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Comparison of packing materials in biofilter system for the biological removal of hydrogen sulfide: Polypropylene fibrils and volcanic stone

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Abstract−In order to develop a method for the removal of hydrogen sulfide via a biological process, two different packing materials were tested to assess their capabilities as biofilter bed materials under variable conditions of two parameters: inlet gas concentration and inlet gas flow rate. We detected a maximal elimination capacity (critical loading rate) of 515.1 (410.5) g-$H_2S/m^3 \cdot hr$, and 415.5 (80.0) g-$H_2S/m^3 \cdot hr$, respectively, when polypropylene fibrils and volcanic stone were employed as supporting materials. The results of this study show that the application of polypropylene fibrils might be a favorable choice as a packing material in biofilter for the biological removal of hydrogen sulfide.

Key words: Biodegradation, Biofilter, Packing Bed Bioreactor, Hydrogen Sulfide, Packing Material

INTRODUCTION

Pulping sites, petroleum refinery plants, drug manufacturing processes, sewage treatment facilities, and livestock farms all generate a variety of offensive odors. The majority of these odors are attributable to the production and dissemination of sulfur compounds. Among the many types of sulfur-containing compounds associated with malodorous gas emission, hydrogen sulfide (H_2S), methyl mercaptane (CH_3SH), dimethyl sulfide ($(CH_3)_2S$), and dimethyl disulfide ($(CH_3)_2S_2$) are some of the more notorious, and these compounds are generated abundantly in many sites and processes. In particular, hydrogen sulfide, an extremely toxic and corrosive gas, can be employed as a standard indicator for this type of offensive odor [1]. Therefore, the amount of this compound released into the air should be strictly regulated. Substantial quantities of hydrogen sulfide are normally generated in industrial facilities, and an emission concentration in a range between 5 to 70 ppm is expected as the result of these processes, which include wastewater treatment, paper/pulp manufacturing, food processing, and waste treatments. The hazards associated with high concentrations of hydrogen sulfide are fairly well established, but little information is currently available with regard to long-term human exposure to low concentrations of this compound [1,2].

Among the biological processes thus far explored in this regard, biofiltration is one of the successful examples of biotechnological methods used in an environmental engineering context. Biofiltration, which is most frequently utilized in the removal of odoriferous compounds, has proven effective in applications involving the treatment of substantial volumes of air which harbor low odor concentrations [3-5]. These processes have also proven effective in the removal of alcohols, toluene, phenol, ketones, hydrogen sulfide, petroleum fuel vapors, and other assorted volatile organic compounds (VOCs) [1,3,4,6,7]. This biological process has proven effective in applications involving the treatment of substantial air volumes harboring low odor concentrations. These processes tend to be associated with low operating costs, and are also effective in the treatment of large volumes of moist air, which harbors low concentrations of biodegradable compounds [2,8,9].

The packing material within the biofilter serves as a habitat and a supplier of nutrients and water. The performance of the biofilter depends substantially on the nature of the carrier, which is also referred to as the support or packing material, or as the filter bed. Many different types of the packing materials have been used in this system, including natural materials, inert compounds, and mixtures of both. Any effective packing material must possess a number of characteristics in order to ensure maximum biofilter performance. These characteristics include a high specific surface area for gas contact, high microbe immobilization, significant water retention capacity, air permeability, easy removal of deodorization waste, and low economic cost. In addition, the carrier must be very durable, with no clogging/blocking or pressure drops in the packed bed during its operation. Several carriers currently satisfy these characteristics [2, 4,6,10-13].

In previous reports, a variety of packing materials have been employed as biofilter supporters, including peat, wood bark, compost, activated carbon, activated carbon fabric, filamentous carbon, polyvinyl chloride, polyurethane foam, ion-exchange resins, porous ceramic, refractory brick, sintered glass, biosand, lava rock, rock wool, and volcanic stone [12,14]. Polypropylene fibril materials have some advantages such as chemical/physical stability, easy to modify, no loss of material by microorganism, long life time, high air-permeability and high surface area. Also, this type of material is used for packing materials in the field of water treatment for immobilization of cells [15,16]. Jeong et al. [15,16] reported that the attachment of microorganisms and removal of BTX and MEK are pro-

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per in a biofilter system using polypropylene fibril support. Volcanic stone has been used as a filter aid and adsorbent of heavy metal by some properties such as generally high porosity and proper strength. Lee et al. [14] reported that the hydrogen sulfide removal experiments performed to in the biofilter system using scoria derived from Jeju (Korea) as packing material with *Bacillus* sludge.

The maximum elimination capacity of a biofilter is referred to as the maximum load that can be sustained by the system without any inhibition of microbial activity. This variable is expressed in the same units as is the inlet load. These parameters differ under various operation conditions and also vary with the type of biofilter packing materials employed. These parameters, then, were also utilized as key factors in the design of an optimal biofilter [2,11,17].

In an attempt to develop a method for the removal of hydrogen sulfide removal via a biological process, two different packing materials, polypropylene fibrils and volcanic stone, were evaluated with regard to their capabilities as biofilter bed materials under variable conditions of inlet gas concentration and inlet gas flow rate.

MATERIALS AND METHODS

1. Microbes and Nutrient

All of the biofilters were inoculated by using a microbial consortium which was composed primarily of three microbes (*Pseudomonas* sp. TKC, *Pseudomonas* sp. AKC, and *Geotrichum* sp. MKC) [9], which had been isolated from contaminated soil and slurry from industrial complexes in Gwangju, Ulsan, Onsan, and Yeocheon, Korea, as well as *Thiobacillus* sp. IW [1], which was isolated from acid drainage water obtained from coal mines (Hwasoon, Korea). The medium employed for the maintenance of the microbial consortium included NH_4Cl_4 1.0 g/L, K_2HPO_4 4.35 g/L, NaH_2PO_4 3.9 g/L, $MgSO_4 \cdot 7H_2O$ 7.2 g/L, $CaCl_2$ 0.45 g/L, $FeSO_4$ 0.45 g/L, $MnCl_2$ 0.45 g/L, $CuCl_2$ 0.45 g/L, and $NaMoO_4$ 0.45 g/L. The medium was adjusted to neutral pH and then autoclaved for 20 minutes at 121 °C and 1.5 atm. The nutrients for the growth of the microbes in the biofilter were supplied with a minimal salt medium (MSM) solution, containing KH_2PO_4 1.50 g/L, Na_2HPO_4 6.00 g/L, $(NH_4)_2SO_4$ 3.00 g/L, $MgSO_4$ 0.05 g/L, $CaCl_2$ 0.01 g/L; and this medium was adjusted to a pH of 7.0.

In order to investigate the removal characteristics of hydrogen sulfide in biofilter, we utilized polypropylene fibrils (PPF; Daehosangi Co., Ltd, Korea) and volcanic stone (VS; B&E Tech., Co., Ltd, Korea) as packing materials in biofilter. The characteristics of the packing materials and operating conditions are described in Tables 1 and 2, respectively.

2. Operation and Construction of Biofilter

The biofilter (Fig. 1) was constructed from a transparent acryl tube, with an inner diameter of 9.4 cm and a bed length of 25 cm. A pore plate was installed in the bed of the support carrier. The biofilter was operated at a temperature of 25-35 °C. Inlet gas was in-

Table 1. Characteristics of experimental matrix of biofilter

	Polypropylene fibril	Volcanic stone
Chemical composition	C, H, O	Si, O, Al, Fe
Surface area	0.8 m^2/m length	148 m^2/cm^{3*}
Porosity (%)	93	75

*This value is obtained from the data of Lee et al. [14].

Table 2. Operating conditions of biofilter

Operating parameter		Operating conditions	
Bed volume (L)		0.8	
Matrix of bed	Kind	PPF*	VS*
	Weight (g)	101.7	110.0
	Volume (cm^3)	456	465
	Packing ratio (%, v/v)	56.5	57.1
	Particle size (cm×cm×cm)	10 cm	1×1×1
SV** (1/h)		120-720	
EBCT*** (sec)		60-10	
Inlet air flow rate (L/min)		1.6-4.8	
Temperature in bed (°C)		25-35	
Initial inlet H_2S concentration (ppm)		25	
Maximum inlet H_2S concentration (ppm)		750-1000	

*Polypropylene fibril (PPF), and volcanic stone (VS).
**SV: space velocity.
***EBCT: empty bed contact time.

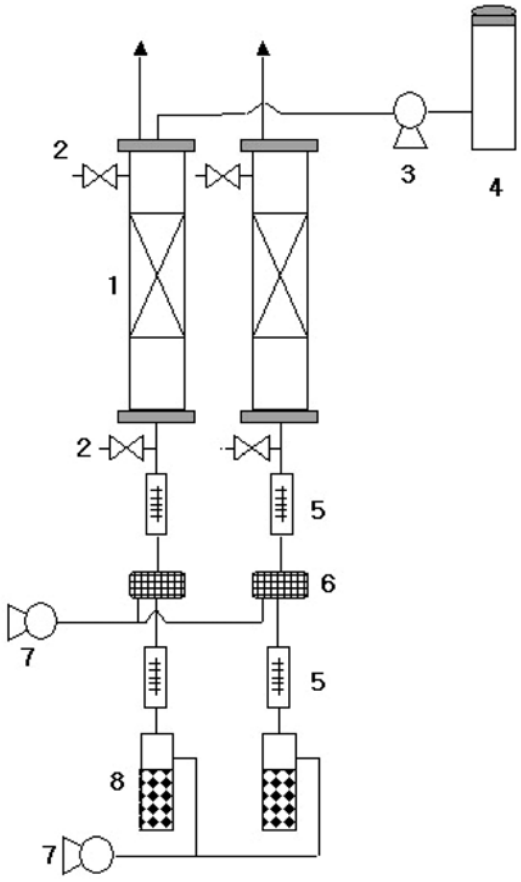

Fig. 1. Schematic diagram of biofilter system.
1. Biofilter reactor
2. Check valve
3. Pump
4. Nutrient tank
5. Mass flow controller
6. Mixing chamber
7. Air compressor
8. Target gas generator

troduced into the bottom of the biofilter and then allowed to escape from the upper portion of the biofilter. Exhaust gases were emitted after passage through an NaOH absorption solution for the removal of residues. The biofilter was inoculated with a microbial consortium, an enriched version of that reported previously [18]. To supply the necessary nutrients to the microorganisms, we periodically fed the system with a nutrient solution using a peristaltic pump. We also sprayed 0.1 L of water into the system at 2-4 hour intervals in order to protect the biofilter bed from drying at the upper nozzle (Full jet nozzle, FF 6.5, orifice diameter 2.31 mm; Hanmi Nozzle, Korea) of the reactors. The inlet gas was prepared by mixing H_2S with the air-flow, and the inlet concentration and amount were controlled with a mass flow controller and a flow meter. Samples were collected from the upper and bottom portions of the assembly, as well as from each of the sampling ports of the biofilter; they were then analyzed. Gas samples were obtained from the inlet and outlet streams, and axially along the length of the biofilter. In this study, we applied hydrogen sulfide to the biofilter system. The EBCT (empty bed contact time) of the biofilter was controlled at 60-5 seconds in order to facilitate biofilm growth, after which the inlet loading was controlled. In order to control the humidity in the input gas, air was passed through the humidifier prior to mixing with prepared gas, and the mixed gas was introduced into the bottom of the biofilter. In order to attain sufficient biomass concentration in the biofilter bed, the biofilter was adapted. The biofilter was initially acclimatized by operating the biofilter at a low concentration (maximum 10 ppm H_2S) and low gas flow rates of 1.6 L/min, for 3 days.

3. Analytical Method

Hydrogen sulfide gas was collected at both the inlet and the outlet of the biofilter system. We used a hydrogen sulfide detector (PGM-30, 0-50 ppm, RAE System, USA) to detect H_2S concentrations of less than 50 ppm. We used a gas detector (4LL (2.5-60 ppm), 4M (25-250 ppm), 4H (100-2,000 ppm), and 4HH (0.1-2%), Gastec, Japan) to detect concentrations in excess of 50 ppm. Data was then calculated on the basis of the five determinants. The H_2S concentrations of the inlet/outlet streams were determined with a gas chromatograph (GC-14B, Shimadzu, Japan) equipped with a TCD. The column used in this study was a packed column (HayeSep D, 100/120 mesh, 10 ft×1/8"). The oven was operated at a temperature of 80 °C. The temperature and electric current of the detector were 200 °C and 100 mA, respectively. Helium was utilized as a carrier gas at a flow rate of 30 mL/min.

4. Calculation of Removal Efficiency and Elimination Capacity

Removal efficiency (RE) and elimination capacity (EC) were the variables used to determine the treatment capacity of the biofilter. Removal efficiency was expressed as the content (%) of odoriferous gas eliminated by the biofilter. Elimination capacity was expressed as the amount of odoriferous gas removed in the bed volume per unit of time.

$$RE\ [\%]=(C_{Gi}-C_{Go})/C_{Gi}\times 100$$

$$EC\ [g/m^3\cdot hr]=(C_{Gi}-C_{Go})\times Q/V_f$$

in which Q is the gas flow rate (m^3/h), V_f is the volume of the filter bed (m^3) and C_{Gi} and C_{Go} are the inlet and outlet hydrogen sulfide concentrations (ppm; g/m^3).

RESULTS AND DISCUSSION

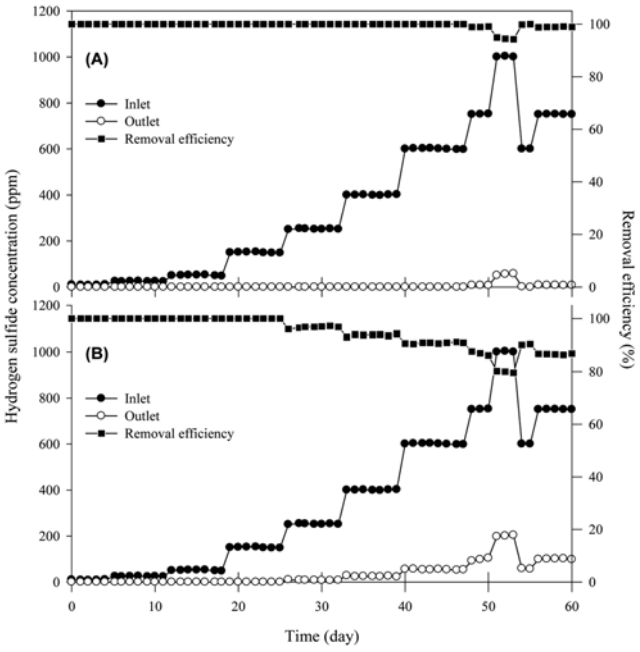

Fig. 2. Hydrogen sulfide removal pattern at constant air flow rate and varying inlet concentrations using different packing materials. (A) polypropylene fibril, (B) volcanic stone. Inlet flow of 4.8 L/min and SV (space velocity) of 360/hr, and then the inlet H_2S concentration was increased in weekly increments, from 25 ppm to a final value of 1,000 ppm.

1. Removal of H_2S at Varying Inlet Concentrations

The two most important factors in a biofilter system for hydrogen sulfide removal are the inlet gas concentration and the inlet gas flow rate [8,19]. The removal characteristics and yields of the H_2S gases tested herein were observed by using two different packing materials within separate biofilter systems (Fig. 2). In order to induce the initial adaptation of the microorganisms, the tested H_2S gas was introduced into the biofilter at low concentrations (maximum 10 ppm) for three days. After the adaptation period was completed, the inlet flow was maintained at 4.8 L/min and SV (space velocity) 360/hr, after which the H_2S concentration at the inlet was increased in weekly increments, from 25 ppm to a final value of 1,000 ppm. Fig. 2(A) shows the inlet/outlet concentrations and H_2S removal efficiency values when polypropylene fibrils were employed as packing material. In this system, we attained a removal efficiency of approximately 99% at an H_2S concentration of up to 750 ppm. However, removal efficiency was reduced to 94% at a H_2S inlet concentration of 1,000 ppm, with an outlet concentration of 57 ppm. After exposure to high concentrations, we observed low removal yields. At a retention time of SV 360/hr, the inlet H_2S was adequately removed by the polypropylene fibrils, up to a concentration of 750 ppm. This system also recovered a high degree of removal efficiency following the reapplication of low H_2S concentrations. As a result, we determined that the H_2S could be treated with polypropylene fibrils in a relatively wide inlet concentration range from low to high, and that no overt growth inhibition occurred as the result of incremental changes in the inlet stream H_2S concentration. In addition, removal efficiency was adequately recovered from the shock load of the short period.

Fig. 2(B) shows the results of different inlet/outlet concentrations and the H_2S removal efficiency values when volcanic stone was utilized as the packing material. As shown in Fig. 2(B), we noted a removal efficiency of approximately 95% with an H_2S concentration of up to 250 ppm when volcanic stone was employed as the packing material. In an H_2S concentration range between 250 and 600 ppm, the removal efficiency was determined to range between 92% and 84%, with an outlet concentration range of 26-139 ppm. However, at an H_2S inlet concentration of 750 ppm, the removal efficiency was reduced to a maximum of 87%. However, Lee et al. [14] reported that a biofilter system using scoria as packing material removed 99.9% of hydrogen sulfide in the range of 1,100 ppm of inlet concentration with EBCT 30 sec.

In conclusion, the H_2S removal efficiency was higher when using polypropylene fibrils as packing material, as compared with volcanic stone, under conditions of constant flow (4.8 L/min, SV 360/hr) and varying inlet concentrations (25-1,000 ppm H_2S). In a report concerning hydrogen sulfide biofiltration conducted by Yang and Allen [20], the removal rate leveled off at approximately 130 $g/m^3 \cdot hr$. This implies that the maximal biological degradation rate was achieved at inlet loading rates in excess of 150 $g/m^3 \cdot hr$. The biofiltration systems that harbored a variety of yard waste compost matter as filter materials were determined to remove H_2S at efficiencies in excess of 99.9% for H_2S inlet concentrations, in a range of 5-2,650 ppmv. In comparison of removal yield, the results of other reports are higher than that of ours. However, the removal yield of hydrogen sulfide may be enhanced by the increase of EBCT in our experiment. More experimentation is required for exact comparison with other systems.

2. H_2S Removal at Varying Inlet Gas Flow Rates

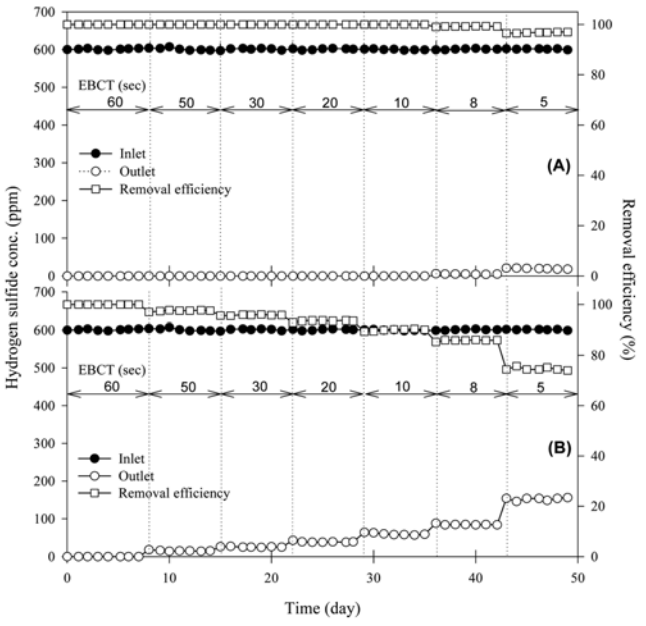

Fig. 3. Hydrogen sulfide removal pattern at a constant inlet concentration and a variable flow rate (EBCT). (A) polypropylene fibril, (B) volcanic stone. H_2S gas was introduced into the biofilters at a concentration of 600 ppm, via incremental changes in the inlet stream flow, at a 0-60 sec retention time for each packing material.

In order to determine H_2S removal characteristics on the basis of changes in the inlet flow rate (changes in retention time), we created biofilters that utilized different packing materials, and also ensured constant H_2S concentration in the inlet stream, while incrementally altering the flow rate. In order to compare the removal efficiencies of several packing materials in the biofilters, H_2S gas was introduced into the biofilters at a concentration of 600 ppm, via incremental changes in the inlet stream flow, in the range of 0-60 sec retention time for each of the packing materials. Fig. 3 shows the inlet/outlet concentration and H_2S removal efficiency values for each packing material under conditions including incremental changes in the inlet load. Fig. 3(A) shows the inlet/outlet concentrations and H_2S removal efficiency values when polypropylene fibrils were employed as packing materials. In this system, a removal efficiency in excess of 99.2% was attained at a retention time of 8-60 seconds. However, the removal efficiency was reduced to 96.8% at an outlet concentration of 19.2 ppm, with an H_2S retention time of 5 sec. Fig. 3(B) shows the results of different inlet/outlet concentrations and the H_2S removal efficiency values when volcanic stone was employed as the packing material. As is noted in Fig. 3(B), we observed a removal efficiency of approximately 95.8% with an H_2S retention time of 30-60 sec, when volcanic stone was employed as the packing material. At an H_2S retention time of 5-20 sec, the removal efficiency was 74.6-93.5%, with an outlet concentration range of 152.4-39 ppm. In comparison, when polypropylene fibrils were used as packing material, the H_2S removal rates were superior to those obtained with the volcanic stone.

Fig. 4 shows H_2S removal efficiency with changes in EBCT and different packing materials. As is shown in Fig. 4, H_2S removal efficiency was higher with polypropylene fibrils as packing material than with volcanic stone, under conditions of constant inlet concentration (600 ppm H_2S) and different inlet loads (EBCT 5-60 sec). In the biofilter, the decrease in the EBCT value translates to increases in the inlet load and inlet flow rate of hydrogen sulfide. This also coincides with a decrease in the size of the biofilter under constant inlet flow rate conditions. With polypropylene fibrils, reductions in retention time did not significantly affect H_2S removal efficiency. However, with volcanic stone as a packing material, the H_2S re-

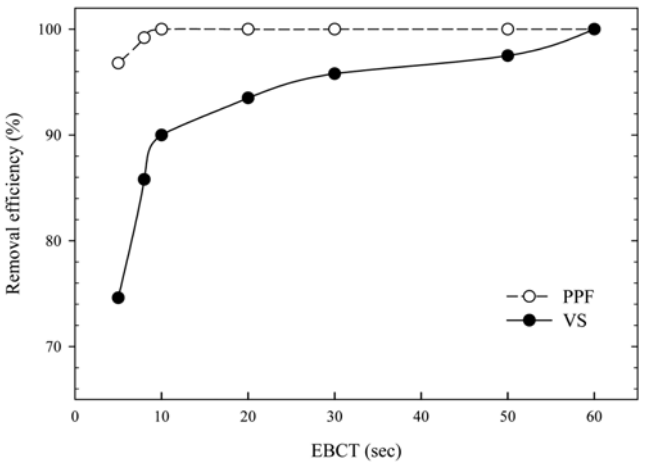

Fig. 4. Changes in removal efficiency with changes in EBCT. Polypropylene fibrils (PPF), and volcanic stone (VS).

moval efficiency abruptly decreased.

In our comparison of two different packing materials at a retention time of 10 seconds, the polypropylene fibrils completely removed the malodorous gases. However, the volcanic stone evidenced a removal efficiency of only 90%. Kam et al. [21] reported that the biofilter in which the activated carbon/polyurethane composite inoculated with *Bacillus* sp. was utilized as a packing material evidenced a stable removal yield of over 99% under a range of EBCT from 15 to 60 sec at a hydrogen sulfide inlet concentration of 300 ppmv. Also, Lee et al. [14] reported that the biofilter with scoria showed stable removal yield of over 99% under the EBCT range from 8.2 to 60 sec at a hydrogen sulfide inlet concentration of 250 ppmv. In comparison with these results, the differences of removal yield were interpreted by the different packing material and inoculated microorganisms.

3. Maximum Elimination Capacity and Critical Loading Rate

The maximum elimination capacity and critical loading rate (critical removal rate) are principal factors considered in biofilter design, and are also primary biofilter performance considerations under identical conditions. Fig. 5 shows the elimination capacity curves for each biofilter system. In the polypropylene fibril system, the maximum elimination capacity was found to be 515.1 g-H_2S/m^3·hr. By way of contrast, the maximum elimination capacity of volcanic stone was determined to be 415.5 g-H_2S/m^3·hr, a figure substantially lower than was attained with polypropylene fibrils as packing material. In comparison with other results, Kam et al. [21] reported that the maximum elimination capacity in the biofilter packed with activated carbon/polyurethane composite media was 157 g-H_2S/m^3·hr. Lee et al. [14] reported that the maximum elimination capacity in the biofilter packed with volcanic rock as packing material was 254 g-H_2S/m^3·hr. Also, Eun et al. [22] reported that the removal efficiency in the rock wool biofilter was mostly above 99%, with a maximum elimination capacity of 1,115 g-sulfur/m^3_{bed}day (49.4 g-H_2S/m^3·hr). Rock wool was a highly efficient packing material in biofilters for hydrogen sulfide treatment at inlet concentrations of up to 250 ppmv at an EBCT of 30 seconds. The maximum elimination capacity of other results showed lower than that of ours. These results estimated the differences of packing material and inoculated microorganisms.

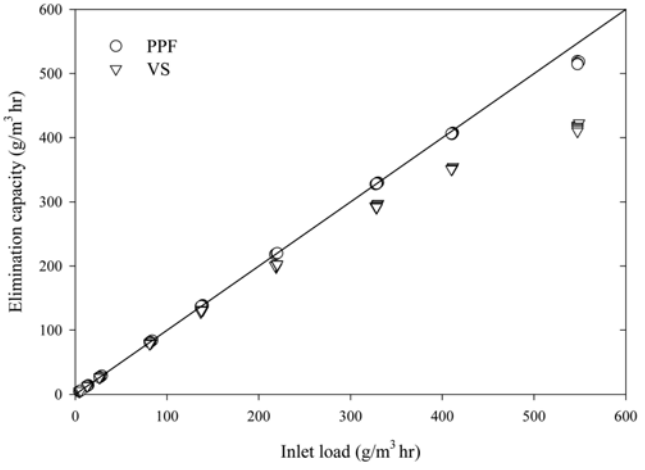

Fig. 5. Inlet load vs. elimination capacity of hydrogen sulfide in varying packing materials. Polypropylene fibril (PPF), volcanic stone (VS).

The diagonal line in Fig. 5 shows a 100% elimination of inlet gas. In this study, the critical loading rate (maximum loading rate to remove 100% of the inlet gas) of hydrogen sulfide using polypropylene fibrils and volcanic stone was calculated at approximately 410.5 g-H_2S/m^3·hr, and 80.0 g-H_2S/m^3·hr, respectively. In the other H_2S removal experiments using a fluidized bed bioreactor, Oh et al. [23] and Kim et al. [24] reported that the critical hydrogen sulfide loading rates were 15 g-H_2S/m^3·hr and 12 g-H_2S/m^3·hr, using *Bacillus cereus* and *Thiobacillis* sp. IW, respectively. Whereas, Yang and Allen [20] reported a critical loading rate of 100 g-H_2S/m^3·hr.

Our results may be attributed to the fact that, in the continuous treatment of hydrogen sulfide using a biofilter inoculated with a microbial consortium, the application of polypropylene fibrils as packing material for the biofilter might represent a viable alternative. In addition, the diversity of biofiltration mechanisms and their interactions with the microbial environment tend to be more complex. Therefore, more research concerning the microbial ecology in biofilters should be conducted, in order to overcome some of the traditional difficulties inherent in the construction of biological treatment systems.

CONCLUSION

The industrial application of biofilter systems requires appropriate packing materials for microbial growth. In order to develop this H_2S removal process, we have attempted to evaluate the capabilities of biofilter support materials under variable inlet concentrations and inlet gas flow rates. H_2S removal efficiency was higher in cases in which polypropylene fibrils were utilized as packing materials than when volcanic stone was used. These results should prove useful in the design of biofilters and the choice of packing materials.

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