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Absorption of lean formaldehyde from air with Na_2SO_3 solution

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Abstract

Formaldehyde is a major indoor pollutant over the world and its high release over the national standards in developing countries, such as China, harms people's health seriously. In this work, an investigation is carried out in a stirred vessel with a plain gas–liquid interface for the absorption of lean formaldehyde from pollution air by Na_2SO_3 solution at 25 °C. Experiments are conducted with Na_2SO_3 concentrations of 0.01–0.30 kmol m^{-3}. The results show that the Na_2SO_3 solution is an effective absorbent to remove lean formaldehyde from polluted air. The experiments reveal that the absorption rate is considerably influenced by the gas flow rate and formaldehyde concentration. The Na_2SO_3 concentration has little effect on the absorption rate, and the loading content of formaldehyde reduces the absorption rate only if the formaldehyde concentration approaches or exceeds the concentration of Na_2SO_3 solution. A theoretical model is developed and used to successfully calculate the absorption rate with the overall relative deviation of less than 15% to the experimental data. A possible process of the absorption method is also proposed in this paper. The analysis shows that the proposed process is feasible in removing formaldehyde from indoor air.

Keywords: Formaldehyde; Indoor air pollution; Absorption rate; Sodium sulfite

1. Introduction

In recent years, volatile organic chemicals (VOCs) have become major indoor air pollutants. Formaldehyde is one of the most serious pollutants among VOCs over the world, especially in newly decorated rooms. It comes from building materials, decorated materials, cigarette smoke, etc. Formaldehyde harms people's health seriously [1,2]. It is pungent to the mucosa of the eye, nose and respiratory tract and acts as a lachrymator to cause sneezing and coughing even at a very low concentration. National Institute for Occupational Safety and Health (NIOSH) has announced that formaldehyde has the possibility of inducing cancers in human being.

The concentration of formaldehyde is commonly observed at part-per-billion (ppb, 10^{-9}) level in air. However, in some developing countries, such as China, the concentration in some newly decorated buildings is far beyond this level and can even reach several part-per-million (ppm, 10^{-6}) [3], due to the use of cheap decorated materials. The formaldehyde concentration could also reach high level (ppm level) at some special occasions such as hospitals and some chemical engineering factories or workshops, both in developing and developed countries.

Some investigators have been focusing on the research of removing formaldehyde from indoor air. Different researchers have indicated that some catalysts [4–6], solid absorbents and adsorbents have effective functions on formaldehyde removal. For instance, metal oxides [7], ammonium salts [8] and Fe–phosphate–ammonium oxalate composites [9] can be good cleaning materials of formaldehyde. However, instability of these materials in removal efficiency and other disadvantages such as low mass transfer flux and low operation life often limit their applications. Ozone [10] as a gas absorbent is also used to remove formaldehyde from air, but itself is harmful to people's health. Nano-TiO_2 shows favorable efficiency on formaldehyde removal under the condition of UV irradiation [11–13], but the low rate of this reaction is not favorable to its indoor use.

The absorption method has been widely used to remove polluted gases since it offers considerable advantages, including high absorption efficiency, large processing capacity, capable of removing almost all kinds of pollutants with certain solutions and so on. However, there are few research efforts on

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Nomenclature

a	specific area of the packing ($m^2\ m^{-3}$)
C_N	concentration of Na_2SO_3 in liquid phase ($kmol\ m^{-3}$)
D	diffusion coefficient ($m^2\ s^{-1}$)
E	chemical reaction enhancement factor
G	gas flow rate of packed column ($kmol\ m^{-2}\ s^{-1}$)
h	height of the packing (m)
k	rate constant of pseudo-first-order reaction (s^{-1})
k_1	rate constant of reaction (1) (s^{-1})
k_2	rate constant of reaction (2) ($kmol^{-1}\ m^3\ s^{-1}$)
k_g	gas-side mass transfer coefficient ($kmol\ m^{-2}\ s^{-1}\ kPa^{-1}$)
k_l	liquid-side mass transfer coefficient ($m\ s^{-1}$)
K	mass transfer coefficient ($kmol\ m^{-2}\ s^{-1}\ kPa^{-1}$)
N_a	absorption rate ($kmol\ m^{-2}\ s^{-1}$)
p	formaldehyde partial pressure (kPa)
p_1	inlet formaldehyde partial pressure of packed column (kPa)
p_2	outlet formaldehyde partial pressure of packed column (kPa)
P	operation pressure of packed column (kPa)
T	temperature (°C)

Subscripts

i	gas–liquid interface
N	Na_2SO_3

such a method to remove formaldehyde from indoor polluted air. Sodium sulfite is believed to react with formaldehyde to form sodium formaldehyde bisulfite [14], which is a non-toxic and biodegradable substance. This work will investigate the removal of lean formaldehyde from indoor air by using Na_2SO_3 solutions.

2. Experimental apparatus and methods

The experiments are carried out in a stirred glass vessel that has a plain gas–liquid interface. The experimental setup is shown in Fig. 1. The vessel has an inner diameter of 80 mm. Double deck stirrers are used in the vessel to agitate the gas and liquid phase, respectively. The stirrers are fan turbines with four blades and placed at the center of each phase. A circulating water jacket is equipped outside the vessel to maintain its temperature at 25 °C.

Formaldehyde vapor is produced by passing nitrogen from a cylinder through a formalin solution, and then diluted by the air from another cylinder. The mixed gas stream is modeled as the formaldehyde polluted air and feeds into the reaction vessel. The inlet or outlet gas stream is collected using a gas collection system and analyzed by the acetylacetone spectrophotometric method [15]. The method mainly consists of following steps: the formaldehyde pollutant gas collected by the gas collection system is absorbed into 20 ml de-ionized water; several milliliters

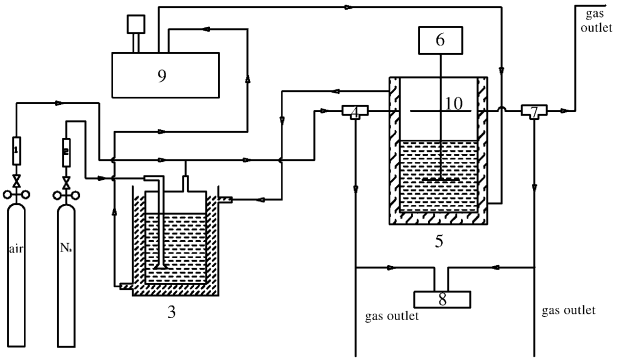

Fig. 1. Schematic diagram of the experimental setup. (1, 2) Flowmeter; (3) formaldehyde producing system; (4, 7) triple valve; (5) stirring vessel; (6) constant speed stirrer; (8) gas collection system; (9) thermostatic bath; (10) double-deck stirrers.

of the solution from the first step are taken out and diluted using de-ionized water to 10 ml, followed by the immediate addition of 2 ml 0.25 wt% acetylacetone solution; the mixed solution from the second step is heated for 3 min in the boiling water bath to develop brilliant yellow color; the spectrophotometry is used at 413 nm to measure the formaldehyde concentration of the colored solution from the calibration curve of standard solutions.

The formaldehyde concentration of the gas could be calculated from that of the colored solution by taking into account the volume of the gas collection system and the dilution ratio. Duplicate determination is done to obtain the average concentration value for each run of the experiment. The accuracy of the spectrophotometric method is found to be within 2%.

The gas volume in the vessel is 590 ml, and the liquid absorbents are 500 ml Na_2SO_3 solutions of different concentrations. The Na_2SO_3 and formaldehyde concentrations used in the experiments are in a range of 0.01–0.30 $kmol\ m^{-3}$ and 7–60 ppm, respectively. The gas flow rate is set at $830 \pm 25\ ml\ min^{-1}$ except those values specially indicated. The stirring speed is kept at 140 rpm for all the experiments.

In order to explore how the stirring speed affects the liquid-side mass transfer, the physical absorption of pure carbon dioxide into water is carried out in the vessel at 25 °C. The liquid-side mass transfer coefficient is obtained from the mass transfer area and the volumes difference of pure CO_2 between the inlet and outlet. The volumes are measured using soap film meters. The gas-side mass transfer coefficient is acquired by performing the chemical absorption experiments of lean carbon dioxide (about 1%) air into various concentrations of NaOH solutions (0.146–0.975 $kmol\ m^{-3}$). The absorption rates are determined from the mass transfer area, the gas flow rate and the concentration difference of CO_2 between the inlet and outlet gases. The concentration of CO_2 is directly read from a Portable Infrared Rays CO_2 Analyzer (Beijing Computer Technology Application Institute, China). The results as shown in Figs. 2 and 3 correlate the gas- and liquid-side mass transfer coefficient with the stirring velocity, n, respectively. These correlations obtained from CO_2 absorption experiments will be used to estimate the mass transfer coefficients versus the stirring speed in the formaldehyde absorption using Na_2SO_3 solutions.

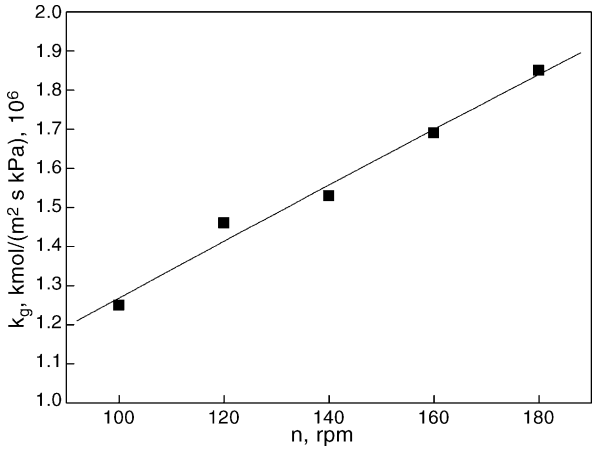

Fig. 2. Gas-side mass transfer coefficient vs. stirring speed.

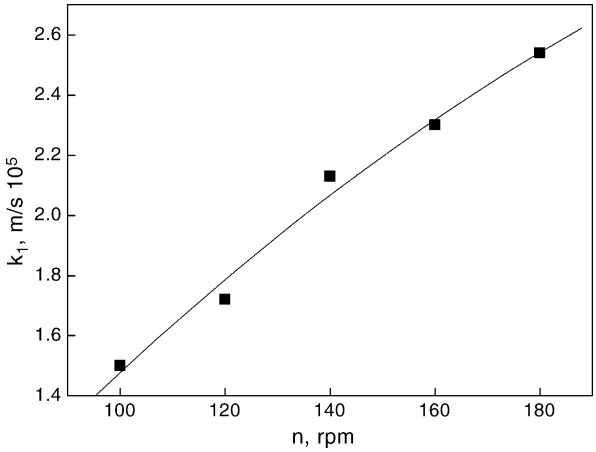

Fig. 3. Liquid-side mass transfer coefficient vs. stirring speed.

3. Theoretical model

3.1. Chemical reactions between formaldehyde and Na_2SO_3 solutions

When formaldehyde is absorbed into a Na_2SO_3 solution and the hydrolyzation of $[SO_3^{2-}]$ is neglected, the following reactions may occur:

$$CH_2O + H_2O \overset{k_1}{\rightleftharpoons} CH_2(OH)_2 \quad (1)$$

$$SO_3^{2-} + CH_2O \overset{k_2}{\rightleftharpoons} CH_2(O^-)SO_3^- \quad (2)$$

where $k_1 = 7800\exp(-1913/T)\,s^{-1}$ [16] and $k_2 = 9.46 \times 10^6\,kmol^{-1}\,m^3\,s^{-1}$ [17] at 25 °C. For the absorption of lean formaldehyde from the polluted air using the Na_2SO_3 solution of 0.01–0.3 kmol m^{-3}, $k_2[SO_3^{2-}] \gg k_1$, reaction (1) can be neglected when comparing with reaction (2).

$CH_2(O^-)SO_3^-$ in reaction (2) is not the final product, it will continue to react with water,

$$CH_2(O^-)SO_3^- + H_2O \overset{\text{fast}}{\rightleftharpoons} CH_2(OH)SO_3^- + OH^- \quad (3)$$

reaction (2) is the rate-determining step when compared with reaction (3) [17]. Since the formaldehyde concentration in this study is several ppm and the gas flow rate is set at about 830 ml min^{-1}, the concentration of reacted $[SO_3^{2-}]$ is not comparable to the concentration of initially added Na_2SO_3 which is in the range of 0.01–0.3 kmol m^{-3}. Therefore, the change of $[SO_3^{2-}]$ in the solution could be ignored.

Then the overall reaction rate of formaldehyde absorption into the solution can be described as:

$$r = k[CH_2O] \quad (4)$$

where $k = k_2[SO_3^{2-}]$, and the whole process can be regarded as a pseudo-first-order reaction.

3.2. Model derivation

Before deducing the theoretical model, it is necessary to obtain the gas- and liquid-side mass transfer coefficients of formaldehyde in the reaction system. These coefficients can be calculated using the following correlations [18],

$$k_{g,CH_2O\text{–air}} = k_{g,CO_2\text{–air}}\left(\frac{D_{CH_2O\text{–air}}}{D_{CO_2\text{–air}}}\right)^{2/3} \quad (5)$$

$$k_{l,CH_2O\text{–}H_2O} = k_{l,CO_2\text{–}H_2O}\left(\frac{D_{CH_2O\text{–}H_2O}}{D_{CO_2\text{–}H_2O}}\right)^{2/3} \quad (6)$$

From Figs. 2 and 3, $k_{g,CO_2\text{–air}}$ and $k_{l,CO_2\text{–}H_2O}$ can be obtained as 1.53×10^{-6} kmol m^{-2} s^{-1} kPa^{-1} and 2.13×10^{-5} m s^{-1} at the stirring speed of 140 rpm, respectively. $D_{CO_2\text{–air}}$ and $D_{CO_2\text{–}H_2O}$ are obtained from literature [19], while $D_{CH_2O\text{–air}}$ and $D_{CH_2O\text{–}H_2O}$ are calculated from the Fuller empirical formula [20] and another empirical equation [19]. Since it is indicated that the ionic strength has little effect on the diffusion coefficient of formaldehyde [21], the diffusion coefficients in this study can be considered as constants. Then, the mass transfer coefficients in the reaction system can be determined with the help of Eqs. (5) and (6), t. Table 1 shows the values of these coefficients, together with the parameter used to calculate them.

The model derivation starts from the mass transfer rate of formaldehyde into a reaction solution, which can be written as:

$$N_a = k_g(p - p_i) \quad (7)$$

or

$$N_a = Ek_l(c_i - c) \quad (8)$$

Table 1
Mass transfer coefficients and diffusion coefficients used in the study at 25 °C

$D_{CO_2\text{–air}}$ (m^2 s^{-1})	1.57×10^{-5}
$D_{CH_2O\text{–air}}$ (m^2 s^{-1})	1.72×10^{-5}
$D_{CO_2\text{–}H_2O}$ (m^2 s^{-1})	1.53×10^{-9}
$D_{CH_2O\text{–}H_2O}$ (m^2 s^{-1})	2.34×10^{-9}
$k_{g,CO_2\text{–air}}$ (kmol m^{-2} s^{-1} kPa^{-1})	1.63×10^{-6}
$k_{g,CH_2O\text{–air}}$ (kmol m^{-2} s^{-1} kPa^{-1})	1.53×10^{-6}
$k_{l,CO_2\text{–}H_2O}$ (m s^{-1})	2.13×10^{-5}
$k_{l,CH_2O\text{–}H_2O}$ (m s^{-1})	2.82×10^{-5}

where N_a is the mass transfer rate, E the chemical reaction enhancement factor and the subscript i means the gas–liquid interface. At low formaldehyde concentration, the gas–liquid equilibrium equation for formaldehyde could be expressed as [20]:

$$[\mathrm{HCHO(aq)}] = 10^X[\mathrm{HCHO(g)}]^Y) \quad (9)$$

where $X = [(4538/T) - 11.34]$, $Y = [(252.2/T) + 0.2088]$. Combining Eq. (9) with Eq. (8), leads to Eq. (10):

$$N_a = Ek_l 55.56 \times 10^X \left[\left(\frac{p_i}{p_0}\right)^Y - \left(\frac{p^*}{p_0}\right)^Y\right] \quad (10)$$

As stated above, the equilibrium constant of reaction (2) is so large that the concentration of free formaldehyde in the liquid side is negligibly small. Thus, Eq. (10) can be simplified as:

$$N_a{}^{1/Y}[(55.56Ek_l 10^X)^{-1/Y}]p_0 = p_i \quad (11)$$

Inserting Eq. (7) into Eq. (11), the following equation can be derived,

$$N_a k_g^{-1} + N_a{}^{1/Y}[(55.56Ek_l 10^X)^{-1/Y}]p_0 = p \quad (12)$$

Since the reaction between formaldehyde and Na_2SO_3 can be regarded as pseudo-first-order reaction, the chemical reaction enhancement factor, based on the film-theory [22], can be written as:

$$E = \frac{k_l'}{k_l} = \frac{(kD_{CH_2O-H_2O}/k_l^2)^{1/2}}{\tanh[(kD_{CH_2O-H_2O}/k_l^2)^{1/2}]} = \frac{r}{\tanh(r)} \quad (13)$$

where $k = k_2 C_N$, C_N is the concentration of Na_2SO_3, and r is the combined parameter which is defined as:

$$r = \left(\frac{kD_{CH_2O-H_2O}}{k_l^2}\right)^{1/2} \quad (14)$$

Note that the lowest concentration of Na_2SO_3 used in this experiment is 0.01 kmol m^{-3}, the smallest r-value calculated from Eq. (14) is 528. Since the value of r is much greater than 3, the enhancement factor, E, is approximately equal to r. Therefore, the absorption equation can be established as

$$N_a k_g^{-1} + N_a{}^{1/Y}[55.56(kD_{CH_2O-H_2O})^{1/2}10^X]^{-1/Y} p_0 = p \quad (15)$$

At the temperature of 25 °C, the parameters of X, Y and k_g, k_2, $D_{CH_2O-H_2O}$ as shown in Table 1 is substituted into Eq. (15), resulting in:

$$N_a(1.63 \times 10^{-6})^{-1} + N_a{}^{0.94}(324.39C_N^{0.47})^{-1} = p \quad (16)$$

The left hand side of Eq. (16) consists of two terms: the liquid phase and gas phase resistance contributions. It is found that the gas phase resistance is several orders of magnitude greater than the liquid phase resistance. Therefore, when C_N is in the range of 0.01–0.30 kmol m^{-3}, Eq. (16) can be further simplified by neglecting the liquid phase resistance contribution as

$$N_a = 1.63 \times 10^{-6} p \quad (17)$$

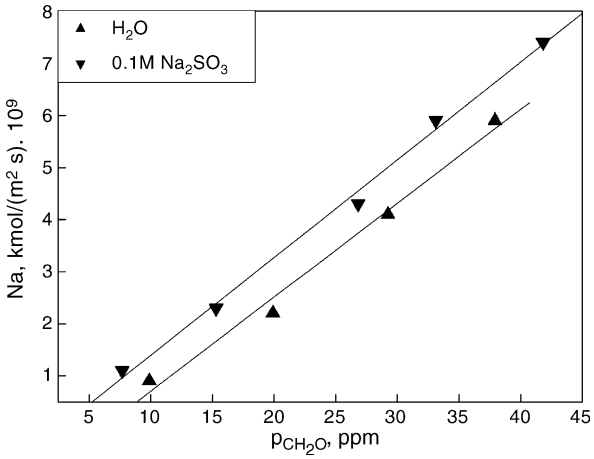

Fig. 4. Comparison the absorption rate of 0.1 M Na_2SO_3 with water at the gas flow rate of 830 ± 25 ml min^{-1}.

Eq. (17) is the final simplified model expression that can be used to predict the mass transfer rate of absorption of lean formaldehyde from air into Na_2SO_3 solutions.

4. Results and discussion

The experiments were performed in the glass vessel with Na_2SO_3 concentration of 0.01–0.30 kmol m^{-3} and formaldehyde concentration of 7–60 ppm at 25 °C, to explore the influencing factors of the absorption process.

Fig. 4 presents the relationship between the absorption rate and the concentration of formaldehyde. Na_2SO_3 holds a higher absorption rate than water does. Therefore, Na_2SO_3 solution is regarded as effective absorbent for the removal of formaldehyde from polluted air.

Fig. 5 shows that the concentration of Na_2SO_3 solution has little effect on the absorption rate. The increase of Na_2SO_3 concentration is believed to increase the reaction rate between formaldehyde and Na_2SO_3, which leads to improve the chemical reaction enhancement factor of this process. However, with the increase of the chemical reaction enhancement factor, the

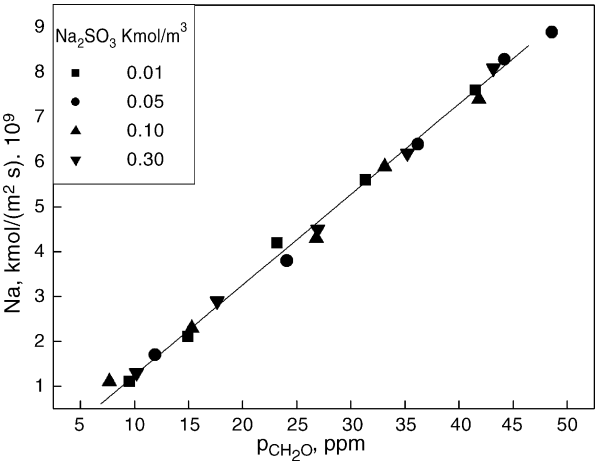

Fig. 5. Effect of the Na_2SO_3 concentration on the absorption rate at the gas flow rate of 830 ± 25 ml min^{-1}.

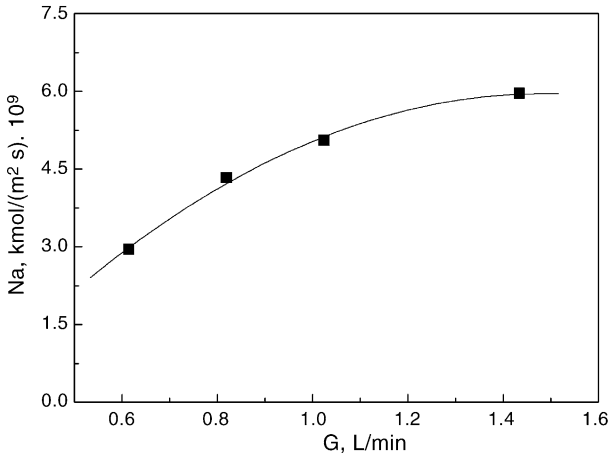

Fig. 6. Effect of gas flow rate on the absorption rate at the formaldehyde concentration of 60 ± 2 ppm and Na_2SO_3 of 0.1 kmol m^{-3}.

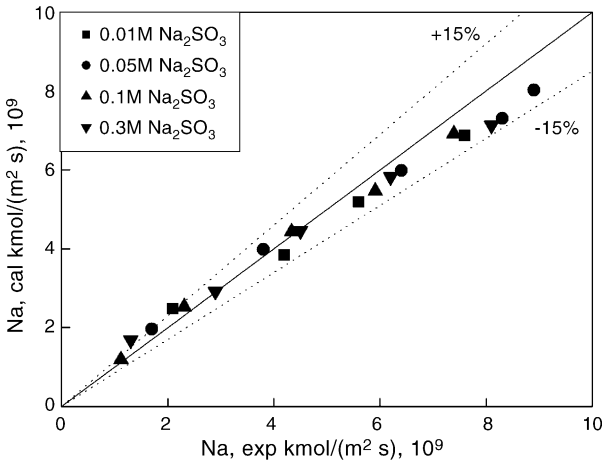

Fig. 8. Comparison of experimental data with calculated values.

absorption rate changes very little. This suggests that the absorption is gas-side controlled process at Na_2SO_3 concentration of 0.01–0.30 kmol m^{-3}. Fig. 5 also indicates that the absorption rates at different Na_2SO_3 concentrations increase linearly with the increase of formaldehyde concentration in the gas phase. These experimental data can be predicted well with the theoretical model, as shown by the line in the figure.

Fig. 6 shows that the absorption rate increases with the increasing of gas flow rate at the Na_2SO_3 solution concentration of 0.1 kmol m^{-3}. This is mainly attributed into two parts: on one hand, the residence time of gas in the vessel will be reduced with the increasing of gas flow rate, and the formaldehyde concentrations in the gas phase will be increased under the same inlet gas concentration. As a result, the absorption rate will be enhanced. On the other hand, the mass transfer rate of gas is depended on the thickness of the boundary layer. With the increase of the air velocity in the gas phase, the boundary layer tends to be thin, which will also benefit to the absorption rate. Therefore, the increase of gas flow rate will lead to the increase of absorption rate.

Fig. 7 is the experimental data of absorption rate with respect to the loading concentration of formaldehyde in the solution.

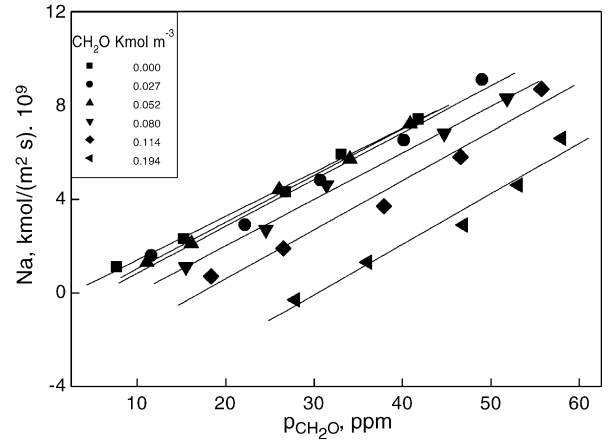

Fig. 7. Effect of the loading of formaldehyde in the solution on the absorption rate at Na_2SO_3 concentration of 0.1 kmol m^{-3}.

When the loading concentration of formaldehyde is far below the concentration of Na_2SO_3 solution, the absorption rate was hardly influenced by the loaded formaldehyde. In contrary, when the loading content of formaldehyde approaches or exceeds the concentration of Na_2SO_3 solution, a reduction of the absorption rate could be apparently observed.

Oxygen in air can also react with Na_2SO_3 solution. However, if there is no heavy metal ion as a catalyst in the solution, the reaction rate will be very slow, and the effect of this reaction on the absorption rate of formaldehyde can be neglected [23].

Fig. 8 shows the comparison of the theoretical data with the experimental ones at Na_2SO_3 concentrations of 0.01, 0.05, 0.10, 0.30 kmol m^{-3}. The calculated values are in good agreement with the experimental data with an average deviation of 9.14%, indicating that the mathematical model based on the film-theory and the chemical reaction enhancement factor is valid for such a mass transfer process of low formaldehyde partial pressure.

5. Consideration of practical absorption process

Although the formaldehyde concentration could reach ppm level in some serious heavily polluted cases, the commonly observed formaldehyde concentration is at ppb level. However, the precision of analysis method used in this experiment limits the study of ppb level. We will predict the absorption of ppb level formaldehyde into Na_2SO_3 solution by theoretical calculation.

The reaction equilibrium constant in Eq. (2) can be written as:

$$k_2 = \frac{[CH_2(O^-)SO_3^-]}{[SO_3^{2-}][CH_2O]} \tag{18}$$

where $k_2 = 9.46 \times 10^6$ kmol^{-1} m^3 s^{-1} [17] at 25 °C. Combining Eq. (9) with Eq. (18), we have

$$[CH_2O(g)]^Y = (55.56 \times 10^X \times k_2)^{-1} \frac{[CH_2(O^-)SO_3^-]}{[SO_3{}^{2-}]} \tag{19}$$

Substituting the data of k_2, X and Y into Eq. (19) generates Eq. (20),

$$[CH_2O]^{1.06} = 2.45 \times 10^{-13} \times \frac{[CH_2(O^-)SO_3{}^-]}{[SO_3{}^{2-}]} \quad (20)$$

Even when the depleted Na_2SO_3 is 90%, the volume fraction of the equilibrium formaldehyde concentration is calculated from Eq. (20) to be 1.01×10^{-11}, which is about two orders of magnitude below the ppb level (10^{-9}). Therefore, we believe that the Na_2SO_3 solution is an effective absorbent for the removal of formaldehyde from polluted air even the formaldehyde concentration was at ppb level.

Since the absorption rate almost depends on the gas phase resistance, the design of the absorption process will focus on how to increase the agitation of gas phase and the mass transfer area. The vessel used in this study is suitable for the experimental determination of mass transfer rate of the absorption, but it seems too simple to fulfill the requirements above. Instead, a mini-type packed tower of high mass transfer area and separation efficiency should replace the vessel for the practical applications.

It is assumed that the room to be cleaned is of 40 m^2 and 3 m height. The formaldehyde concentration should be reduced from 1000 to 5 ppb, and the indoor air that has a total volume of 120 m^3 should be circulated twice each day and cleaned in 3 h. The designed packed column is loaded stainless steel wire mesh rings and has an inner diameter of 200 mm. The specific area, a, of the packing is about 1200 $m^2\,m^{-3}$. The pollutant gas and the absorbent are in counter current contact with each other in the column, the gas and liquid flow rates are 0.0158 kmol $m^{-2}\,s^{-1}$ and 1.50l $m^{-2}\,s^{-1}$, respectively. The cleaning operation will be done under atmospheric pressure and room temperature.

With the use of the absorption model in this study, the height of the packing, h, can be calculated from Eq. (21) [24]:

$$h = \frac{G}{aPK_G} \ln \frac{p_1}{p_2} \quad (21)$$

where G is the gas flow rate, P the operating pressure, K_G represents the absorption transfer coefficient and p_1 and p_2 are the partial pressure of the inlet and outlet pollutant gas, respectively. By substituted the parameters, the height of the packing could be obtained as 0.42 m, and the total height of the equipment is believed within about 1 m. This value located at a quite suitable size for indoor room applications.

The life expectancy of the absorbent is also an important factor in practical applications. Provided that the circulation volume of Na_2SO_3 solution is 20 l with the concentration of 0.5 kmol m^{-3} and the maximum consumption of the formaldehyde in the solution is set to 90% of the originally loaded amount, the theoretical life expectancy of the absorbent could be predicted as:

$$\frac{0.5 \times 20 \times 90\%}{2 \times 40 \times 3 \times (1000 - 5) \times 10^{-9} \times 1000/22.4} \approx 844 \text{ days} \quad (22)$$

As can be seen from the calculation, the method developed in this study is believed to be a feasible way for practical indoor applications. The experimental determinations of the scaled-up processes will be further investigated in our following work.

6. Conclusions

The absorption rate of lean formaldehyde in air by the use of Na_2SO_3 solutions is measured in a stirred vessel with a plain gas–liquid interface. The results of this study show that the Na_2SO_3 solution is an effective absorbent to remove lean formaldehyde from polluted air. The gas flow rate and the formaldehyde concentration are the most important factors that affect the mass transfer rate. The Na_2SO_3 concentration is found to have little effect on the absorption rate of formaldehyde, while the loading content of formaldehyde reduces the absorption rate only if the formaldehyde concentration approaches or exceeds the concentration of the Na_2SO_3 solution.

A theoretical model based on the film-theory and chemical reaction enhancement factor has been successfully derived to represent the experimental absorption rates with average relative deviation less than 10%. With the use of the absorption model in the study, the minitype packed column has been selected for practical application. The size of the equipment and the life of the absorbent have been calculated and are believed feasible for practical indoor usage.

Acknowledgment

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