pall ring of 16 mm diameter.

### 2.2. Experimental process

Acrylonitrile wastewater was prepared by dissolving acrylonitrile in water at ambient temperature and pressure. The acrylonitrile in wastewater was at a concentration of 3,000 ± 100 mg $L^{-1}$.

Schematic diagram of high gravity air stripping acrylonitrile wastewater was demonstrated in Fig. 1. Acrylonitrile wastewater was pumped from the stock tank into the RPB through a liquid inlet and sprayed the inner edge of the packing via a liquid distributor. The air was introduced into the RPB through a gas inlet. Acrylonitrile wastewater and air contacted countercurrently in the RPB. Consequently, acrylonitrile in the liquid stream was stripped in the gas stream. The wastewater moved outward and left through the outer edge of the packing under the centrifugal force. After

Table 1
Details of rotor

| Inner diameter (mm) | Outer diameter (mm) | Height (mm) |
|---|---|---|
| 30 | 64 | 30 |

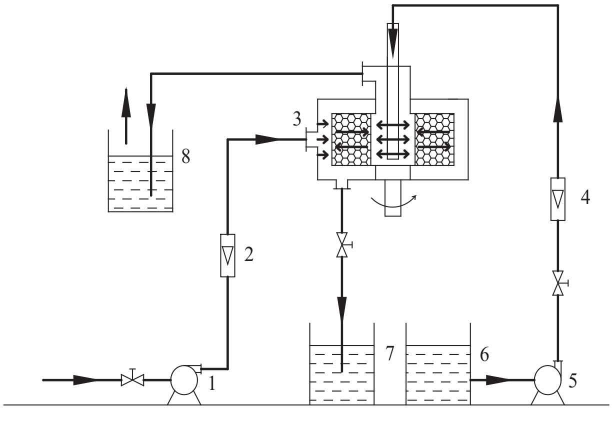

Fig. 1. Schematic diagram of experimental equipment procedure (1) blower; (2, 4) rotor flow meter; (3) counter-flow RPB; (5) pump; (6) stock tank; (7) waste solution storage; (8) absorption solution storage.

Table 2
Details of analytical conditions

| Parameter | Value (℃) | Parameter | Value |
|---|---|---|---|
| Injection-port temperature | 140 | Carrier | 30 mL min$^{-1}$ N$_2$ |
| Detector temperature | 180 | Hydrogen | 40 mL min$^{-1}$ |
| Column temperature | 90 | Air flow | 400 mL min$^{-1}$ |

high gravity air stripping, gas stream containing acrylonitrile was absorbed by the sodium hydroxide solution. The temperature and the pressure in the RPB were about 20℃ and 0.1 MPa (absolute pressure), respectively.

## 2.3. Analysis

### 2.3.1. Determination of the acrylonitrile concentration in the liquid phase

The acrylonitrile concentration in the inlet and outlet liquid stream is measured by gas chromatography. The gas chromatography instrument of GC7900, detector of FID, and packed column (1 m × 3 mm) is used. 60–80-mesh GDX-502 is packed in column. The detailed analytical conditions are given in Table 2. The injection size is 2.0 μL.

### 2.3.2. Determination of liquid overall mass transfer coefficient $K_xa$

Air stripping process for acrylonitrile wastewater treatment in RPB is the stable gas–liquid mass transfer process. A radius of $r$, thickness of d$r$, and packing layer axial height of $h$ is taken as an element of volume, which is shown in Fig. 2. Material balances for the element of volume are calculated as follows.

According to the material balances, the loss of solute in liquid is equal to the gain of solute in gas and equal to the solute transferred from the liquid phase to the gas phase, that is:

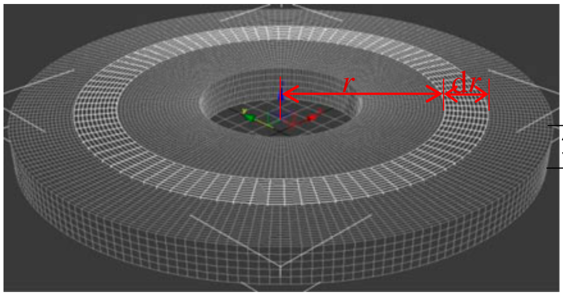

Fig. 2. Material balances calculation for packing layer respectively, mol mol$^{-1}$.

$$-L\mathrm{d}X = G\mathrm{d}Y = N_{\mathrm{AN}} \cdot a \cdot 2\pi r \cdot h \cdot \mathrm{d}r \tag{1}$$

where $G$, $L$—the molar flow of gas and liquid, respectively, mol s$^{-1}$; $Y$, $X$—the molar ratio of gas and the liquid phase, respectively, mol mol$^{-1}$; $h$—packing layer axial height, m; $a$—effective mass transfer specific surface area, m$^2$ m$^{-3}$; $N_{\mathrm{AN}}$—the mass transfer rate of acrylonitrile, mol m$^{-2}$ s$^{-1}$.

$N_{\mathrm{AN}}$ can be calculated as follows:

$$N_{\mathrm{AN}} = K_{\mathrm{X}}(X - X^*) \tag{2}$$

where $K_{\mathrm{X}}$—liquid overall mass transfer coefficient, mol m$^{-2}$ s$^{-1}$; $X^*$—the equilibrium molar ratio of acrylonitrile in the liquid phase, mol mol$^{-1}$; $X$—the molar ratio of acrylonitrile in the liquid phase, mol mol$^{-1}$.

If the two-film theory is used to describe it, this stripping process is controlled by gas film and liquid film commonly. Eq. (2) also can be expressed as:

$$N_{\mathrm{AN}} = k_{\mathrm{X}}(X_{\mathrm{i}} - X) = k_{\mathrm{Y}}(Y - Y_{\mathrm{i}}) \tag{3}$$

where, $k_{\mathrm{X}}$, $k_{\mathrm{Y}}$—liquid and gas mass transfer coefficient, respectively, mol m$^{-2}$ s$^{-1}$; $X$, $X_{\mathrm{i}}$—the molar ratio of acrylonitrile in the liquid phase and gas–liquid interphase, respectively, mol mol$^{-1}$; $Y$, $Y_{\mathrm{i}}$—the molar ratio of acrylonitrile in the gas phase and gas–liquid interphase respectively, mol mol$^{-1}$.

Then, the liquid overall mass transfer coefficient is given by the following expression

$$\frac{1}{K_{\mathrm{X}}} = \frac{1}{mk_{\mathrm{Y}}} + \frac{1}{k_{\mathrm{X}}} \tag{4}$$

From Eq. (4), $K_{\mathrm{X}}$ was affected by $k_{\mathrm{X}}$, $k_{\mathrm{Y}}$, and $m$ mainly. Their measurements are more complicated and difficult, although they can be used to analyze the mass transfer process. Therefore, liquid overall mass transfer coefficient is mainly determined in this experiment.

Eq. (2) is put into Eq. (1) to make out Eq. (5).

$$-L\mathrm{d}X = K_{\mathrm{X}}(X - X^{*}) \cdot a \cdot 2\pi r \cdot h \cdot \mathrm{d}r \quad (5)$$

Eq. (5) is solved using a definite integral. Boundary conditions are

(1) $r = r_1$, $X = X_1$;
(2) $r = r_2$, $X = X_2$;

Consequently, Eq. (6) is developed as follows:

$$2\pi h \int_{r_1}^{r_2} r\mathrm{d}r = \int_{X_1}^{X_2} \frac{-L}{K_{\mathrm{X}}a} \cdot \frac{\mathrm{d}X}{(X - X^{*})} \quad (6)$$

Because the concentration of acrylonitrile in the gas and liquid phase, mass transfer surface area, and flow velocity varies radially, the average liquid overall mass transfer coefficient is used.

$$K_{\mathrm{X}}a = \frac{L}{\pi h \cdot (r_2^2 - r_1^2)} \int_{X_2}^{X_1} \frac{\mathrm{d}X}{(X - X^{*})} \quad (7)$$

The molar ratio of acrylonitrile in the liquid phase is generally low ($X_1 < 1\%$), and therefore $X$ is equal to $x$ approximately. The equilibrium molar ratio of acrylonitrile in the liquid phase $X^{*}$ tends to 0, and $X - X^{*}$ is equal to $X$ approximately. Thus, the average liquid overall mass transfer coefficient is

$$K_{\mathrm{x}}a = \frac{L}{\pi h \cdot (r_2^2 - r_1^2)} \ln \frac{x_1}{x_2} \quad (8)$$

### 2.3.3. Determination of removal rate of acrylonitrile

The removal rate of acrylonitrile is expressed as Eq. (9):

$$\eta = \frac{C_{\mathrm{in}} - C_{\mathrm{out}}}{C_{\mathrm{in}}} \times 100\% \quad (9)$$

where $C_{\mathrm{in}}$ and $C_{\mathrm{out}}$ represent the mass concentration of acrylonitrile in liquid stream entering and leaving the RPB, respectively, mg $\mathrm{L}^{-1}$.

## 3. Results and discussion

### 3.1. High gravity factor $\beta$

Mass transfer in RPB is affected by speed significantly, that is by the strength of high gravity field. In order to characterize the strength, the high gravity factor $\beta$ is proposed, and it is defined as the ratio of the centrifugal acceleration and gravitational acceleration. The definition formula is

$$\beta = \frac{\omega^2 r}{g} \quad (10)$$

$\omega$ is the angular speed of the RPB, $r$ is the geometric average radius of the packing, and $g$ is the acceleration of gravity (9.8 m $\mathrm{s}^{-2}$).

In Fig. 3, the liquid overall mass transfer coefficient and acrylonitrile removal rate obviously increase with an increase in high gravity factor $\beta$ ranging from 35 to 50. However, when the high gravity factor $\beta$ is further increased, only a small effect on the $K_{\mathrm{x}}a$ and $\eta$ is observed. The possible reasons include three aspects. First, in RPB, with an increase in high gravity factor $\beta$, the centrifugal force is strengthened, so that the wastewater in packing layer is sheared in the smaller droplets, liquid filament, and liquid film [24]. The mass transfer surface area is increased continually and mass transfer distance from the liquid phase to gas phase is reduced. Second, the gas–liquid relative velocity and turbulent increase with increase in high gravity factor $\beta$. Third, the renewal rate and times of mass transfer surface are increased. Therefore, the mass transfer coefficient increases. Meanwhile, the removal rate of acrylonitrile is improved. When high gravity factor $\beta$ is higher, wastewater dispersion is difficult to further increase. The effective mass transfer surface area has been restricted by the dispersion of the liquid. Retention time is reduced under a higher $\beta$, which is unfavorable to stripping process. So, the mass transfer coefficient and removal rate of acrylonitrile increase slowly. Consequently, in terms of economic benefits, the high gravity factor $\beta$ of 50–55 is optimal for the RPB in this study.

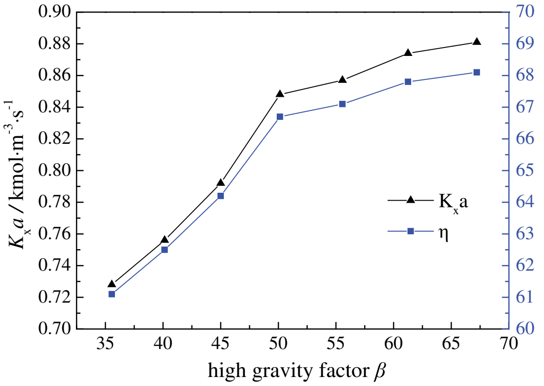

Fig. 3. Effect of high gravity factor $\beta$ on $K_{\mathrm{x}}a$ and $\eta$.

### 3.2. Gas–liquid ratio R

Gas–liquid ratio is served as a major concern in the design and investment of RPB, blower, pump, and pipeline. Researchers have demonstrated that gas–liquid ratio will directly influence the operation and cost. Meanwhile, it is believed to be a powerful way to adjust the stripping performance.

Figs. 4 and 5 show the effect of gas–liquid ratio $R$ on $K_xa$ and $\eta$ at different high gravity factor $\beta$. The gas–liquid ratio $R$ is controlled by changing the air flow rate at a fixed wastewater flow rate. As a whole, the liquid overall mass transfer coefficient and removal rate of acrylonitrile increase with the rise of gas–liquid ratio $R$. The increase in the gas–liquid ratio $R$ means more gas per unit of wastewater. The increase in gas flow rate results in increased not only gas–liquid contact area, but also mass transfer force, which is beneficial for the mass transfer. Thus, the liquid overall mass transfer coefficient and removal rate of acrylonitrile increase. Furthermore, the increase in high gravity factor $\beta$ from 35 to 65 enhances the liquid overall mass transfer coefficient, as well as increases the removal rate of acrylonitrile. The positive effect of increasing high gravity factor $\beta$ is related to a large shear force, which is favorable to air stripping process. So, the gas–liquid ratio $R$ should be kept around 1,300 $m^3\ m^{-3}$ in this high gravity air stripping technology to ensure good mass transfer performance and an optimal removal rate.

### 3.3. Liquid spray density U

In RPB, liquid spray density is an important design parameter for selecting liquid flow, which is defined as:

$$U = \frac{L}{2\pi r_m h} \qquad (11)$$

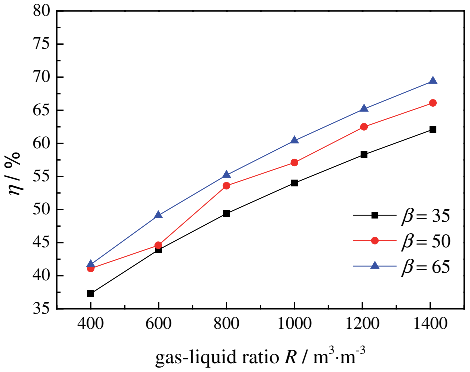

Fig. 5. Effect of gas–liquid ratio $R$ on $\eta$.

$L$ is the liquid flow, $r$ is the geometric average radius of the packing layer, and $h$ is the height of packing layer.

Figs. 6 and 7 display the effect of liquid spray density $U$ on the liquid overall mass transfer coefficient and removal rate of acrylonitrile at different high gravity factor $\beta$. The liquid overall mass transfer coefficient increases with increase in liquid spray density $U$. This will lead to the formation of thin liquid boundary layer. Increase in liquid flow rate will lead to the formation of thin liquid boundary layer and the corresponding reduction of mass transfer resistance, thereby benefiting the increase of liquid side mass transfer. On the one hand, the turbulence of liquid and the renewal rate of liquid film increase as the liquid spray density $U$ increases. On the other hand, the packing is wetted insufficiently at a smaller liquid spray density $U$. At this time, the packing can be wetted fully when the liquid spray density $U$ increases. However, after the packing wetted fully, the liquid film becomes thicker as liquid spray density $U$ increases, which enlarge liquid phase mass transfer resistance. As expected, high gravity factor $\beta$ also

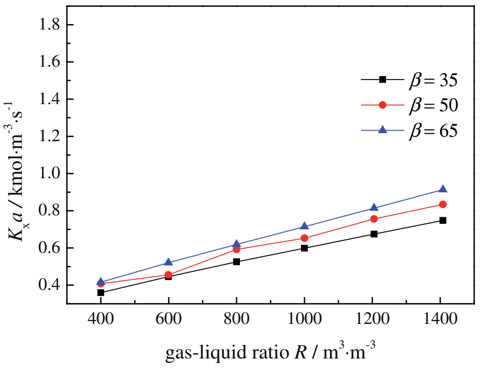

Fig. 4. Effect of gas–liquid ratio $R$ on $K_xa$.

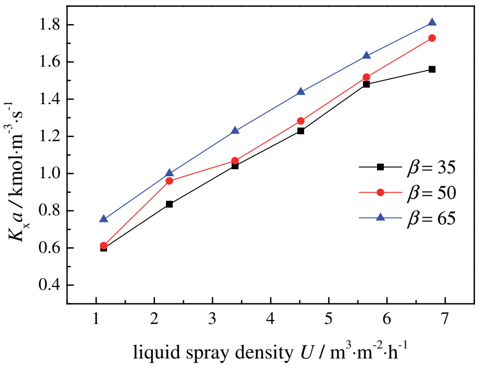

Fig. 6. Effect of liquid spray density $U$ on $K_xa$.

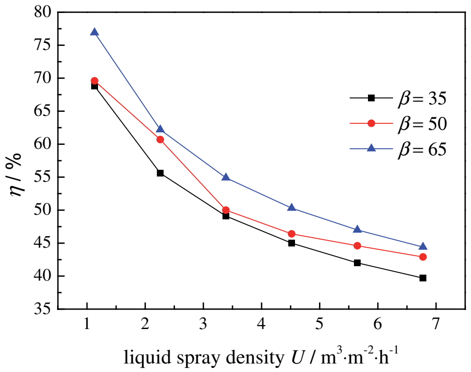

Fig. 7. Effect of liquid spray density $U$ on $\eta$.

plays an important role on mass transfer process as $K_xa$ increases as $\beta$ is increased. An increase in high gravity factor $\beta$ will lead to a smaller droplets [25], resulting in the complete contact of air and wastewater. So, the optimum liquid spray density $U$ is in the range of 2–3 m$^3$ m$^{-2}$ h$^{-1}$ for this high gravity air stripping technology.

### 3.4. Initial concentration of acrylonitrile $C_{in}$

Figs. 8 and 9 present the effect of initial concentration $C_{in}$ on $K_xa$ and $\eta$ at different high gravity factor $\beta$. $K_xa$ and $\eta$ reach the highest value at a initial concentration $C_{in}$ of 3,000 ± 100 mg L$^{-1}$. The high or low initial concentrations are not favorable to the mass transfer process and removal of acrylonitrile. Under the initial concentration $C_{in}$ of 3,000 mg L$^{-1}$, the liquid phase mass transfer force increases with increase in initial concentration of acrylonitrile. But the acrylonitrile concentration in the gas phase is close to equilibrium when the initial concentration continues to increase. The change of acrylonitrile concentration in wastewater is small, so the liquid overall mass transfer coefficient and removal rate of acrylonitrile decrease. Therefore, within the experimental range, the initial concentration of 3,000 ± 100 mg L$^{-1}$ is suitable.

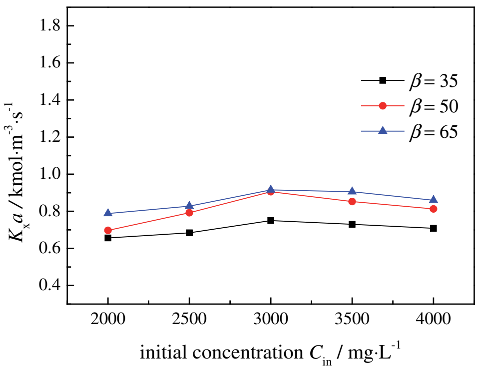

Fig. 8. Effect of initial concentration $C_{in}$ on $K_xa$.

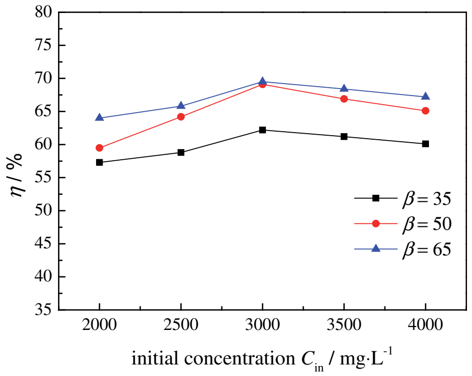

Fig. 9. Effect of initial concentration $C_{in}$ on $\eta$.

### 3.5. Correlation of liquid overall mass transfer coefficient $K_xa$ and removal rate of acrylonitrile $\eta$

Seen from the above experimental studies, the effect of operating parameters on $K_xa$ and $\eta$ is more complex. Based on initial concentration of acrylonitrile and some easily adjustable operating parameters on industrial application such as high gravity factor $\beta$, gas–liquid ratio, and liquid spray density $U$, $K_xa$, and $\eta$ correlations are correlated as

$$K_xa = A \cdot \beta^B \cdot R^C \cdot U^D \cdot C_{in}^E \tag{12}$$

$$\eta = A \cdot \beta^B \cdot R^C \cdot U^D \cdot C_{in}^E \tag{13}$$

where, $A$, $B$, $C$, $D$, and $E$ are undetermined coefficient. $A$ includes RPB structure, acrylonitrile wastewater characteristics, and other factors. During the experimental range, $K_xa$ and $\eta$ are calculated by software. The relative errors of $K_xa$ and $\eta$ are given in Table 3. The values and standard errors of fitting coefficients are given in Table 4.

The comparisons of $K_xa$ and $\eta$ between fitting and experimental results are shown in Figs. 10 and 11, respectively. Experimental results are used as abscissa, and fitting results are used as vertical coordinate. The points represent the corresponding fitting results under the operational conditions of experimental results. Figs. 10 and 11 show that the correlation $K_xa$ and $\eta$ are available. The average relative errors are 3.689 and 2.25%, respectively. The $R^2$ are 0.98 and 0.96, respectively. Apparently, the correlations are reasonable and could be used to predict the mass

Table 3
Relative errors

| Items | Average relative error (%) | Maximum relative error (%) | Minimum relative error (%) |
|---|---|---|---|
| $K_xa$ | 3.69 | 12.47 | 0.0090 |
| $\eta$ | 2.25 | 6.73 | 0.14 |

Table 4
Fitting coefficients

| Items | | $K_xa$ | $\eta$ |
|---|---|---|---|
| $A$ | Value | $2.8295 \times 10^{-4}$ | 0.6403 |
| | Standard error | $3.0406 \times 10^{-4}$ | 0.3473 |
| $B$ | Value | 0.3201 | 0.1800 |
| | Standard error | 0.1027 | 0.0539 |
| $C$ | Value | 0.6301 | 0.4050 |
| | Standard error | 0.0673 | 0.0295 |
| $D$ | Value | 1.1551 | 0.0986 |
| | Standard error | 0.0673 | 0.0341 |
| $E$ | Value | 0.1946 | 0.1182 |
| | Standard error | 0.1059 | 0.0553 |
| $R^2$ | | 0.98 | 0.96 |

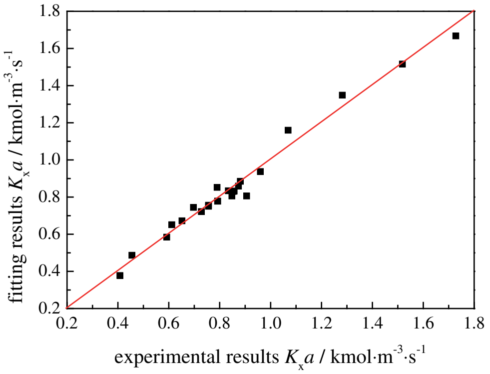

Fig. 10. Comparison of $K_xa$ between fitting and experimental results.

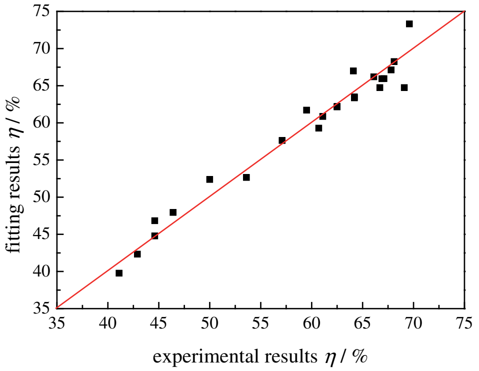

Fig. 11. Comparison of $\eta$ between fitting and experimental results.

transfer performance and removal rate of RPB with experimental data.

### 3.6. Comparison with conventional technology

The comparison of high gravity technology and conventional technology are presented in Table 3. For better mass transfer performance and higher removal rate, the stripping column is 700 times the height of the RPB. $K_xa$ in the RPB is eight times over that in the stripping column. The pressure drop in the RPB is one-fourteenth lower than that in the stripping column. As can be seen in Table 5, the treatment of acrylonitrile wastewater by the high gravity technology has unique advantages over the conventional technology.

Table 5
Technologies comparison

| Items | High gravity technology | Conventional technology |
|---|---|---|
| Reactor type | Rotating packed bed | Stripping column |
| Packing layer | ID 0.03 m × OD 0.064 m × H 0.03 m | D 1.07 m × H 21.20 m |
| High gravity factor $\beta$ | 55 | 0 |
| Gas–liquid ratio $R$ (m$^3$ m$^{-3}$) | 1,300 | 144 |
| Liquid spray density (m$^3$ m$^{-2}$ h$^{-1}$) | 2–3 | 254 |
| Initial concentration $C_{in}$ (mg L$^{-1}$) | 3,000 | 500 |
| Operating temperature (℃) | 20–25 | 25 |
| Wastewater flow rate (L h$^{-1}$) | 20–30 | 227,124 |
| $\eta$ (%) | 69.1 | 99.8 |
| $K_xa$ (kmol m$^{-3}$ s$^{-1}$) | 0.906 | 0.114 |
| Pressure drop (Pa) | 3,340 | 45,061 |

## 4. Conclusions

In this study, RPB is successfully employed in high gravity air stripping process for the treatment of acrylonitrile wastewater. Increase in the high gravity factor $\beta$, the gas–liquid ratio $R$, and liquid spray density $U$ are beneficial to the enhancement of mass transfer process. The high or low initial concentrations are not favorable to mass transfer process, resulting in a low acrylonitrile removal rate. The high gravity factor $\beta$ of 50–55, gas–liquid ratio $R$ of around 1,300 $m^3\,m^{-3}$, and liquid spray density $U$ of 2–3 $m^3\,m^{-2}\,h^{-1}$ is suitable for the initial concentration of $3{,}000 \pm 100\,mg\,L^{-1}$ of acrylonitrile wastewater. Under the following conditions, the liquid overall mass transfer coefficient and acrylonitrile removal rate could reach $0.906\,kmol\,m^{-3}\,s^{-1}$ and 69.1%, respectively. The average relative errors of $K_xa$ and $\eta$ correlations are 3.689 and 2.25%, respectively, which indicate that fitting results fit well with experimental results. Compared with conventional technology, high gravity air stripping technology can significantly reduce wastewater treatment costs when the removal rate of acrylonitrile remains higher. Therefore, it could be envisioned that RPB exhibits a great potential for the industrial application of acrylonitrile wastewater treatment via high gravity air stripping technology.

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