

Coupled Biotreatment of Waste Gas Containing H₂S and VOCs in a Compound Biofilter

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Abstract. A pilot-scale compound biofilter was installed in a pharmaceutical factory to purify waste gas containing H₂S and VOCs. The results confirmed that compound biofilter had the both advantages of biotrickling filter (BTF) and biofilter (BF), with high efficiency, energy conservation and low material-consumption in dealing with mixture waste gas containing H₂S and VOCs. Under the conditions of empty bed residence time(EBRT) at 18 s, temperature 30°C, pH 2~4, average H₂S removal efficiency reached above 97%, with removal load of 71.2 g/m³·h, which was mainly degraded by BTF, the lower layer of compound biofilter. Meanwhile, average VOCs removal efficiency reached above 80%, with removal load of 94.3 g/m³·h, which was mainly degraded by BF, the upper layer of the system. Thus it was found that EBRT had a more obvious effect on VOCs removal efficiency than that on H₂S.

Keywords: Hydrogen sulphide; Volatile organic compounds; Coupled biotreatment; Compound biofilter.

1. Introduction

The odor gas including hydrogen sulfide (H₂S) and volatile organic compounds (VOCs) and other toxic and harmful ingredients, discharged from the urban sewage treatment plants and pharmaceutical factory sewage station, which seriously polluted the surrounding environment. H₂S was a colorless, highly toxic and highly toxic gas, which had great harm to human body even at low concentration. Most of VOCs was carcinogens, which led to photochemical smog and serious air pollution. Biological purification technology of waste gas was the research focus and hotspot in the air pollution control field. The pollutants was degraded into carbon dioxide, water, sulfate radical and other non-pollution through microorganism metabolism, then generated new microbial cells[1,2,3]. Comparing to absorption, adsorption, catalytic combustion, neutralization and oxidation, biological method had some advantages of good removal efficiency, stable operation, low operation cost, and no secondary pollution. It was also especially suitable for processing the exhaust gas with large flow and low concentration [4, 5, 6].

At present, the domestic and international studies on biotrickling filter (BTF) or biofilter (BF) purifying H₂S or VOCs were more and more applied in the exhaust gas biotreatment industry. Degradation of H₂S and VOCs using BTF, BF or both series process in two devices were reported [7, 8, 9]. Treatment of urban sewage station mixed exhaust gas containing 200 mg/m³ H₂S and 2200 mg/m³ VOCs was researched by Cox [10, 11, 12]. When EBRT was 36 s, the highest removal loads were 20.0 g/m³·h and 70.0 g/m³·h, respectively. The experiment of purify the VOCs and H₂S using a biofilter was



researched by Converse [13]. When the H_2S inlet load was $8.3 \text{ g/m}^3 \cdot \text{h}$ under EBRT 15 s and the VOCs inlet load was $33 \text{ g/m}^3 \cdot \text{h}$ under EBRT 60 s, the pollutants can almost be removed completely. However, the related researches were few reported in china.

The coupled biotreatment of waste gas containing H_2S and VOCs from a pharmaceutical factory in a compound biofilter will be studied, which include the biotrickling filter and biofilter in a single experimental installation. In this paper, the removal efficiency, running performance, removal load and microbial community structure will be researched in this new compound biofilter.

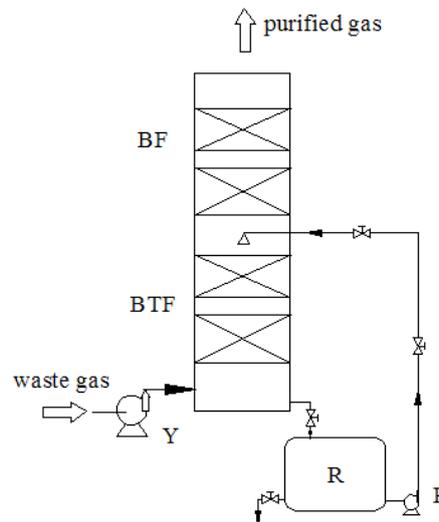
2. Materials and Methods

2.1. Waste Gas Source

The source of the experimental waste gas was collected from the comprehensive regulation pool and the anaerobic pool in a pharmaceutical factory. The main component of the waste gas was H_2S and VOCs, with the concentration of H_2S 191.0~891.5 mg/m^3 and VOCs 265.5~945.0 mg/m^3 .

2.2. Experimental Installation

The experimental installation was a new compound biofilter; the process flow was shown in figure 1.



Y- blower; R-Circulating tank; P-Circulating pump; biofilter(BF) ;biotrickling filter(BTF)

Figure 1. Schematic of compound biofilter

The main structure of compound biofilter was made of organic glass column, which was divided into upper and lower two layers. The inertness packing and biological packing were filled in the lower layer and the upper layer, respectively. The nutrient solution was supplied with continuous spray in the lower layer and carried moisture into the upper layer for microorganisms' growth. The installation was operated by gas-liquid countercurrent operation. Nutrient solution was sprayed by a pump from the circulating tank to the middle part of the device, and returned to the bottom and the circulating tank. The waste gas was entered from the bottom and discharged from the top of the tower. The pollutants in the waste gas were degraded by biomembrane on the packing surface. Spray intensity, circulation liquid level and pH were controlled by the PLC automatic system. Design and operation parameters of compound biofilter were shown in table 1.

Table 1. Design and operation parameters of compound biofilter

Design parameters	The upper layer	The lower layer
Tower size (internal diameter × height) / mm	380×1150	
Packing type	Biological packing (natural plant fiber)	Inertness packing (Porous polymer)
Packing height / mm	342	320
pH	No control	PLC automatic control
Spray water / (m ³ ·m ⁻² ·d ⁻¹)	No spray	4.41(continuous spray)

2.3. Analysis Method

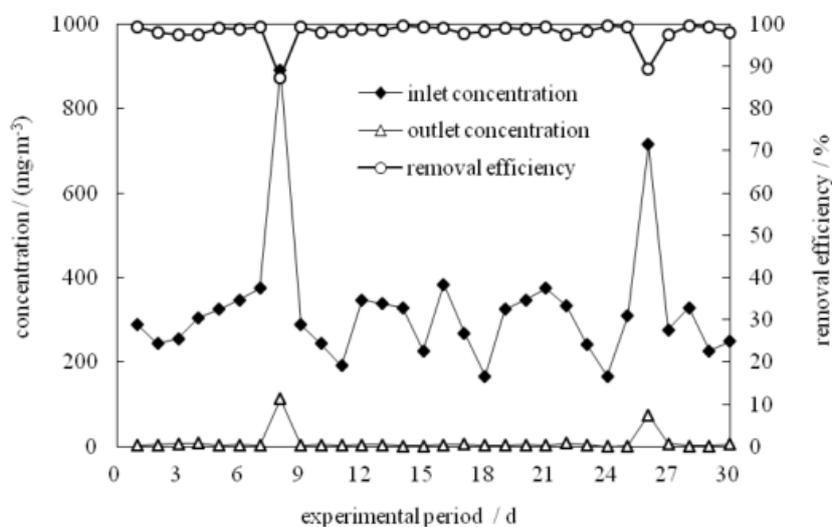
H₂S concentration was mensurated by iodometric method when H₂S concentration was higher than 10 mg/m³, and mensurated by methylthionine chloride spectrophotometric method when H₂S concentration was less than 10 mg/m³. The VOCs concentration was mensurated by VOCs portable analyzer (Huarui science instruments company, MiniRAE 2000), pH was mensurated by pH/ORP analyzer (HACH science instruments company, p53).

The biofilms were extracted and cultured from the packing of the upper and lower layer, then the biofilms dyed with crystal violet, observed the morphology, size and characteristics by microscope, and the bacterial colony was counted.

3. Results and Discussion

3.1. Startup and Operation of Compound Biofilter

The startup and stable running study of the compound biofilter was operated for 30 days. Experimental operating conditions as follows: the empty bed residence time (EBRT) was 18 s, pH of circulating fluid was 2~4, waste gas flow was 15m³/h, temperature of circulating liquid was 30°C, and one fifth circulating fluid was replaced every day. The ingredients of nutrient solution as follows: K₂HPO₄ 100 mg/L, KH₂PO₄ 100 mg/L, MgSO₄ 20 mg/L, CaCl₂ 10 mg/L, NH₃NO₃ 600 mg/L, MnSO₄ 5 mg/L, CuSO₄ 2 mg/L, FeSO₄ 2 mg/L. The operation of compound biofilter were shown in Figure 2~3.

**Figure 2.** H₂S removal efficiency change in compound biofilter

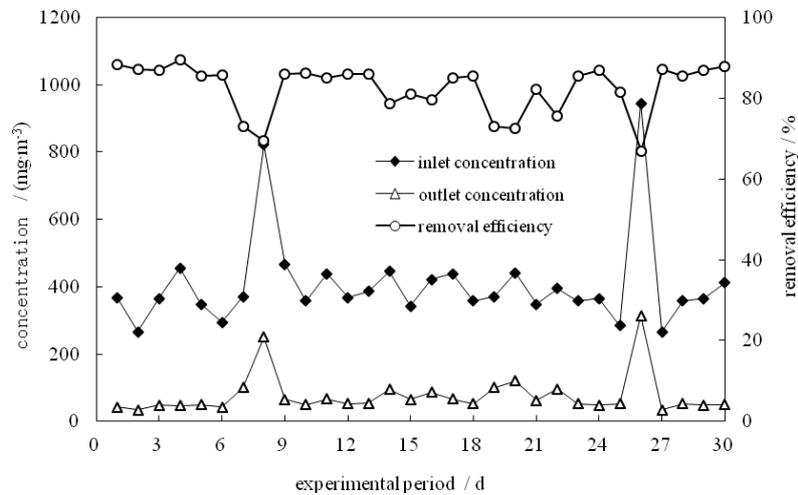


Figure 3. VOCs removal efficiency change in compound biofilter

H₂S and VOCs removal efficiency changes in compound biofilter during startup and stable running were displayed in figure 2~3. It was found that the startup period of the compound biofilter was only 7 days. The H₂S average inlet concentration was 332.9 mg/m³ with removal efficiency above 97%, and the VOCs average inlet concentration was 406 mg/m³ with removal efficiency above 80%. The highest concentration of H₂S was 891.5 mg/m³ during this period, and removal efficiency fell to 87.3%, because of the production process changed in the workshop of pharmaceutical factory. At the same time, the highest concentration of VOCs was 945 mg/m³, and removal efficiency fell to 66.8%. However it happened, H₂S and VOCs removal efficiency returned to normal, when the concentration returned to normal range in the next day. Obviously, the compound biofilter had the ability to resist the impact of high concentration load, which was great significance in application of waste gas biotreatment.

3.2. Effect of Inlet Load on Removal Efficiency

In this paper, effect of inlet load (I_L) on removal efficiency (R_E) and removal load (R_L) was researched during the experimental period. The expressions as follows:

$$I_L = C_{g.in} \times Q/V \quad (\text{g/m}^3 \cdot \text{h}) \quad (1)$$

$$R_E = (C_{g.in} - C_{g.out}) / C_{g.in} \quad (2)$$

$$R_L = (C_{g.in} - C_{g.out}) \times Q/V \quad (\text{g/m}^3 \cdot \text{h}) \quad (3)$$

where $C_{g.in}$ = inlet concentration(g/m³), $C_{g.out}$ = outlet concentration(g/m³), Q = waste gas flow(m³/h), V = packing volume(m³).

Effect of H₂S and VOCs inlet load on removal efficiency in compound biofilter were shown in Figure 4~5. As shown in Figure 4, The H₂S average removal efficiency was 96.5% and the average removal load was 71.2 g/m³·h, when the waste gas flow was 15.0 m³/h, temperature was 30°C and EBRT was 18 s. As shown in Figure 5, The VOCs average removal efficiency was 81.2% and the average removal load was 94.1 g/m³·h at the same experimental conditions.

As shown in Figure 4~5, The H₂S and VOCs removal load in compound biofilter increased linearly with inlet load increasing, and the H₂S and VOCs removal efficiency were more than 97% and 80% under the low inlet loads, respectively. The H₂S and VOCs removal efficiency gradually decreased and deviated from the linear under the high inlet loads.

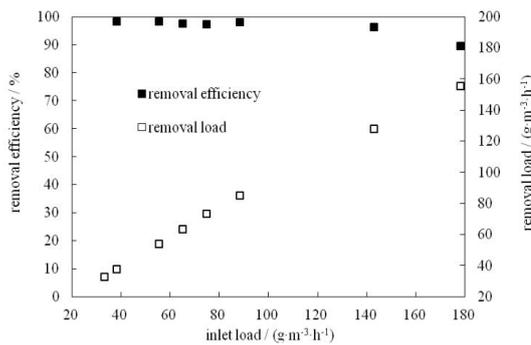


Figure 4.

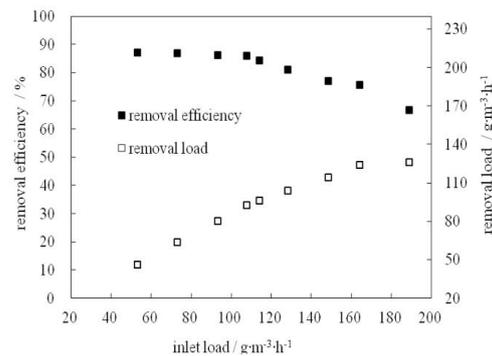


Figure 5

Figure 4. Effect of H₂S inlet load on removal efficiency in compound biofilter
Figure 5. Effect of VOCs inlet load on removal efficiency in compound biofilter

3.3. Study on the Layered Removal Efficiency and Load in Compound Biofilter

When studying on the layered removal efficiency, the experimental operating conditions was controlled as follow: pH 2~4, temperature 30°C, EBRT 18 s. H₂S and VOCs layered removal efficiency and layered removal load in compound biofilter were shown in Figure 6~7.

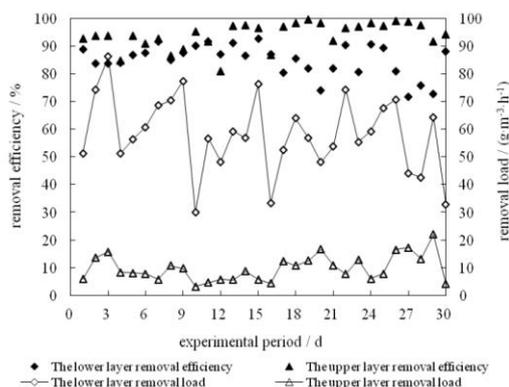


Figure 6.

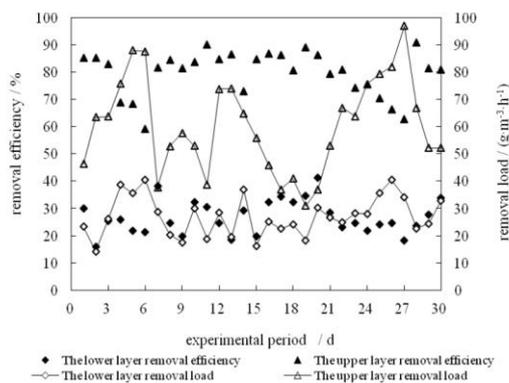


Figure 7

Figure 6. H₂S layered removal efficiency and load in compound biofilter
Figure 7. VOCs layered removal efficiency and load in compound biofilter

As shown in Figure 6, during the experimental period, the H₂S average removal efficiency of the lower layer was 85%, and the average removal load was 61.3 g/m³·h. Meanwhile, the H₂S average removal efficiency of the upper layer was 94%, though the average removal load was only 9.9 g/m³·h. It can be known that although the H₂S average removal efficiency of lower layer was lower than that of the upper layer, the H₂S average removal load of the lower layer was significantly higher because of the high inlet load of this layer. Therefore, it can see that most of the H₂S load was degraded by the BTF process.

As shown in Figure 7, during the experimental period, the VOCs average removal efficiency of the lower layer was 26.8%, and the average removal load was 27.6 g/m³·h. Meanwhile, the VOCs average removal efficiency of the upper layer was 79.8%, and the average removal load was 66.7 g/m³·h. It can be known that the VOCs average removal efficiency and load of the upper layer was both higher than that of the lower layer, although the inlet load in the lower layer was higher. Therefore, it can see that most of the VOCs load was degraded by the BF process.

In conclusion, the BTF process in compound biofilter was more suitable to purify the hydrophilic pollutants. The process of biodegradation can be explained by the theory of “absorb-biofilm”. The

dissolved load of H₂S was increased when the nutrient solution increasing, which was beneficial to purify. The BF process in compound biofilter was more suitable to purify hydrophobic pollutants, and the process of biodegradation can be explained by the theory of “adsorption-biofilm”. It was not beneficial to removal of VOCs while the amount of circulating fluid was too large, so it was no sprayed in the upper layer in the experimental period.

The compound biofilter had both advantages of BF and BTF, which were the focus to the different purification task. Therefore, it had advantages in purifying the mixed waste gas.

3.4. Bacterial Colony Count and Analysis

During the experimental period, the biofilm of the upper and lower layers in the compound biofilter was extracted and stained, and the bacterial colony was counted. Bacterial colony type and count were shown in table 2. Scanning electron microphotograph of biotic community on the packing were shown in Figure 8~9.

Table 2. Bacterial colony type and count CFU·g⁻¹

Bacterial colony type	The upper layer(BF)	The lower layer(BTF)
Bacteria	7.0×10 ⁵	9.2×10 ⁷
Actinomycetes	9.9×10 ⁶	1.1×10 ⁵
Fungi	4.4×10 ⁶	6.0×10 ⁵

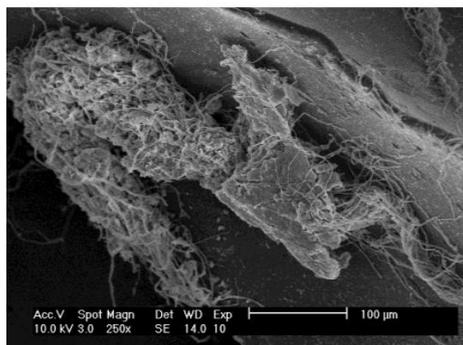


Figure 8.

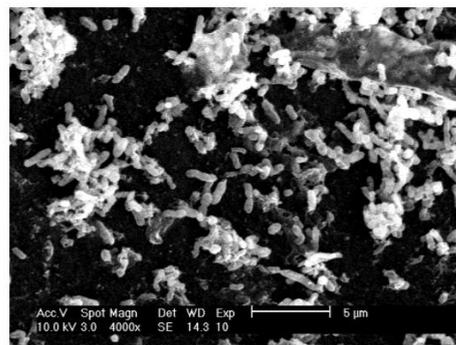


Figure 9

Figure 8. Scanning elctron micro photograph of biotic community on upper packing

Figure 9. Scanning elctron micro photograph of biotic community on lower packing

The results showed that the bacterial colony type containing bacteria, actinomycetes and fungi in the upper and lower layers were almost the same. It was mainly filamentous bacteria in the surface of the upper layer, but was mainly bacillus in the surface of the lower layer.

4. Conclusion

(1)At the stable operation period, H₂S and VOCs average removal efficiency were more than 97% and 80% , meanwhile, the average removal load was 71.2 g/m³·h and 94.3 g/m³·h , respectively.

(2)During the experimental period, the H₂S average removal load was 61.3 g/m³·h in the lower layer, though the average removal load was 9.9 g/m³·h in the upper layer. Most of the H₂S load was degraded by the BTF process. The VOCs average removal load was 27.6 g/m³·h in lower layer. Meanwhile, the VOCs average load was 66.7 g/m³·h in the upper layer. Most of the VOCs load was degraded by the BF process. The compound biofilter had both advantages of BF and BTF.

(3)The bacterial colony type contained bacteria, actinomycetes and fungi in the compound biofilter. It was mainly filamentous bacteria in the surface of the upper layer, but was mainly bacillus in the surface of the lower layer.

5. Acknowledgements

This work was supported by the Science and technology project of Zhejiang province (2017C37071), the open fund of priority among priorities of biological engineering in Zhejiang province (KF2017004), Zhejiang college students' science and technology innovation activity plan (2017R420009), and the Science and Technology project of Ningbo city (2015C50002).

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