Research on Influencing Factors of Biological Filtration Tower Treating Toluene Gas

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Abstract. Through the orthogonal experimental design, the optimal combination of Triton X-100, nitrogen source, Fe$^{2+}$, temperature, concentration of antibiotics, pH and spray quantity was determined with surfactants, nitrogen and iron elements as additive, by which the key influencing factors were determined. In the test, the removal efficiency of the second groups was higher than that of the eighth groups, which were 89% and 87%, respectively. The best combination of a group of removal was as follows: nitrogen source concentration was 2 g·L$^{-1}$, antibiotic concentration was 300 U·mL$^{-1}$, the concentration of Triton X-100 was 0.05 mL·L$^{-1}$, Fe$^{2+}$ concentration was 14 mL·L$^{-1}$, pH was 7, the temperature was 34 ℃, spray amount was 6 L·h$^{-1}$. The antibiotic concentration was the most important factor on the removal efficiency of the toluene. The concentration of gas in each layer of toluene was detected; the curve of the outlet concentration in the optimal combination and the average state was obtained. The removal efficiency of the optimal combination was much better than the average, and it was found that the removal rate decreased with the increase of the height of the filling layer. The change of oxygen content in each layer was detected which was no significant change. It showed that oxygen was not the limiting factor of the removal of toluene by microorganisms.

Keywords: surfactants; biological filtration tower; toluene; orthogonal test

1. Introduction
Volatile Organic Compounds (VOCs) are a class of chemical compounds which are extremely complex. They are not only easily lead to photochemical smog which is harm to human and plant health, but also, after entering the atmosphere, they are participate in a series of chemical reactions to produce ozone and destroy the atmosphere, thus affect the global climate environment. Toluene is the most common volatile organic compounds, belonging to the benzene series, and is a very important chemical raw material. Benzene has been identified as a strong carcinogen by the World Health Organization (WHO) [1], and long-term exposure may cause anemia or leukemia [2-3].

There are many kinds of methods to be used to remove or reduce VOCs levels, the conventional physical and chemical methods include: catalytic incineration, adsorption, absorption, condensation, etc [4-5]. Among the new methods such as non-equilibrium plasma method, photo catalytic method and biological method, biological method because of its good effect, simple facilities, low cost, environmental protection, easy to manage and so on, it has been paid more and more attention by scholars both at home and abroad. With the characteristics of strong and fast adaptation to the
pollutants, biological method domesticates the microorganisms. Pollutants are used as microorganisms’ carbon source and energy, and finally been transmuted into harmless substances (H₂O, CO₂, etc.).

The process of biological treatment of volatile organic compounds mainly has three kinds of methods: biological washing, biological filtration and biological trickle filtration. This study mainly aimed at the biological trickle filtration method in which the gas is from the bottom of the tower. In the process of flow, gas is purified by contacts the biological membrane. The recycled spraying cleaning solution enters the tower from the top of the packing layer, then sedimentation at the bottom of the tower through the biological membrane.

In this study, the activated carbon was used as filler together with the domesticated strain in the treatment of toluene. Using of additives to improve the water solubility of hydrophobic organic compounds and microbial activity. Therefore, we can provide scientific basis for the application of the biological filter in the future.

2. Device and method

2.1. Experimental installation
The biological trickle filtration is made of transparent organic glass and in the type of sectional detachable. Tower diameter is 120mm the height of each layer is 150mm, and total of 4 layers. The activated carbon was used as filler. The experiment uses gas-liquid countercurrent operation so the nutrient solution flows from the top of the tower and flows out from the bottom of the tower. The waste gas, on the contrary, flows from the bottom to the top.

2.2. Experimental method

2.2.1. Detection method. Toluene detected by People’s Republic Of China environmental protection standard of “Ambient air—Determination of benzene and its analogies by activated charcoal adsorption carbon disulfide desorption and gas chromatography”.

The Bruke 400-GC gas chromatograph was used with BR-SWax (Carbowax® polyethylene glycol) (30 m×0.32 mm×0.5 µm) capillary column, injection port temperature was 150°C, split ratio 1:100; kept the temperature of column heater at 50°C for six minutes, then heated it at a rate of 25 °C•min⁻¹ up to 125°C and kept 3 minutes at this temperature; the flow rate of column was kept steady at 0.70 mL•min⁻¹; the detector temperature was 270°C and the sensitivity of the detector was 12. Nitrogen flow rate: 25 mL•min⁻¹. Hydrogen flow rate: 30 mL•min⁻¹. Air flow rate: 300 mL•min⁻¹.

2.2.2. Additive selection. Surface active agent was chosen, nitrogen element and iron element as additives. Because the surface active agent has the hydrophilic base and the oil base, so a small quantity of it can reduce the mass transfer resistance between gas and liquid. As the result, the solubility of hydrophobic gas such as toluene in aqueous solution will be increased [6-7].

The non-ionic surfactant named Triton X-100 was selected as a solubilizer in order to increase the contact of toluene and microorganisms which was advantageous to improve the efficiency of removing [8]. At the same time, the study of Berti had discovered that by adding 1% of methyl oleate into Triton X-100 can form the oil-in-water emulsion and capacity of micellar solution [9]. The emulsion increased the gas-liquid contact area of organic phase [10]. So the effects would be more obvious if we add 1% of methyl oleate.

Trace metal ions was the activator or active group of enzyme [11-13]. Nitrogen and Fe²⁺ were used to improve activity of microorganisms. Carbon source and nitrogen source were essential nutrient elements for microbial growth, so microbes absorbed carbon source by capturing toluene in the liquid and improved the biological activity by adding external nitrogen source. After Fe²⁺ dissolving into water, hydrated ions with water molecules such as Fe(H₂O)₅Cl was formed. When they met the surface active agent in the water, the metal ion chelate may be formed, which can accelerate the dissolution of organic matter [14]. At present, there are few studies about the effect of adding Fe²⁺, so the
concentration research of Fe\textsuperscript{2+} had innovative.

2.2.3. Experiment design scheme. Choose L18 (3\textsuperscript{7}) orthogonal table which used orthogonal scientific to arrange experimental scheme with multi-level and multi-factor as a design table [15]. The suitable combination of various factors was found out through the analysis and treatment of the experimental results [16].

In addition to choose the above three kinds of additives, we choose the antibiotic concentration, temperature, pH, spray volume to explore the main factors that affect the efficiency of removal. Xu Lei’s study discovered that when degrading toluene’s concentration lower than 100 mg·m\textsuperscript{-3}, fungus was about 20% higher than bacteria [17]. Add antibiotics to inhibit the growth of bacteria could benefit the growing of fungi. Suitable temperature, pH, spray volume could improve the efficiency of microbial degradation as well. Through the combination of various factors at different levels, we can find out the group with highest removal efficiency as the best combination.

The experimental designed with this conditions: the inlet concentration of toluene was 1000 mg·m\textsuperscript{-3}, nitrogen source concentration was 2–6 g·L\textsuperscript{-1}, antibiotic concentration was 0–600 U·mL\textsuperscript{-1}, Triton X-100 concentration was 0.02–0.08 mL·L\textsuperscript{-1}, Fe\textsuperscript{2+} concentration was 8–20 mL·L\textsuperscript{-1}, pH was 5.5–8.5, temperature was 29–39 ℃, spray rate was 2–10 L·h\textsuperscript{-1}. The air intake method was countercurrent. The experimental scheme was designed according to the table 1.

| Group | A   | B    | C   | D   | E   | F   | G   |
|-------|-----|------|-----|-----|-----|-----|-----|
| 1     | 2   | 0    | 0.02| 8   | 5.5 | 29  | 2   |
| 2     | 2   | 0    | 0.02| 8   | 5.5 | 29  | 2   |
| 3     | 2   | 600  | 0.08| 20  | 8.5 | 39  | 10  |
| 4     | 4   | 0    | 0.02| 14  | 7   | 39  | 10  |
| 5     | 4   | 300  | 0.05| 20  | 8.5 | 29  | 2   |
| 6     | 4   | 600  | 0.08| 8   | 5.5 | 34  | 6   |
| 7     | 6   | 0    | 0.05| 8   | 8.5 | 34  | 10  |
| 8     | 6   | 300  | 0.08| 14  | 5.5 | 39  | 2   |
| 9     | 6   | 600  | 0.02| 20  | 7   | 29  | 6   |
| 10    | 2   | 0    | 0.08| 20  | 7   | 34  | 2   |
| 11    | 2   | 300  | 0.02| 8   | 8.5 | 39  | 6   |
| 12    | 2   | 600  | 0.05| 14  | 5.5 | 29  | 10  |
| 13    | 4   | 0    | 0.05| 20  | 5.5 | 39  | 6   |
| 14    | 4   | 300  | 0.08| 8   | 7   | 29  | 10  |
| 15    | 4   | 600  | 0.02| 14  | 8.5 | 34  | 2   |
| 16    | 6   | 0    | 0.08| 14  | 8.5 | 29  | 6   |
| 17    | 6   | 300  | 0.02| 20  | 5.5 | 34  | 10  |
| 18    | 6   | 600  | 0.05| 8   | 7   | 39  | 2   |

A—Nitrogen/g·L\textsuperscript{-1}  
B—Antibiotic concentration/U·mL\textsuperscript{-1}  
C—Surface active agent/mL·L\textsuperscript{-1}  
D—Fe\textsuperscript{2+}/mL·L\textsuperscript{-1}  
E—pH  
F—Temperature/℃  
G—Spray volume/L·h\textsuperscript{-1}
3. Experimental results and analysis

The text of your paper should be formatted as follows:

The experimental scheme was designed according to the table 1; the results obtained were shown in Figure 1 and table 2.

From the Figure 1, in the design of orthogonal experiment, the removal efficiencies of the second and the eighth groups were 89% and 87%. Therefore, the conclusion was as follows.

Condition 1: nitrogen source concentration was 2g·L⁻¹, antibiotic concentration was 300 U·mL⁻¹, Triton X-100 concentration was 0.05mL·L⁻¹, Fe²⁺ concentration was 14 mL·L⁻¹, pH was 7.0, temperature was 34℃, spray rate was 6 L·h⁻¹.

Condition 2: nitrogen source concentration was 6g·L⁻¹, antibiotic concentration was 300 U·mL⁻¹, Triton X-100 concentration was 0.08mL·L⁻¹, Fe²⁺ concentration was 14 mL·L⁻¹, pH was 5.5, temperature was 39℃, spray rate was 2 L·h⁻¹.

| Group | Removal rate/% | Group | Removal rate/% |
|-------|----------------|-------|----------------|
| 1     | 73.60          | 10    | 71.59          |
| 2     | 88.59          | 11    | 78.23          |
| 3     | 83.71          | 12    | 66.64          |
| 4     | 74.20          | 13    | 49.26          |
| 5     | 76.73          | 14    | 69.41          |
| 6     | 65.37          | 15    | 70.77          |
| 7     | 56.28          | 16    | 74.99          |
| 8     | 87.27          | 17    | 65.31          |
| 9     | 64.58          | 18    | 74.72          |

Through the date analysis the results of the removal rates in table 2, it can be concluded that the range of each factor called R. The bigger the range of substances, the bigger the effect for toluene removing, on the contrary, the smaller the range of substance, the smaller the effect for toluene removing[18].

Methods of computation: the test date obtained from the first group were added together and average the results, named the summation as $K_i$, the average as $k_i = \frac{K_i}{6}$, $R_i$ was the range of the i factor. The calculated mode is:

$$R_i = \max [k_{i1}, k_{i2}, k_{i3}] - \min [k_{i1}, k_{i2}, k_{i3}]$$  \hspace{1cm} (1)

Take the nitrogen source as an example, after into the above formula, we can draw a conclusion:

$$K_{A1} = y_1 + y_2 + y_3 + y_{10} + y_{11} + y_{12}$$
\[ k_{A_1} = \frac{k_{A_1}}{6} = 0.771 \] (2)

In the same ways, \( k_{A_2} = 0.676, k_{A_3} = 0.705 \)

\[ R_A = \max[k_{A_1}, k_{A_2}, k_{A_3}] - \min[k_{A_1}, k_{A_2}, k_{A_3}] \]

\[ = \max[0.771, 0.676, 0.705] - \min[0.771, 0.676, 0.705] = 0.095 \] (3)

The calculation results are shown in Table 3.

### Table 3. Experimental data and calculation analysis table

| Number | A   | B   | C   | D   | E   | F   | G   |
|--------|-----|-----|-----|-----|-----|-----|-----|
| k_1    | 0.771 | 0.667 | 0.711 | 0.696 | 0.679 | 0.710 | 0.758 |
| k_2    | 0.676 | 0.776 | 0.687 | 0.771 | 0.738 | 0.697 | 0.702 |
| k_3    | 0.705 | 0.710 | 0.754 | 0.685 | 0.735 | 0.746 | 0.693 |
| R      | 0.095 | 0.109 | 0.067 | 0.086 | 0.059 | 0.049 | 0.065 |

A—Nitrogen/g•L-1  
B—Antibiotic concentration/U•mL-1  
C—Surface active agent/mL•L-1  
D—Fe^{2+}/mL•L-1  
E—pH  
F—Temperature/℃  
G—Spray volume/L•h-1

According to the range R from table, we could identify the primary and secondary order of the factors affecting the removal efficiency of toluene:  

B-A-D-C-G-E-F

The order of influencing factors on removal efficiency of toluene is: antibiotic concentration > nitrogen > Fe^{2+} > surface active agent > spray volume > pH > temperature.

The concentration of toluene in each layer of the outlet was detected through the sampling port, and then we could obtain the concentration of toluene in the optimal combination and the average state of all combinations. The concentration of toluene in each layer of the filter is shown in Figure 2.

Removal efficiency = \( \frac{\text{inlet concentration} - \text{outlet concentration}}{\text{inlet concentration}} \), the removal of toluene in the unit time of tower was called removal rate. Removal quantity was the main factor affecting the removal efficiency because the structure and size of each layer were the same and the gas in each layer had the same residence time. The removal quantity and removal rate of each layer of the tower are shown in Table 3.

![Figure 2. Concentration change of each layer in the biological filter tower](image-url)
a—average combined polynomial
b—optimal combined polynomial

Two regression curves could be obtained by fitting the curve in Figure 2. Using regression calculation, regression curve equation for the average concentration of all combinations and the number of layer of the filter tower could be obtained as:

\[ y = 20.008x^2 - 361.64x + 1377.7 \]  \hspace{1cm} (4)

The curve equation between the optimal combination concentration and the number of layers is:

\[ y = 41.849x^2 - 477.68x + 1457.5 \]  \hspace{1cm} (5)

**Table 4. Removal rate and removal rate of each layer**

| Name Number | Average removal | Optimal combined removal | Average removal rate/% | Optimal combination removal rate/% |
|-------------|-----------------|--------------------------|------------------------|-----------------------------------|
| 1           | 259.07          | 302.46                   | 24.89                  | 29.97                             |
| 2           | 236.69          | 343.39                   | 30.28                  | 48.58                             |
| 3           | 149.50          | 134.16                   | 27.43                  | 36.92                             |
| 4           | 99.62           | 114.13                   | 25.1                   | 49.79                             |

The fitting coefficient of regression curve equation R² were 0.9994 and 0.9942 respectively which were close to 1. Conclusions can be drawn from Table 4. Along with the microorganisms remove the toluene from waste gas; the toluene partial pressure in the exhaust gas was getting lower and lower, so as it was in the liquid phase, as the result, removal quantity of toluene decreased with the reaction. It was believed that this is due to the resistance of hydration film in the process of toluene in gas-liquid mass transfer. The average removal rate of each layer was relatively stable.

There was no significant change in the oxygen content of each layer, so it could be determined that oxygen will not be the limiting factor for the removal of toluene by microorganisms.

4. Conclusions

1) In the design of orthogonal experiment, the removal efficiency was significantly different among the combinations, the removal efficiencies of the second and the eighth groups were 89% and 87%. The optimal condition was: nitrogen source concentration was 2g•L⁻¹, antibiotic concentration was 300 U•mL⁻¹, Triton X-100 concentration was 0.05mL•L⁻¹, Fe²⁺ concentration was 14 mL•L⁻¹, pH was 7.0, temperature was 34°C, spray rate was 6 L•h⁻¹.

2) According to the range R from table, we could identify the primary and secondary order of the factors affecting the removal efficiency of toluene: antibiotic concentration> nitrogen>Fe²⁺> surface active agent> spray volume> pH> temperature. The effect of antibiotic concentration on the removal efficiency was largest.

3) Fitting curve of the concentration of toluene in the optimal combination and the average state of all combinations was obtained, removal quantity of toluene decreased with the increase of the height of the layer. There was no significant change in the oxygen content of each layer, so it could be determined that oxygen will not be the limiting factor for the removal of toluene by microorganisms.

4) When the inlet toluene concentration was 1000 mg•m⁻³, the outlet concentration was about 400 mg•m⁻³, which had not reached the National emission standard (below 60 mg•m⁻³). Therefore, it can be connected to two stage treatment equipment in the treatment of high concentration of waste gas in order to meet the requirements of emission standards.
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