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Research article

The performance of a two-layer biotrickling filter filled with new mixed packing materials for the removal of H_2S from air

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ABSTRACT

In the work described here, a two-layer biotrickling filter filled with new packing materials was used to remove H_2S from air. The upper layer of the filter was packed with activated carbon-loaded polyurethane, whereas the lower layer was filled with modified organism-suspended fillers. The effects of inlet load, empty bed residence time (EBRT) from 79 s to 53 s, pH and contaminant starvation time were investigated. For loads of 15–50 g/(m^3 h), the average removal efficiency (RE) was higher than 96% under a consistent supply of pollutants. The critical elimination capacity was 39.95 g/(m^3 h) for an EBRT of 53 s with an RE of 99.9%. The two-layer BTF was capable of withstanding contaminant starvation periods for 1.5 d and 7 d with only a few hours of recovery time. The biodegradation kinetics was studied using Michaelis–Menten type equations under different EBRTs. At an EBRT of 66 s, the optimal kinetic constants r_{max} and K_m were 333.3 g/(m^3 h) and 0.93 g/m^3, respectively. During the operation, the two-layer BTF performed well under various reasonable conditions.

1. Introduction

The influence of air pollution has become a major issue and a nuisance to the communities surrounding the sources of pollutant emissions. Some pollutants are odorous, and of these compounds, hydrogen sulfide (H_2S) is one of the most common. H_2S is a colorless gas found in the off-gas produced by textile manufacturing, waste management, dye production, heavy water production and petroleum refining, and it can be removed primarily by chemical and physical practices. H_2S typically smells of rotten eggs and has an odor threshold of 0.5 ppb to 0.3 ppm. It can lead to death at concentrations of over 500 ppm due to its broad spectrum toxicity (Busca and Pistarino, 2003). In recent decades, biological methods have been incorporated into the field of H_2S research. Among the available biotechnologies, which include biofilters, biorubbers and biotrickling filters, the biotrickling filter (BTF) is the most suitable for controlling H_2S. Although biological treatments are ecologically and environmentally favorable, they are still insufficient compared with chemical methods. Though chemical scrubbing is the most well-established technology for H_2S removal in Waste Water Treatment Plants (WWTPs), this method consumes many chemicals, which increase both the operating costs and the carbon footprint of the treatment (Mannucci et al., 2012).

By contrast, a biotrickling filter can remove H_2S under various operating conditions without the use of so many chemicals. The main advantages of BTFs are their ease of control, low costs and high removal efficiency (RE). Though there is abundant literature on the process modeling and biomass accumulation of BTFs used in the removal of volatile organic compounds (VOCs) (Devinny and Ramesh, 2005; Yang et al., 2010), further study is needed regarding the removal of H_2S. Several studies have demonstrated that BTFs can run normally even under extreme conditions. Kim et al. (Kim et al., 2005) proved that trickle-bed air biofilters could be used under periodic backwashing and cyclical nonuse periods to treat styrene-containing air at a styrene load of 0.64–3.17 kg chemical oxygen demand (COD)/(m^3 day). Moreover, Namini et al. (Namini et al., 2012) evaluated the performance of a bio-trickling filter employing *thiobacillus thiparus* immobilized on polyurethane foam under various starvation regimens, showing that the BTF was subjected to various starvations such as non-contaminant loading, idleness, and complete shutdown.

The selection of packing materials is of great importance in BTFs.

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Fine packing materials usually present high REs and better stability over the long run. Due to the importance of the packing materials, many types of materials have already been studied, including molecular sieves, polyurethane foam, sugarcane bagasse, coconut fiber, activated carbon, ceramic rings and compost (He et al., 2009; Filho et al., 2010; Dorado et al., 2010). In addition, some researchers have developed a number of new packing materials. Dumont et al. (Dumont et al., 2008) compared a new packing material called UP20, which contains CH_4N_2O, H_3PO_4, $CaCO_3$ (C/N/P molar ratio: 100/5/1) and an organic binder, with pine bark for the removal of H_2S, showing that UP20 could be effective in treating large concentrations of pollutants. Packing materials with larger specific areas and longer life spans are more suitable, as they have better physical and chemical stabilities.

Current research is focused on using BTFs with lower residence times to treat mixed gasses. Hernández et al. (Hernández et al., 2013) tested the startup and long-term performance of two biotrickling filters packed with polyurethane foam and polar wood chips in the treatment of a mixture of ethylmercaptan, H_2S and NH_3. The result indicated that both filters could completely remove NH_3 in the long run, while the H_2S removal efficiencies exceeded 90%. The removal of ethylmercaptan differed obviously between the two BTFs. In terms of residence time, Gabriel et al. (Gabriel and Deshusses, 2003) investigated the performance of a full-scale biotrickling filter in treating H_2S over a gas contact time period of 1.6–2.2 s. Under continuous operation over one year, the BTF showed stable performance and high removal efficiency, although the inlet H_2S concentration was relatively low (30–60 ppm). The results of that study proved the feasibility of using BTFs to remove low concentrations of H_2S over short periods of gas contact time.

The objective of the present study was to investigate the performance of treating H_2S-contaminated air streams with a two-layer biotrickling filter filled with new packing materials. These materials consisted of modified organism-suspended fillers and activated carbon-loaded polyurethane inoculated with activated sludge. The empty bed residence time (EBRT) was also controlled on the basis of an actual situation. Finally, the pilot scale BTF was shown to run normally and maintain a high RE.

2. Materials and methods

2.1. Microorganism and cultivation medium

The microorganisms used in the present study were obtained from activated sludge collected from the secondary sedimentation tank in the sewage treatment works of Sinopec Yangzi Petrochemical Company, China. First, some of the microorganisms obtained from the supernatant of the activated sludge were cultured in solid medium, which was then used to enrich the autotrophic sulfur-oxidizing microorganisms. The media (per liter) contained 0.4 g NH_4Cl, 10 g $Na_2S_2O_3$, 4 g KH_2PO_4, 4 g K_2HPO_4, 0.8 g $MgSO_4 \cdot 7H_2O$, 20 g agar powder, and 10 ml trace elements solution. The composition of the trace elements solution (per liter) was 250 mg Na_2-EDTA, 55 mg $ZnSO_4 \cdot 7H_2O$, 36.7 mg $CaCl_2 \cdot 2H_2O$, 12.5 mg $MnCl_2 \cdot 4H_2O$, 2.5 mg $CoCl_2 \cdot 6H_2O$, 2.5 mg $(NH_4)_6Mo_7O_{24} \cdot 4H_2O$, 2.5 mg $FeSO_4 \cdot 7H_2O$, and 1 mg $CuSO_4 \cdot 5H_2O$. The microorganisms grown on the solid medium were transferred to a new shake-flask containing the medium described above except without agar powder. Finally, the cultures and the activated sludge were mixed together.

2.2. Description of packing materials

In the two-layer BTF, the activated carbon-loaded polyurethane and modified organism-suspended fillers were packed into the upper and bottom layers, respectively. The modified organism-suspended filler prepared with base material of polythene had much better mechanical strength and weaker resistance than the polyurethane filler, which was beneficial for mass transfer. Consequently, it was packed into the bottom layer. The polyurethane loaded with activated carbon was porous, benefiting gas distribution and reasonably prolonging gas contact times. The total packing percentage was 36%. The basic characteristics of the modified organism-suspended filler are listed in Table 1. The modified organism-suspended filler was prepared as a microorganisms attaching carrier usually using in wastewater treatment, which would present suspended state in wastewater operation process. But here it was filled into fixed bed to immobilize bacteria for H_2S biodegradation.

2.3. Biotrickling filter system

A schematic of the two-layer BTF is presented in Fig. 1. The BTF was made up of a transparent Plexiglas column with an inner diameter of 250 mm and a total height of 1800 mm. The biotrickling filter system contained three zones: the spraying zone for nutrient feeding and discharge of the gas stream; the packing zone for loading the packing materials, including the 300 mm layer of activated carbon-loaded polyurethane and the 350 mm layer of the modified organism-suspended filler; and the gas distribution zone for distributing the influent gas streams and discharging the effluent. Perforated Plexiglas was placed at the base of the BTF to support the packing materials and to apportion the gas streams. Three gas sampling ports were used to detect the H_2S concentration in the spraying zone and the gas distributing zone.

Countercurrent air–water flow was used in the BTF. The gas entering the BTF was divided into two streams: the air was supplied by a compressor, whereas the H_2S was supplied by a H_2S cylinder. Before delivery to the BTF, the two streams were premixed in a mixing tank and went through a buffering tank to dilute the H_2S gas. The flow rate of the total streams was regulated by a gas flow meter. The gas streams were then delivered to the gas distribution zone and entered the packing zone, where the pollutant was absorbed and degraded by the microorganisms on the packing materials. After the treatment, the gas streams were delivered to the upper layer, which was the same as the bottom layer. Each layer was equipped with a nozzle in the spraying zone for adding nutrients. The performance of the system was estimated by calculating the elimination capacity (EC) of H_2S and the RE at different inlet loads according to the following equations: $EC = Q(C_{in} - C_{out})/V$ and $RE = 100\% (C_{in} - C_{out})/C_{in}$, where Q, C_{in}, C_{out}, and V are the gas stream inlet flow rate, inlet H_2S concentration, outlet H_2S concentration, and BTF volume, respectively (Duan et al., 2007).

2.4. Immobilization method

According to our findings, in the lab-scale test, it was found that bacteria accumulation time was longer than normal activated sludge culture, probably because H_2S degraded autotrophic bacteria were not so much in the original activated sludge. Therefore, in the present study, sulfur-oxidizing microorganisms were isolated from the original activated sludge and expanded before immobilization. In the bottom layer, the cultures and activated sludge were transferred to the BTF, and meanwhile sufficient amount of nutrients were added to immerse the packing materials. After aeration for approximately 20 h, the mixture was left to stand for several hours and then was discharged. As a result, microorganisms adhered to the surface of the packing material. The supernatant of the mixture was then separated and used for immobilization in the upper layer. The supernatant was sprayed circularly onto both layers as air was blown into the BTF at a low flow rate. To form the