

Construction of an aeration stripping reactor based on a novel filler and stripping kinetics of carbon tetrachloride in groundwater

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ABSTRACT

To target the problems of easy scaling of filler, easy clogging of flow channels and low mass-transfer efficiency in traditional fixed packed-bed aeration stripping technology, a combined blade structure blow-off filler was invented, and the hydrodynamic characteristics of the internal gas-liquid interface were simulated and analyzed by computational fluid dynamics simulation software. The result showed that the combined air-blade structure blow-off filler had a significant effect on the mass transfer of carbon tetrachloride (CCl_4) in the gas-liquid phase. A pilot-scale study on the removal efficiency of CCl_4 in groundwater was carried out by a self-designed fixed-bed aeration stripping pilot system. The gas-water ratio and filling ratio had a significant effect on the removal rate of CCl_4 and the gas-liquid mass-transfer coefficient $K_L a$, whereas the initial concentration had little effect on these parameters. To promote the technology to China's water-supply plants, it is necessary to formulate a cost-effective plan based on the water-quality characteristics and to select an appropriate gas-water ratio and filling ratio to achieve the desired removal effect.

Keywords: Fixed packed bed; Aeration stripping; Pilot-scale experimental study; Carbon-tetrachloride removal; Combined vane-structure stripping filler

1. Introduction

In recent years, carbon tetrachloride (CCl_4) has been found to be a relatively common volatile chlorinated hydrocarbon organic pollutant in groundwater. It is usually stable in nature, not easy to degrade by environmental microorganisms, and it is lipid-soluble and easily absorbed by the skin. It is teratogenic, carcinogenic, and mutagenic and threatens human health [1–4]. Researchers have carried out extensive research to develop efficient CCl_4 treatment technology [5–7]. The main technologies that have

been studied include activated-carbon adsorption, membrane filtration, photochemical oxidation, zero-valent iron reduction, and aeration stripping [8–11]. Aeration stripping has become a topic of great interest to scholars in many countries recently because of its unique advantages [12]. Zhang et al. [13] used the combined technology to treat CCl_4 pollution in groundwater. The results showed that the effluent could reach the limit of the “drinking water hygiene standard” (gb5749-2006) stably for a CCl_4 concentration in raw water of 11.30 g/L and gas-water ratio of 5.75. Benner et al. [14] used aeration stripping to remove volatile

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organic compounds in groundwater from Chicago in the United States, with a removal rate of 88%. Zamarron [15] treated disinfection by-product trihalomethanes in drinking water in the same way. The removal rate of total trihalomethanes could exceed 90% when the gas-water ratio reached or exceeded 60. Compared with other processes, fixed packed-bed aeration has advantages of sufficient gas-liquid contact and a flexible change of gas-liquid ratio in a wide range. The innovation in this study lies in the independent design of a fixed-bed aeration stripping pilot system and the invention of a new composite packing, combined with a vane-structure stripping filler. The packing units can be combined in the system according to a certain filling ratio. Driven by water flow and airflow, the impeller can rotate in the packing, which improves the gas-liquid mass-transfer efficiency significantly. This study demonstrates the effect of the gas-water ratio, initial concentration and packing filling on the removal efficiency of the raw water that contains CCl_4 , and uses computational fluid dynamics (CFD) to simulate the conditions inside the packing unit, to provide related data for the promotion of aeration stripping in China.

2. Materials and methods

2.1. Apparatus and operating conditions

The experimental device that was used in this study is shown in Fig. 1. The field experiment was conducted at the Dongyuan Water Plant in Jinan City where the fixed-bed aeration stripping pilot-system treatment scale was

$5 \text{ m}^3/\text{h}$. The device was 135 cm long, 65 cm wide, and 180 cm high, the material of which was carbon structural steel. It was divided into three chambers I, II, and III (the filler was used only in chambers I and II in the experiment). Aeration discs were located at the bottom of the device, and each chamber corresponded to two discs. The upper portion of the aeration plate was a support grille, and the filler could be stacked on the upper part of the grille. The linear low-density polyethylene water tank had an effective volume of 10 m^3 . The inner submersible pump was a model 50WQ8-15-1.1; the dosing pump model was a DCM0408; the static-mixer model was an SK-68650; the gas glass rotor flowmeter was a model LZB-25 with a $2.5\text{--}25 \text{ m}^3/\text{h}$ range; the liquid glass rotor flowmeter was a model LZB-50 with a $0.6\text{--}6 \text{ m}^3/\text{h}$ measuring range and the Roots blower was a model RSS-40. The activated-carbon adsorption device was a columnar activated-carbon absorber.

Groundwater from the Dongyuan water plant was stored in the water tank and pumped into the pipeline by a submersible pump. The prepared halogenated hydrocarbon solution was transported to the pipeline by a dosing pump, where it joined with the raw water to enter the static mixer. After complete mixing, the water entered the main device of the system. Inside the device, the water was blocked by the folding plate, the flow direction changed twice, and finally, it flowed out through the outlet. Roots that fanned from the bottom delivered air to form bubbles through the aeration disk, and the air was pumped into the device above. During the rising process, the bubbles were in contact with the water in the packing layer. The chlorinated

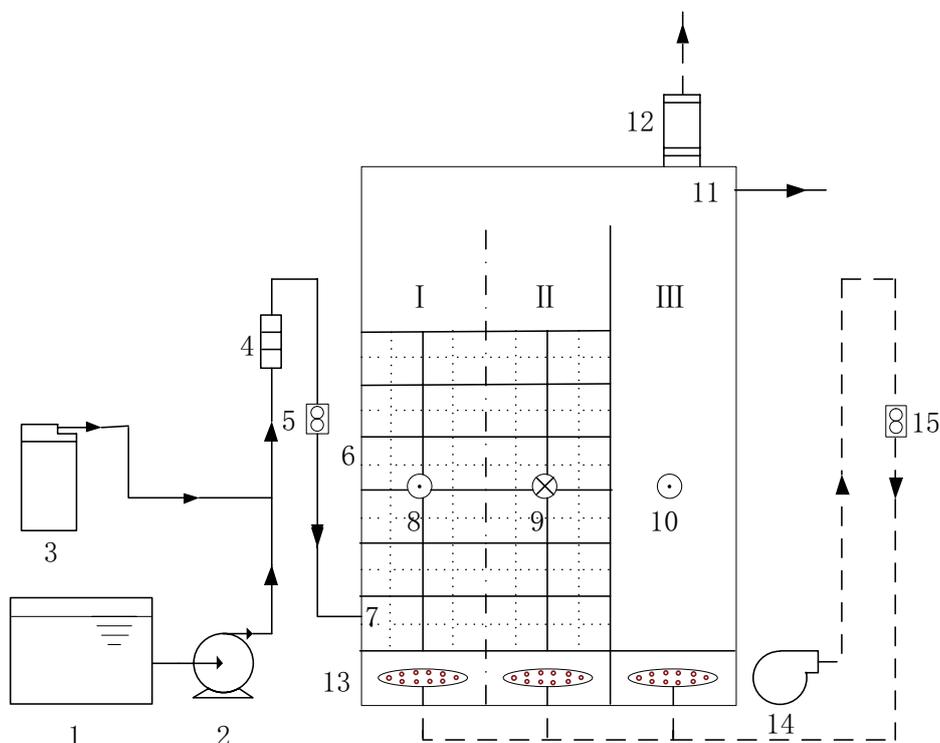


Fig. 1. Pilot-scale experimental system. (1) Water tank, (2) centrifugal pump, (3) chemical addition unit, (4) static mixer, (5) liquid rotameter, (6) packing layer, (7) water inlet, (8–10) correspond to sampling ports (1–3), respectively, (11) water outlet, (12) activated carbon adsorption device, (13) disc diffuser, (14) blower, and (15) gas rotameter.

hydrocarbons in the water were carried away by the bubbles, and the waste gas was absorbed by the activated carbon and discharged into the atmosphere. During the process, samples were taken from the inlet, outlet, and sampling ports at intervals to test the concentration of chlorinated hydrocarbons.

2.2. Combined vane-structure stripping filler

Self-designed combined vane-structure blow-off packing was used in the device. The structure of the packing unit is shown in Fig. 2. The packing unit was composed of an outer frame, an impeller, a column pin, an upper platen, and a lower platen. The outer frame was a hollowed-out cubic structure that was composed of nine square hollow columns and reinforcing bars, and the bottom was provided with a base plate. The framework was lined up with four-wheel groups that each included nine layers of the impeller. Each layer in the middle of the impeller was equipped with a central hole. The angle between the blades and the horizontal plane ranges from 10° to 30° . The nine-layer impeller was connected by the column pin and could rotate around the column pin. The upper part of the four impeller sets was fixed by an upper pressing plate, and the lower part was fixed by a lower pressing plate.

The outer dimensions of the frame were $200 \text{ mm} \times 200 \text{ mm} \times 200 \text{ mm}$, and the thickness of the polypropylene bottom plate was 2 mm. The blades of the impeller formed a spiral, and ten blades were distributed evenly on the outer cylinder. The outer diameter of the impeller was 90 mm, the diameter of the outer cylinder was 20 mm, the height was 14 mm, the inner cylinder was 21 mm high, and the diameter of the central hole was 10 mm with a height of 21 mm. The thickness was 1 mm. The pin size had a 5 mm diameter and was 195 mm long. The rib section was $5 \text{ mm} \times 6 \text{ mm}$ and was arranged along the upper and lower sides of the frame. The upper pressing plate had a protrusion, and the lower pressing plate had a groove, and the filling unit

could be spliced through the protrusion and the groove to achieve a combination of the packing units.

Because the angle between the blade and the horizontal plane was between 10° and 30° , rotation occurred under the action of the water flow driving force. The rotation speed (symbol: n , unit: r/s) followed the water flow velocity (symbol v , unit: m/s) increases and had the following relationship: $v = \pi n d$. As the flow rate of the water increased, the flow rate increased, and thus, the rotation speed of the blade increased, the renewal speed of the gas–liquid contact surface inside the packing unit was accelerated, and the mass transfer was accelerated. The process was conducted efficiently. The gas–liquid contact area of each impeller was $S = 0.25 \pi d^2$. Each group had nine layers of impellers, and the corresponding gas–liquid contact area was $S' = 2.25 \pi d^2$. Each packing unit was composed of four sets of impellers, and the total gas–liquid contact area was $S'' = 9 \pi d^2$. If the impeller rotated under the action of the water-flow driving force and the rotation speed was n , the contact area $S''' = 9 n \pi d^2 = 9 v d$ with rotation. Under normal circumstances, the flow rate of the general aeration stripping tower was 0.7–1 m/s, the outer diameter of the packing was $d = 9 \text{ cm}$, and the gas–liquid contact area during rotation was 0.57–0.81 m^2/s . Compared with traditional fixed packing, the gas–liquid contact area improved during the same contact time. The rotation of the impeller promoted the uniformity of the gas–liquid distribution in the packing layer, and the mass-transfer efficiency was improved because the packing had larger pores. Wastewater after the completion of mass transfer out of the pores was recycled to ensure the continuity of the system operation.

2.3. Analytical methods

CCl_4 in the water was determined by headspace gas chromatography. An electron-capture detector in the GC-2010 gas chromatograph (Shimadzu, Japan) was used [16].

To explore changes in the flow state in the device, CFD software was used to analyze a selected packing unit under

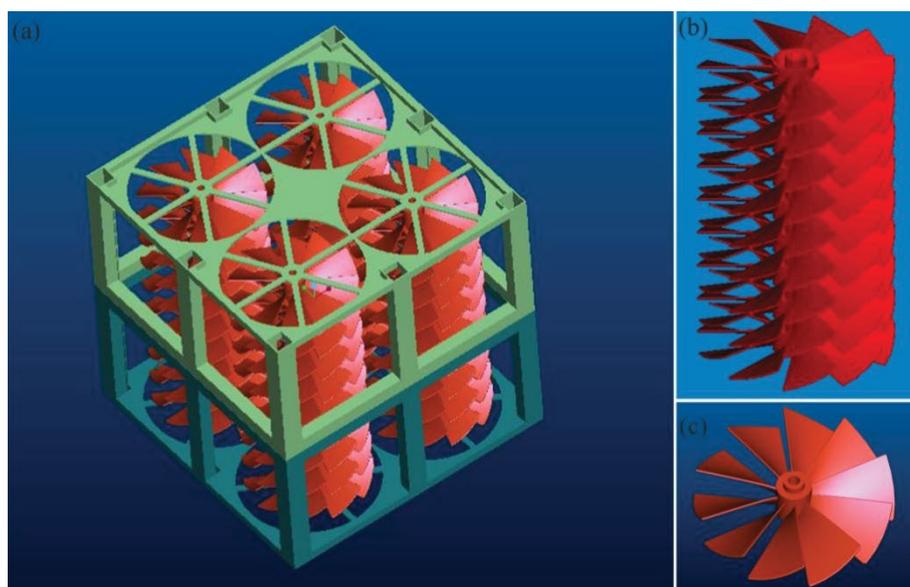


Fig. 2. Combined vane-structure stripping filler.

the stated condition (the filling ratio of the device was 60%, the inlet flow was 5 m³/h, and $Q_G/Q_L = 5$).

We measured the removal rate of the CCl₄ η and the gas–liquid mass-transfer coefficient $K_L a$ that corresponds to different conditions.

2.4. Calculation of air stripping efficiency and mass-transfer coefficient of CCl₄

The removal rate of CCl₄ η is given by:

$$\eta = \frac{(C_0 - C_t)}{C_0} \times 100\% \quad (1)$$

where C_0 is the initial concentration of CCl₄ in the raw material water, mg/L, and C_t is the concentration of CCl₄ in the liquid phase at any time t in the device, mg/L.

Muller et al. [17] showed that in an aeration stripping system, the mass-transfer expression of volatile organic compounds A per unit volume of water is given by:

$$-\ln \frac{C_{A_t}}{C_{A_0}} = \frac{Q_G H_A}{V_L} \left[1 - \exp \left(-\frac{K_L a V_L}{H_A Q_G} \right) \right] t \quad (2)$$

Among them, C_{A_t} and C_{A_0} are the concentrations of volatile organic compounds A in the liquid phase at time t and initial time, g/m³; H_A is the Henry's constant, which is a dimensionless quantity; K_L is the total liquid mass-transfer coefficient, m/min; a is the unit interfacial area of the volumetric liquid, m² m⁻³; V_L is the total volume of the liquid, L; Q_G is the gas flow rate, l/min; and t is the stripping time, min.

When $K_L a V_L / H_A Q_G \ll 1$, Eq. (2) can be converted to Eq. (3):

$$-\ln \frac{C_{L,A_t}}{C_{L,A_0}} = K_L a t \quad [18] \quad (3)$$

This situation only occurs when the two-phase gas–liquid in the device has not reached equilibrium and the concentration of CCl₄ is far below saturation.

In this test, the influent flow rate was set to 5 m³/h, and after 20 min of blow off treatment, the treated raw water flowed out of the device. Therefore, the residence times at the water inlet; sampling ports 1, 2, and 3; and the water outlet were 0, 5, 10, 15, and 20 min, respectively. After the test, the concentration of CCl₄ in the gas phase before sampling port 3 did not reach saturation. Therefore, before the 15 min residence time, the gas–liquid mass-transfer coefficient $K_L a$ was estimated by Eq. (3).

3. Results and discussion

3.1. CFD simulation

The fluid-mechanical simulation of the gas–liquid flow state before and after the addition of the filler unit to the fixed-fill aeration device was carried out by CFD simulation. The results are shown in Fig. 3.

Fig. 3 shows that the pressure of the gas–liquid contact surface increased gradually from top to bottom, and the pressure at the inlet was highest. The upper and lower

pressure difference after the filler addition is larger than that without the filler. The increased pressure difference increases the rising speed of the bubble significantly and contributes to a renewal of the gas–liquid contact surface. Without filler, the gas–water interface is concentrated in the upper part. After filler addition, impeller rotation causes the bubble to diverge to the surroundings, and the distribution of the gas–water interface becomes more dispersed throughout the packing unit. After filler addition, the flow state of the gas–liquid contact surface was more disordered than that without filler. The simulation results show that the ratios of total bubble area to total bubble volume in the packed unit S_Q/V_Q are 208.7 and 124.3, respectively, under the condition of with filler and without filler, and the ratio of with filler was 40% higher than that of without filler. The results are explained as follows. The impeller inside the packing unit rotates under the action of water flow, and the airflow bulges from below. Under the cutting action of a rotating blade, big bubbles are broken into tiny bubbles, and the amount of which increases gradually. In the process, CCl₄ is removed. Gas–liquid mass transfer occurs from the liquid phase to the gas phase, and tiny bubbles increase the gas–liquid contact area, so the gas–liquid mass-transfer process can be completed to a greater extent. Therefore, filler addition improves the CCl₄ removal effect significantly.

To describe the distribution characteristics of the gas–liquid flow velocity in the local part of the packing, a velocity plot at $X = 0.048$ is shown in Fig. 4. Figs. 4a and b show that in the state without filler, the large gas and liquid flow rates are concentrated mostly in the upper part; and Figs. 4c and d show that, with the filler, the positions where the gas and liquid flow rates are large are relatively dispersed. The maxima exist in the middle of the two sets of impellers. This occurs mainly because the impeller rotation increases the gas-diffusion area, and the tiny bubbles and the liquid are in full contact in the middle of the two sets of impellers. The impeller rotation accelerates the renewal of the gas–liquid contact surface, which allows the mass-transfer process to be more fully completed. When the gas–water ratio was 1, 2, 3, and 5, the impeller speed was 10, 20, 30, and 40 rpm, respectively. As the gas–water ratio increased, the impeller speed increased gradually. The frequency of the bubble breakage also increased gradually, the gas–liquid flow turbulence increased gradually, and the mass-transfer effect improved significantly.

3.2. Effect of Q_G/Q_L

The effect of the gas–water ratio on the removal rate of CCl₄ is shown in Fig. 5. An increase in residence time increases the removal rate of CCl₄ gradually, and the increasing trend is obvious in 0–15 min, and slow in 15–20 min, the gas–water ratio and residence time are the main factors that affect the effect of blowing off. Some studies had shown that within a certain range, the aeration time, gas–water ratio, and the removal rate of chlorinated hydrocarbon were positively correlated, and the increase of the gas–water ratio and the prolonging of the aeration time will slow down the growth trend of removal rate [19]. The behavior may result because the CCl₄ concentration in the gas–liquid phase of the device reaches an equilibrium state within ~15 min,

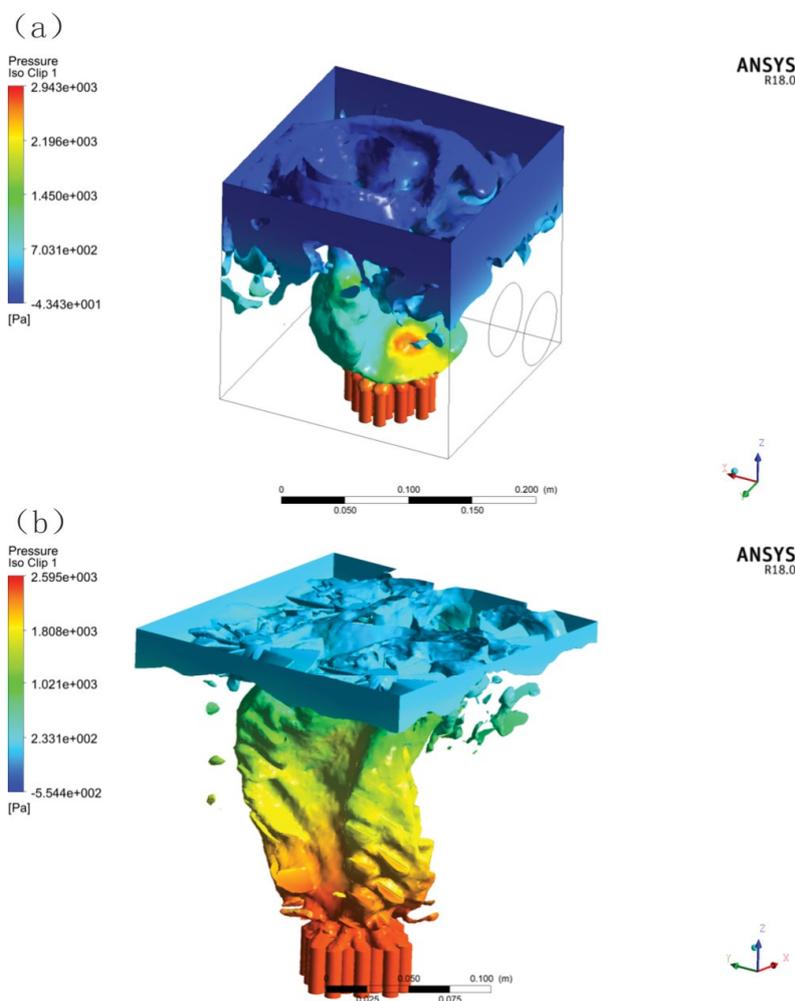


Fig. 3. Pressure-distribution diagram of gas-water interface (a) no filler and (b) with filler.

and the effect of an increase in aeration time on the removal rate is almost insignificant. The device has a good removal effect on CCl_4 . When the gas-water ratio is 1 and the residence time is 20 min, the removal rate of CCl_4 reaches 68%. For the same residence time, with an increase in gas-water ratio, the removal rate of CCl_4 increased. When the residence time was 15 min, the removal rate increased by 9% from 66% to 75% as the gas-water ratio changed from 1 to 2. When the gas-water ratio changed from 2 to 3, the removal rate changed from 75% to 91%, which represents an increase of 16%. When the gas-water ratio changed from 3 to 5, the removal rate changed from 91% to 94%, which is an increase of only 3%. A gas-water ratio of 3 was optimal, and the concentration of CCl_4 in the effluent could meet the “drinking water hygiene standard” requirements (gb5749-2006).

As mentioned above, for a 15 min residence time, the concentration of CCl_4 in the gas and liquid phases reaches an equilibrium state. The change in CCl_4 in the device conforms to Eq. (3) during 0–15 min. The plot of $-\ln(C_i/C_0)$ vs. the time allowed us to obtain the gas-liquid mass-transfer coefficient ($K_L a$) for different gas-water ratios through a linear fitting, as shown in Fig. 6. The linear fitting result was ideal ($R^2 = 0.9935\text{--}0.9984$). With the increase in gas-water ratio,

$K_L a$ also increased gradually, which is similar to the effect of the gas-water ratio on the removal rate of CCl_4 . When the gas-water ratio changed from 1 to 2, the $K_L a$ changed slightly from 0.07117 to 0.0935 min^{-1} . When the gas-water ratio changed from 2 to 3, the $K_L a$ increased significantly from 0.0935 to 0.15879 min^{-1} . When the gas-water ratio changed from 3 to 5, the changes were not obvious. Therefore, when the gas-water ratio was 3, the diffusion rate of CCl_4 from the liquid phase to the gas phase was faster, which can be used as the optimal production condition.

A combination of Figs. 5 and 6 show that the gas-water ratio of 2 is a critical value. When the gas-water ratio is less than 2, it has little influence on the removal rate of CCl_4 and $K_L a$, but when it exceeds 2, it has a very obvious influence. The reasons for this behavior are as follows. When the gas-water ratio is less than 2, less gas exists per unit volume of water, and the formed shear stress cannot resist the resistance of the gas-liquid contact surface. When the gas-water ratio exceeds 2, more gas is present. The shear stress can offset the resistance to a large extent. Air enters the liquid phase and carries the CCl_4 in the liquid, which increases the removal rate significantly [20]. Airflow drives the impeller inside the packing to rotate. With the increase in gas flow

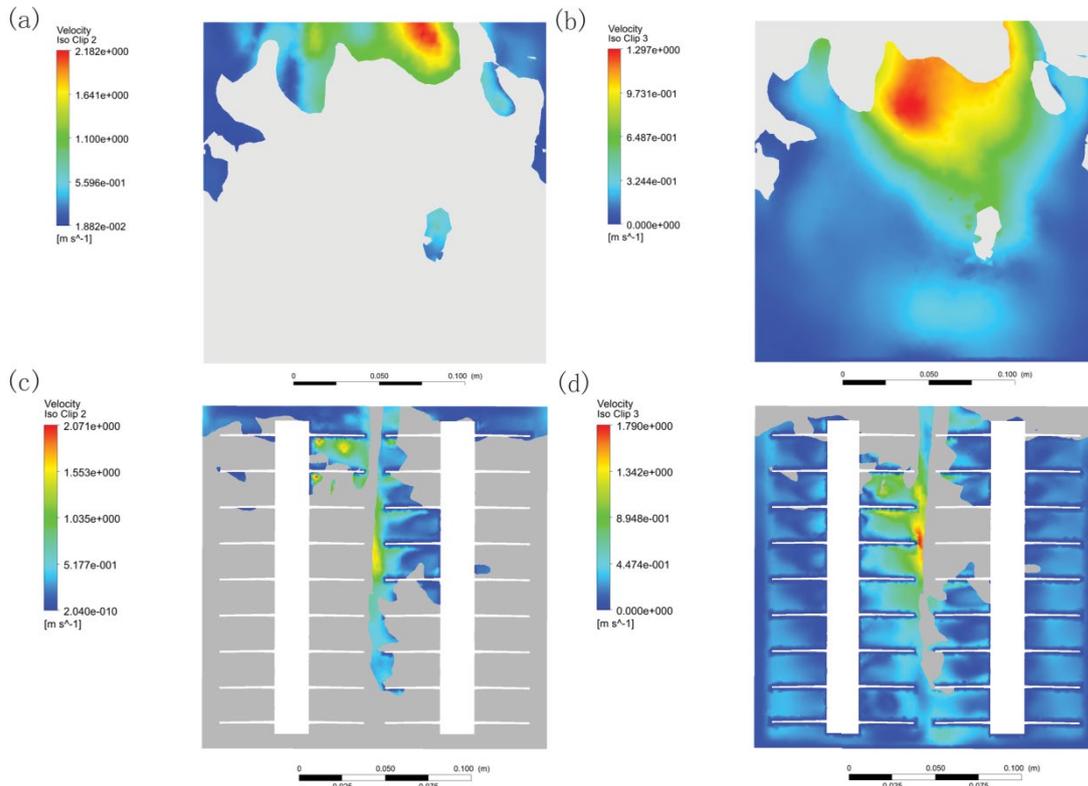


Fig. 4. Velocity plot at $X = 0.048$, (a) no filler, gas, (b) no filler, liquid, (c) with filler, gas, and (d) with filler, liquid.

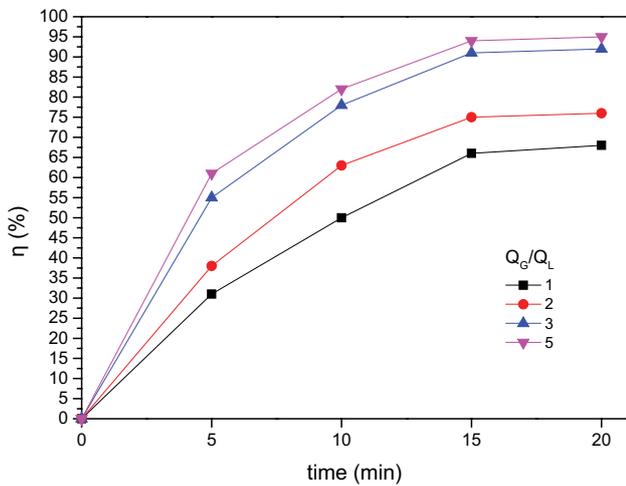


Fig. 5. Effect of Q_G/Q_L on air stripping of CCl_4 . Experimental conditions: fill ratio = 60%, $Q_L = 5 \text{ m}^3/\text{h}$, initial CCl_4 concentration 0.01 mg/L.

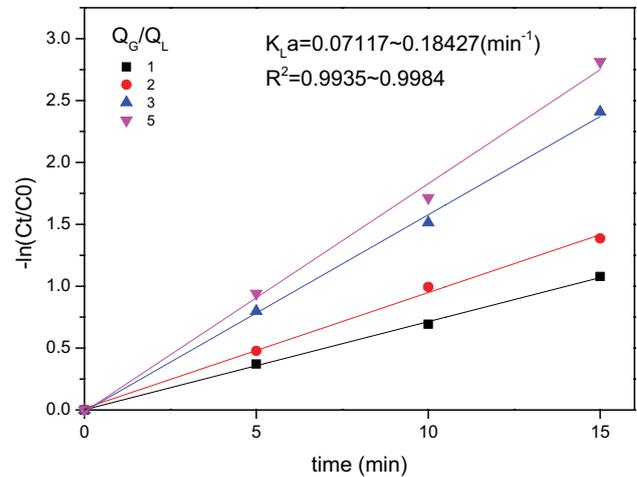


Fig. 6. Effect of Q_G/Q_L on the mass-transfer coefficient of CCl_4 removal. Experimental conditions: fill ratio = 60%, $Q_L = 5 \text{ m}^3/\text{h}$, initial CCl_4 concentration 0.01 mg/L.

rate, the speed of the impeller rotation is accelerated and the rotating impeller agitates the water flow, so the flow converts from laminar flow to turbulence. During the rising, bubbles that rotate around the impeller are broken into smaller bubbles, which increases the gas–liquid contact area significantly. Therefore, the mass transfer of CCl_4 between the gas and liquid phases increases significantly by a combination of the two effects [21].

3.3. Effect of initial concentration

The effect of the initial concentration on the CCl_4 removal rate and the $K_L a$ is shown in Figs. 7 and 8. The removal rate of CCl_4 increases with an increase in residence time. At 10 min, the removal rate of CCl_4 at each initial concentration exceeded 85%, and the concentration of CCl_4 from two sampling ports met the requirements of the “drinking water hygiene standard” (gb5749-2006).

An increase in the initial concentration did not change the removal rate of CCl_4 significantly, and the gas-liquid mass-transfer coefficient $K_L a$ changed only within the range of 0.18892–0.20422 min^{-1} . Therefore, the initial concentration has little effect on the removal of CCl_4 , and this is a remarkable feature of the blow-off method [22]. This behavior can be explained by the mass-transfer process. Eq. (3) can be arranged to yield Eq. (4).

$$\ln(1 - \eta) = -K_L a t \quad (4)$$

The $K_L a$ that corresponds to the highest and lowest concentrations in the same blowing period is almost a fixed value. Therefore, according to Eq. (4), the removal rate of the highest and lowest concentrations is almost constant. However, with an increase in initial concentration, the mass-transfer rate of the CCl_4 in the gas-liquid phase also

increases slightly, which occurs mainly because of the increase in concentration. This change can improve the mass transfer of CCl_4 molecules, and yield a higher removal rate of CCl_4 .

3.4. Effect of fill ratio

The effect of filling ratio on the removal rate and $K_L a$ is shown in Figs. 9 and 10. The filling ratio of the filler has a significant impact on the removal rate of CCl_4 . The removal rate of CCl_4 increased gradually with an increase in the filling ratio. When the filling ratio was 20%, 40%, and 60%, the final removal rate of CCl_4 in the effluent water was 79%, 84% (with a difference of only 5%), and 95%, respectively.

Under the same gas-water ratio, the effect of fixed packed bed aeration on the removal of carbon tetrachloride

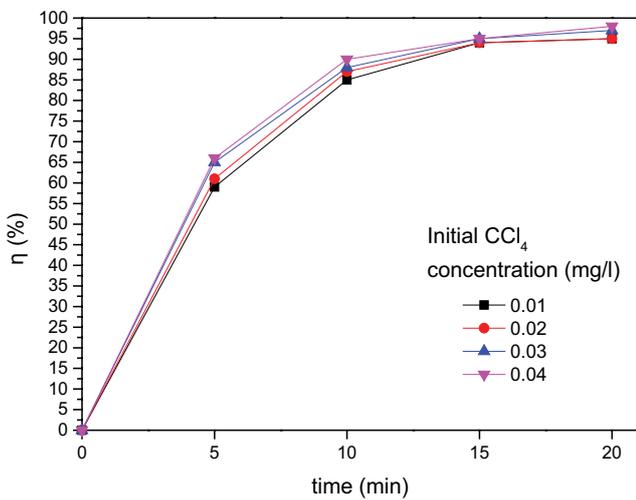


Fig. 7. Effect of initial CCl_4 concentration on air stripping of CCl_4 . Experimental conditions: fill ratio = 60%, $Q_L = 5 \text{ m}^3/\text{h}$, $Q_G/Q_L = 5$.

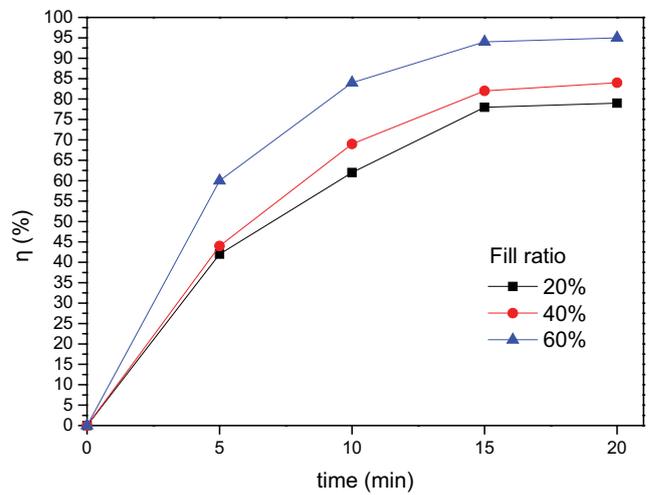


Fig. 9. Effect of fill ratio on air stripping of CCl_4 . Experimental conditions: initial CCl_4 concentration of 0.01 mg/L, $Q_L = 5 \text{ m}^3/\text{h}$, $Q_G/Q_L = 5$.

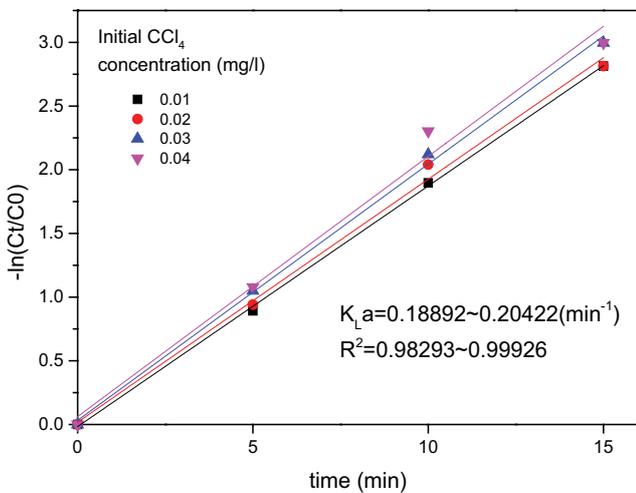


Fig. 8. Effect of initial CCl_4 concentration on the mass-transfer coefficient of CCl_4 removal. Experimental conditions: fill ratio = 60%, $Q_L = 5 \text{ m}^3/\text{h}$, $Q_G/Q_L = 5$.

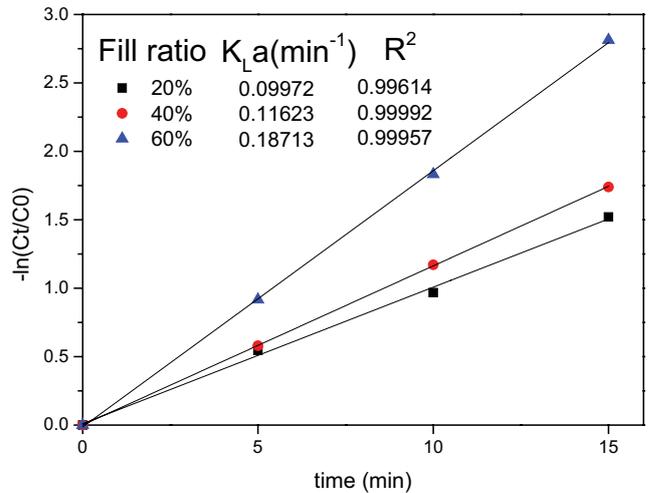


Fig. 10. Effect of fill ratio on the mass-transfer coefficient of CCl_4 removal. Experimental conditions: initial CCl_4 concentration of 0.01 mg/L, $Q_L = 5 \text{ m}^3/\text{h}$, $Q_G/Q_L = 5$.

was better than that of empty tower aeration and suspended packing aeration, and the removal rate of carbon tetrachloride was increased by about 5%–10%, which was the same as that of spray tower. Fixed packed bed aeration process on chloroform, trichloroethylene, and tetrachloroethylene and other volatile substances removal effect increased more obvious, removal rate increased about 10%–20% [23].

4. Conclusions

Aeration stripping technology has broad application prospects in the field of groundwater volatile organic compounds processing. To resolve problems of easy scaling of the aerated blow-off packing, easy clogging of the flow passage, and low mass-transfer efficiency of the traditional fixed packed bed, a combined vane-structure stripping filler was invented. The filler has a large porosity, can be combined and has a regular flow path. Its scale-up is difficult, the internal impeller can be rotated, and its mass-transfer efficiency is high. The CFD simulation results show that the combined vane-structure blow-off packing can distribute the bubbles in the device evenly and reduce their size. The gas–liquid contact-surface renewal speed is increased, and the gas–liquid phase turbulence is also increased. The liquid mass-transfer efficiency and the removal of CCl_4 were improved.

The removal efficiency of CCl_4 was studied by a fixed-bed-aeration stripping pilot system, which could remove CCl_4 . When the residence time was ~15 min, the two phases of gas and liquid reached equilibrium, and with the increase in gas–liquid ratio and filler filling ratio, the removal rate of CCl_4 η and the gas–liquid mass-transfer coefficient $K_L a$ increased significantly before 15 min. The initial concentration had little effect on the removal rate and the mass-transfer coefficient. Under present conditions, the gas–water ratio was 3, and the CCl_4 concentration reached the national standard limit when the filler filling ratio was 40%. When the technology is extended to water plant application, a cost-effective plan should be formulated according to the quality characteristics of the water to be treated in the plant, and the appropriate gas–water ratio and filling ratio should be selected to achieve the desired removal effect.

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