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Study on hydrogen sulfide removal based on bench-scale experiment by bio-trickling filter*

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Abstract: A bench-scale experiment for control of hydrogen sulfide (H_2S) emissions was carried out continuously for nearly four months by using bio-trickling filter packed with ZX01 stuffing. The results suggested that the bio-trickling filter had proven excellent performance over substantial operational periods. Removal efficiency of H_2S was nearly 100% when volumetric loading of the bio-trickling filter varied from 0.64 g/(m^3·h) to 38.20 g/(m^3·h) and metabolism products of H_2S were mainly composed of SO_4^{2-}. When inlet concentration of H_2S was 250 mg/m^3, the optimum gas retention time was 30 s and the optimum spray water flow rate was 0.005 9~0.012 L/(cm^2·h). The bio-trickling filter had good ability to resist shock of high volumetric loading, and was not blocked during experiments for nearly four months during which resistance was maintained at relatively lower value, so that the bio-trickling filter need not carry out back washing frequently and can be operated steadily for long-term.

Key words: Bio-trickling filter, Hydrogen sulfide, Carrier, Deodorization
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INTRODUCTION

H_2S is a highly toxic air pollutant with very low odor threshold value. Considerable amounts of H_2S are produced and emitted in industrial processes, such as petroleum refining, rendering, wastewater treatment, paper and pulp manufacturing, food processing, and in the treatment of sour gas and other fuels. H_2S is frequently the main component of most observable odorous emissions.

The removal of malodorous reduced sulfur emissions has been traditionally accomplished using physical or chemical methods (Bouzaza *et al*., 2004; Meeyoo *et al*., 1998; Nishikawa and Takahara, 2001; Przepiórski *et al*., 1999). Chemical methods such as incineration, chlorination and combustion involve relatively complicated procedures, require the addition of chemicals or fuels, and involve considerable expense. Physical methods such as activated carbon adsorption and scrubbing are also costly and involve regeneration or transportation of hazardous wastes. These control technologies are usually uneconomical if large flow rates and low contaminant concentrations characterize the waste air streams. Mostly importantly, all the above mentioned methods generate secondary wastes that may require further treatment or disposal, thereby creating additional environmental problems. More and more, purification processes are based on the ability of some microorganisms to oxidize a variety of inorganic and organic compounds into mineral end-products because they have many advantages over physical and chemical methods. Besides their high removal efficiency, low investment,

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safe operating conditions and low energy consumption, they generate less secondary pollution (Shinabe *et al*., 2000; Park *et al*., 2002; Malhautier *et al*., 2003; Potivichayanon *et al*., 2006).

Biofilters have been used for approximately 50 years to treat odorous off-gases from wastewater treatment plants and other sources. In the early 1980s application of the technology to treatment of volatile organic compounds (VOCs) in industrial off-gases began (Ottengraf *et al*., 1986). It is currently a well established air pollution control technology. In the US, the first systematic research on biofiltration involved the treatment of odor resulting from hydrogen sulfide gases in sewage works. Studies on the efficiency of removing odor were conducted using a lab-scale biofiltration unit (Chung *et al*., 2005; Hilger *et al*., 2000; Lee *et al*., 2006; Mohseni and Allen, 2000; Syed and Henshaw, 2003; Morgan-Sagastume and Noyola, 2006). At the same time, researches on biological methods were also reported in China. For example, Yin *et al*.(2003) used a peat biofilter to remove H_2S with the removal efficiency being higher than 99% when the inlet concentration was 1.51~9.57 g/(m^3·h) and gas retention time was 30 s. Guo *et al*.(1999) developed a fixed-film filter to remove H_2S with removal efficiency being able to reach 100% when volumetric loading was lower than 23.33 g/(m^3·h). Wang *et al*.(2001) employed a peat biofilter to dispose of complex malodorous gas containing H_2S. Removal efficiency of H_2S was about 100% and could be kept for a long time at volumetric loading lower than 45 g/(m^3·h). Jiang *et al*.(2001) and Shao *et al*.(1999) also studied the setup and mechanism of biological deodorization. Bio-trickling filtration is newer than biofiltration with its bench-scale study and industrial application being still relatively less. Bio-trickling filters enable higher pollutant elimination rates to be obtained for a broader range of pollutants than biofilters. Bio-trickling filtration is especially well-suited for the control of relatively poorly biodegradable VOCs or pollutants such as chlorinated and reduced sulfur compounds (Cox and Deshusses, 1998) that release acids upon degradation.

A number of microbial processes for H_2S removal have been studied by using some special stuffing as carriers. In the bioreactors, the packing materials commonly used consist of peat, compost, soil, calcium alginate, polyethylene, glass, heather branches and bark chips. Activated carbon is also used as microorganism support for removing undesirable molecules from wastewater and gaseous effluents. However, these processes have neither satisfied removal efficiency nor desirable removal capacity, especially for removal of high concentration H_2S.

In this study, removal efficiency of the H_2S was investigated by using bio-trickling filter with ZX01 stuffing. The research focused on operational parameter, removal efficiency, ability to resist loading change, pressure drop and working life of the bio-trickling filter. The research results also laid foundation for optimal design and operation of the full scale bioreactor.

MATERIALS AND METHODS

Deodorization setup

The deodorization equipment provided by Shanghai Best Environmental Engineering Co. (China) has a volume of 2800 mm×800 mm×1650 mm with the whole system being automatically controlled (Fig.1). For example, frequency and flow rate of spray water are controlled by electromagnetism valve. The deodorization equipment is made up of Plexiglas. The most important part of the equipment is the biological tower packed with ZX01 stuffing layer with diameter and height of 0.3 m and 1.65 m, respectively. The ZX01 carrier is also made in Shanghai Best Environmental Engineering Co. (China) and consists of many round fiber balls with diameter of 30~35 mm and bulk density of 82 kg/m^3. The carrier was strongly resistant to acidification and erosion. The void fraction of the bioreactor is 0.70. In order to check the removal efficiency of H_2S at different tower heights, the tower is divided into three sections (A, B and C) with each section being equipped with a sampling port (H_A=270 mm, H_B=452 mm and H_C=340 mm). The odor containing H_2S is pumped to a buffer gas pot by air compressor and then is introduced into the biological tower from air distributor after flowing through the gas flow meter. The gases treated by biological tower exhaust from outlet port. Liquid in the storage water tank automatically flows to buffer water pot due to the gravity, then can be sprayed regularly to biological tower by centrifugal water pump from liquid distributor, and finally drains off from effluent

water port. The liquid is tap water added with nutrients or effluent wastewater from secondary sedimentation tank, either of which can supply nutrients and moisture for the growth of microorganisms.

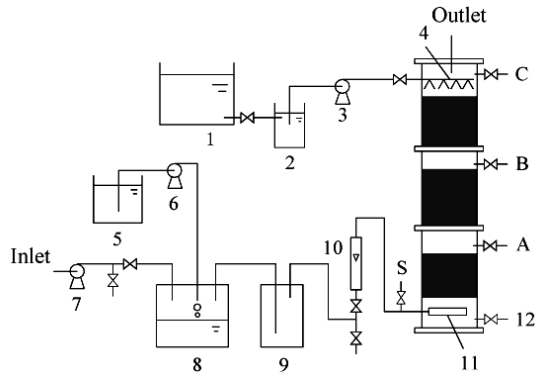


Fig.1 Schematic diagram of the biological deodorization process

1: Storage water tank; 2: Buffer water pot; 3: Water pump; 4: Liquid distributor; 5: Na_2S liquid pot; 6: Stoichiometric pump; 7: Air compressor; 8: H_2SO_4 pot; 9: Buffer gas pot; 10: Gas flow meter; 11: Air distributor; 12: Effluent water port; S, A, B, C: Sampling ports

The deodorization experiment was carried out continuously under ambient temperature and pressure condition.

Biofilm formation

In this study, ZX01 stuffing provided by Shanghai Best Environmental Engineering Co. (China) was randomly placed in the bio-trickling filter. The deodorization experiments were started after inoculation with domestic activated sludge from wastewater treatment plant, and H_2S concentrations were supplied with 5 mg/m^3 up to about 150 mg/m^3 after 15 d.

At the same time, fresh medium was continually added to the bio-trickling filter so that biofilm containing sulfur oxidizing bacteria was formed on the packing carrier surface after two weeks.

Analytical methods

H_2S concentration in the odor was measured by gas chromatography with a flame photometric detector (Hewlett Packard 6890, USA, 3 m×3 mm i.d. column was packed with ODTN 25%). Sulfate concentration was measured by ion chromatograph (Dionex 4500i, USA). The separating column was an inorganic negative-ion column of HPIC-AG4AAS4A-SC. The mobile phase was composed of 1.8 mmol/L Na_2CO_3 and 1.7 mmol/L $NaHCO_3$. Both total N and total P were respectively measured by potassium persulfate oxidation-UV spectrophotometric method and potassium persulfate digestion-colorimetry of stannous chloride reduction. Pressure drop was measured by a water manometer with minimum division length reading of 1-mm water column.

RESULTS AND DISCUSSIONS

Startup of the bio-trickling filter

Deodorization experiments were started after inoculation and H_2S was flown through the bioreactor at 5 m^3/h of gas flow rate and 5 L/h of spray water flow rate. At the same time, fresh medium was continually added to the bio-trickling filter. The fresh medium consisted of (g/L): KH_2PO_4, 2; $MgCl_2{\cdot}6H_2O$, 0.2; K_2HPO_4, 2; NH_4Cl, 0.4; $FeSO_4{\cdot}7H_2O$, 0.01 and glucose, 9.

During the startup period, inlet H_2S concentration was enhanced gradually at lower than 5 mg/m^3 of outlet concentration. Fig.2 shows the operation results of the bioreactor during the startup phase. The bioreactor was successfully started when inlet concentration was increased to 150 mg/m^3 from 5 mg/m^3 and biofilm containing sulfur-oxidizing bacteria was obviously formed on the stuffing surface after about two weeks. Fig.3 shows the carrier shape before and after the biofilm formation, which also suggested the bioreactor was successful started.

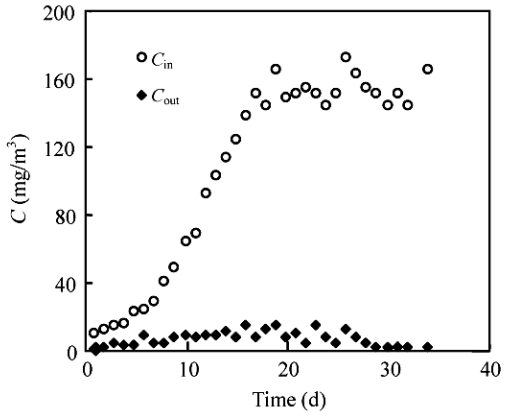
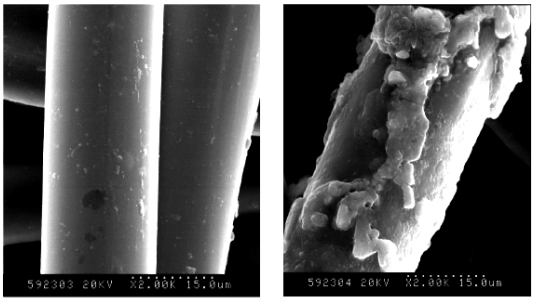


Fig.2 Operation results of the bio-trickling filter during the startup phase

C_{in}: Inlet concentration; C_{out}: Outlet concentration



(a) (b)

Fig.3 Micrograph of scanning electronic microscope of packing carrier before (a) and after (b) the biofilm formation

Effect of volumetric loading on the removal efficiency of H_2S

The effect of volumetric loading on the removal efficiency of H_2S was investigated at spray water flow rate of 5 L/h. Fig.4 shows the removal efficiency and outlet concentration of H_2S with the change of volumetric loadings.

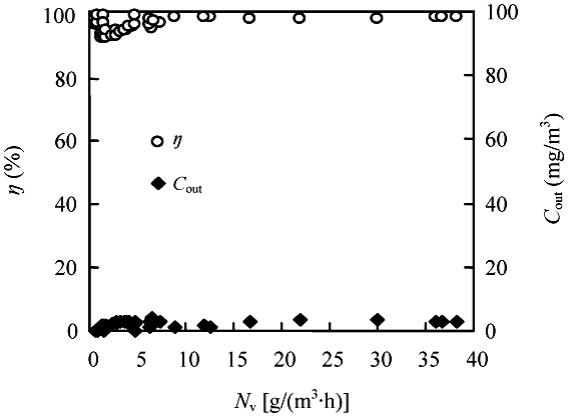


Fig.4 Effect of volumetric loading (N_v) on removal efficiency (η) and outlet concentration (C_{out}) of H_2S

Outlet concentration of H_2S was lower than 5 mg/m^3 when volumetric loading varied from 0.64 to 38.20 g/(m^3·h) and removal efficiency of H_2S was nearly 100%, especially under high loading condition. When volumetric loading was relatively lower, microorganisms were in starvation state and had not achieved optimum digestion or absorption capacity so that the elimination capacity of the bio-trickling filter increased gradually with increasing volumetric loading.

To further investigate the effect of higher volumetric loading on removal efficiency of H_2S, volumetric loading of Section A and removal efficiency of Section A were evaluated when removal efficiency of the bioreactor was nearly 100% and inlet gas flow rate was 6 m^3/h. Table 1 shows removal efficiency and volumetric loading of Section A with the change of inlet concentration.

Table 1 Effect of volumetric loading of Section A on removal efficiency of H_2S

Concentration (mg/m^3)		η_A (%)	$N_{v,A}$ (g/(m^3·h))	N_v (g/(m^3·h))
C_{in}	$C_{out,A}$			
54.89	0	100	17.25	4.39
68.61	0	100	21.54	5.48
82.34	0	100	25.90	6.59
109.78	0	100	34.50	8.78
137.23	0.14	99.90	43.11	10.97
164.68	1.24	99.24	51.75	13.17
192.12	1.81	99.06	60.40	15.37
219.57	4.32	98.08	69.05	17.57
247.01	10.98	95.56	77.65	19.76
274.46	16.47	94.00	86.30	21.96
301.91	28.82	90.45	94.90	24.15
329.35	35.68	89.17	103.55	26.35

C_{in}: Inlet concentration; C_{out}: Outlet concentration; η: Removal efficiency; N_v: Volumetric loading

Removal efficiency of Section A decreased gradually and outlet concentration increased gradually with increasing volumetric loading. Removal efficiency of Section A was higher than 90% when volumetric loading of Section A was lower than 94.90 g/(m^3·h). Otherwise, when volumetric loading of Section A was lower than 43.11 g/(m^3·h), removal efficiency of Section A could reach 100% and Section B and Section C microorganisms were nearly in starvation state and did not remove H_2S gas. Volumetric loading of Section A was about 3.93 times that of the volumetric loading of the bio-trickling filter so that the bioreactor had enormous potential to increase volumetric loading and elimination capacity of Section B and Section C.

Table 1 also shows the effect of the bio-trickling filter height on the removal efficiency of H_2S. Even though Section A occupied only 22.64% of the whole tower height, it had nearly 90% of removal efficiency when inlet volumetric loading was lower than 26.35 g/(m^3·h).

Metabolism products of microorganisms

Sulfur-oxidizing bacteria could utilize O_2 as electron acceptor and oxidize H_2S to sulfuric acid. Microorganisms could obtain energy during the transfer of H_2S and synthesize the constituent parts of cytoplasm. Oxidation and decomposition processes of H_2S by microorganism are shown as follows:

$$2H_2S+O_2 \rightarrow 2S+ 2H_2O+\text{energy}, \quad (1)$$

$$2S+3O_2+2H_2O \rightarrow 2H_2SO_4+\text{energy}, \quad (2)$$

$$CO_2+H_2O+\text{energy} \rightarrow [CH_2O]+O_2. \quad (3)$$

SO_4^{2-} concentration was continuously detected in the effluent spray water under fluctuating inlet H_2S volumetric loading condition. Fig.5 shows that with the gradual increase of volumetric loading, SO_4^{2-} concentration in the effluent spray water increases. Few yellow solids (generally considered element sulfur) were observed on the carrier surface, which suggested that metabolism products of H_2S were mainly composed of SO_4^{2-} and thus reduced the occurrence of bioreactor jam.

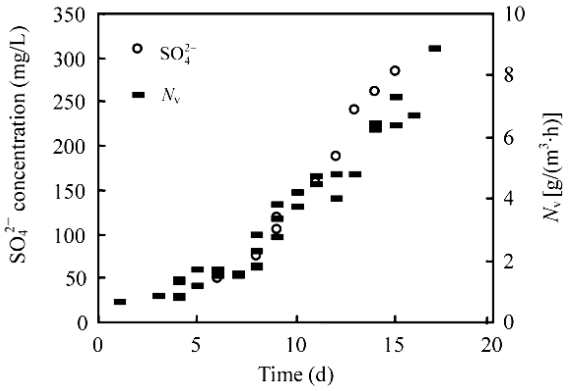


Fig.5 Effect of volumetric loading (N_v) on sulfate concentration

Fig.6 shows the value of pH and removal efficiency with the increase of volumetric loading. SO_4^{2-}, main metabolism products of H_2S, could lead to decrease of pH, which is an important factor affecting the activity of microorganisms. With the increase of volumetric loading, pH in effluent spray water decreased gradually. When volumetric loading was more than 4.83 g/(m^3·h), pH in effluent spray water decreased to 1~2, whereas the removal efficiency of the bioreactor was nearly constant and higher than 95%. Cox and Deshusses (2002) also observed similar results in a single-stage bio-trickling filter. Therefore, low pH did not decrease the removal efficiency of H_2S.

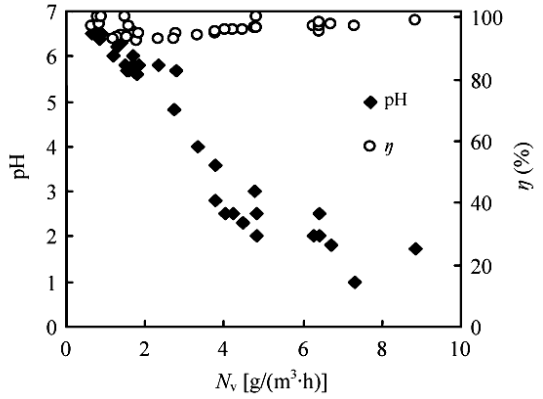


Fig.6 Effect of volumetric loading (N_v) on pH and removal efficiency (η) of H_2S

Effect of gas retention time on H_2S removal

The effect of gas retention time on H_2S removal was studied by varying the gas flow rate through the bioreactor. The results are shown in Table 2. In the first dataset of 8 tests presented in Table 2, the bioreactor inlet gas concentration was maintained constant. With shorter retention time, outlet concentration and elimination capacity of Section A increased gradually. When retention time was longer than 30 s, removal efficiency of Section A was more than 87%. However, removal efficiency of Section A decreased obviously and corresponding outlet concentration of Section A increased significantly from 31.72 mg/m^3 to 45 mg/m^3 when retention time was shorter than 30 s. Furthermore, retention time was in inverse proportion to volumetric loading at constant inlet concentration. Removal efficiency of Section A decreased gradually with decreasing retention time and/or corresponding increasing volumetric loading. Conversely, removal efficiency of Section A could increase when retention time increased and/or corresponding volumetric loading decreased. When retention time increased to 30 s from 27 s and corresponding volumetric loading reduced by nearly 3.33 g/(m^3·h), removal efficiency of Section A increased by about 6%. Compared with the results at higher than 30 s, its change was significant, which indicated that 30 s was a critical value to keep higher removal efficiency of Section A. Thus, it could be seen that, as long as retention time was

long enough, the bioreactor could achieve high removal efficiency at constant inlet concentration.

In the second dataset of 6 tests, the H_2S inlet volumetric loading was kept nearly constant to investigate the effect of retention time on elimination capacity of H_2S. It was obvious that there was no apparent effect of the retention time on H_2S removal during the experiment as long as the retention time was longer than 30 s. When inlet volumetric loading was about 30 g/(m^3·h), removal efficiency of Section A was more than 86.74% and elimination capacity of Section A was about 25.92 g/(m^3·h) at longer than 30 s retention time. However, when the retention time was reduced to 27 s, the removal efficiency of Section A decreased to 82.13% and elimination capacity of Section A decreased to 24.09 g/(m^3·h), which suggested that the reduction of H_2S removal efficiency under shorter residence time was either due to insufficient reaction time between the H_2S and the biomass or the slower step of H_2S diffusion from the gas phase into the liquid phase.

Effect of spray operation on removal efficiency of H_2S

Enzymes generated by microorganisms are the catalysts of deodorization reaction, so it is a key factor to maintain the powerful vigor of microorganisms during the bio-trickling filter operation. The spray operation achieves two significant effects, and can supply microorganisms with necessary nutrients and wash the products such as SO_4^{2-} and element sulfur generated by bacteria metabolism. The washing also improves the stability of the reactor because old biofilms were removed from the packing carrier.

The effect of spray water flow rate on removal efficiency of H_2S was evaluated when the gas flow rate and inlet concentration were 9 m^3/h and 250 mg/m^3, respectively. Fig.7 shows that when spray water flow rate is less than 0.005 9 L/(cm^2·h), removal efficiency of H_2S increases with its increasing. However, with spray water flow rate of more than 0.012 L/(cm^2·h), removal efficiency of H_2S decreases with its increasing.

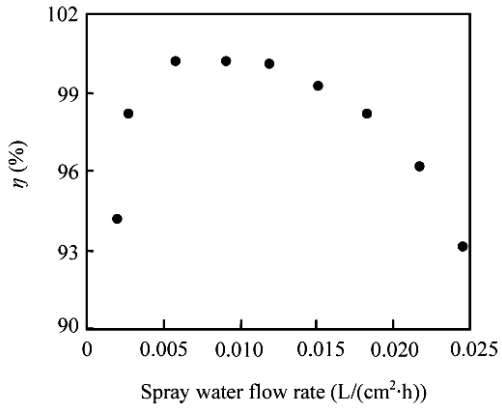


Fig.7 Effect of spray water flow rate on removal efficiency (η) of H_2S

Table 2 Effect of retention time on removal efficiency of H_2S

C_{in} (mg/m^3)	τ (s)	Q (m^3/h)	N_v (g/(m^3·h))	C_{out} (mg/m^3)			η (%)			EC_A [g/(m^3·h)]
				C_A	C_B	C_C	η_A	η_{A+B}	η_{A+B+C}	
250	67.50	4.0	13.33	0	0	0	100	100	100	13.33
250	54.00	5.0	16.67	1.52	0	0	99.39	100	100	16.57
250	45.00	6.0	20.00	6.96	0	0	97.22	100	100	19.44
250	37.50	7.2	24.00	17.83	0	0	92.87	100	100	22.29
250	33.33	8.1	27.00	25.98	0	0	89.61	100	100	24.19
250	30.00	9.0	30.00	31.72	1.00	0	87.31	99.60	100	26.19
250	27.00	10.0	33.33	45.24	8.00	0	81.90	96.80	100	27.30
250	22.49	12.0	40.00	56.68	10.00	0	77.33	96.00	100	30.93
220	27.00	10.0	29.33	39.32	5.20	0	82.13	97.64	100	24.09
249	30.00	9.0	29.88	33.01	3.09	0	86.74	98.76	100	25.92
285	33.75	8.0	30.40	34.32	3.14	0	87.96	98.90	100	26.74
310	36.00	7.5	31.00	35.32	2.95	0	88.61	99.05	100	27.47
325	38.57	7.0	30.33	34.81	3.51	0	89.29	98.92	100	27.08
368	45.00	6.0	29.44	33.81	3.46	0	90.81	99.06	100	26.74

τ: Retention time; Q: Flow rate N_v: Volumetric loading; C_{in}: Inlet concentration; C_{out}: Outlet concentration; η: Removal efficiency; EC: Elimination capacity

The results suggested excessive spray water could result in two negative effects: (1) Thick water film on biofilm surface leads to increasing resistance of transferring H_2S into the biofilm; (2) Biofilms easily fall off.

Pressure drop and work life

Resistance to gas flow is the major factor that determines the amount of energy needed by the blowers to force the contaminated gas through the filter bed material. During the experiment, the pressure drop was continuously monitored using a water manometer, showing that it was approximately 42 mm H_2O/m at gas flow rate of 9 m^3/h. The bio-trickling filter had been running continuously for nearly 4 months without back washing. The study suggests that ZX01 stuffing used is a kind of excellent material to deodorize. Besides its lower density and favorable biological adsorption, it has higher moisture content, high void fraction and mechanical tension, which contribute to maintain lower pressure drop of the bio-trickling filter during the experiment. At the same time, spray operation plays also an important role in favor of shucking old biofilm and metabolic products. Therefore, it is conceivable that the bio-trickling filter can work well and stably for long time without back washing.

CONCLUSION

As an air pollution control technology, the bio-trickling filter for treatment of air streams contaminated with odor is reliable, highly efficient and easy to be operated and maintained. Through continuous deodorization experiment for nearly four months, the following conclusions were obtained:

(1) The bio-trickling filter packed with ZX01 stuffing has high removal efficiency and elimination capacity for H_2S gas. The removal efficiency approaches nearly 100% when inlet volumetric loading of H_2S is lower than 38 g/(m^3·h).

(2) The bioreactor has enormous potential to increase inlet volumetric loading and elimination capacity. Removal efficiency of Section A is higher than 90% when inlet volumetric loading of Section A is lower than 94.90 g/(m^3·h). Section A occupies only 22.64% of the whole tower height and has over 90% of removal efficiency when inlet volumetric loading is lower than 26.35 g/(m^3·h).

(3) Suitable volume of spray water has a positive effect on retaining the vigor of microorganisms with the optimal spray water flow rate in this study being 0.0059~0.012 L/(cm^2·h) at 250 mg/m^3 of inlet concentration. Long-term operational results showed that the biological reactor is not jammed during the operation and that the resistance in the tower is lower than 42 mm H_2O/m when the inlet gas flow rate is lower than 9 m^3/h. Therefore, the bio-trickling filter need not do back washing frequently and can be operated steadily.

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