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Hydrogen Sulfide Removal by *Thiobacillus thioparus* Bacteria on Seashell Bed Biofilters

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Abstract: The aim of this research is to achieve an efficient and cheap methods to remove H_2S from the factories emissions. Four serial cylinders are designed, 40 cm in height and 15 cm in diameter each. They are filled with bivalve seashells with 63% porosity which contains *Thiobacillus thioparus* bacteria to the maximum height of 27.5 cm. By mixing phosphoric acid and sodium sulfide, H_2S gas is released and its concentration is measured as mg m^{-3} before injecting into the cylinders. A permanent measuring instrument is equipped to control the gas coming out of the cylinders. In order to prevent the outdoor environment from pollution, first the gas is sent through two activated carbon columns and then sent through a ferrous chloride scrubber. Finally it is burnt directly by flames. There were 550 sample readings in 15 weeks. The changes in the discharge of the air which carries the gas are considered between 1-12 L min^{-1} and the concentration of the influent pollutant is considered between 1-140 mg m^{-3}. Also the humidity in the atmosphere is fixed between 77-93% and the optimum temperature required for growing of the microorganisms is retained between 20.5-30°C. After feeding the system for three weeks the efficiency started to increase so that by the end of the final week of this research the efficiency reached to 90% with the discharge of 6 L min^{-1} of the carrier gas. The results achieved from this research show that because of not using Filamentous bacteria, clogging did not occur in the biological system in biofilters. The amount of head loss in cylinder was only 2 mm water and during this research, head loss was the same due to unclogging of filter. On the other hand the traditional methods are expensive in terms of using chemicals, carbon recycling and using fuel and etc. Therefore researchers have started new studies in this field. The above mentioned method, according to high efficiency, inexpensiveness and easiness of control and maintenance is considered one of the best methods.

Key words: Air pollution, biofilters, hydrogen sulfide, *Thiobacillus thioparus* bacteria, biological removal

INTRODUCTION

Compounds such as hydrogen sulfide and ammonia are pollutants that come out of many different factories like compost producing plants, food industries, refineries and wastewater treatment plants. There are different methods to remove these troublesome compounds. Some of the most important ways of removing them are; surface adsorption by activated carbon, burning, catalytic oxidation, wet scrubbers and oxidation by heating (Shamansouri *et al*., 2005; Mosqera and Sanchez, 2002; Langenhove, 1986). These methods are often expensive and produce other pollutants as well. That is why hydrogen sulfide removal in industries is restricted (Vanlith, 1997; Pomery, 1996).

These days using Biofilters has been taken into consideration by experts because of its advantages such as low cost, low energy consumption, no chemical usage and no production of contaminated by products (Armand, 1994).

In a biofilter the reaction is similar to a biofilm. In this mechanism the reaction takes place in three stages in substrate. First the chemical substance in gas phase between demarcation lines of gas flow and vacuolar space in solid bed, passes through biofilm, then the chemical substances in biofilm penetrates in the united combination of microorganism and finally the microorganisms get the needed energy from the oxidation of chemical substances in form of primary substrate or co-metabolism (Guillermo *et al*., 2005; Satoh *et al*., 2004). In this process nutrients such as nitrogen, phosphorus, sulfur and oxygen are simultaneously penetrated and used between biofilm media and air flow (Sheridan and Curran, 2002). If the biofilter is properly

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designed almost all the pollutants are removed and changed into CO_2, water, salt and biomass (Kim and Chung, 2002). The gas compounds leave the biofilter environment by spreading into the air flow. The increase of salt and biomass leads to increasing of the thickness of the bed and decreasing of the space for gas to pass (Guillermo *et al*., 2005), which is caused by improper design of the system therefore it can lead to falling of the pressure and decreasing removal efficiency (Malhautier and Grecia, 2003).

Removing odorous substances such as hydrogen sulfide, other sulfurous compounds and nitrogenous compounds such as mercaptan, methanol or ammonia has been one of the main efforts in development of biofilters in industries (Luo *et al*., 1997).

Biofilters with organic bed are usually used for biological treatment of VIC. Microbial types like *Thiobacillus* and *Hyphomicrobium* are the best for removal of sulfides in biofilters (Oyarzun *et al*., 2003). Also volatile organic compounds can be treated by specialized micro-organisms. These compounds consist of halogenated and nonhalogenated aliphatics and aromatic pollutants (Deshusses and Webster, 2002). Full studies on removal of some specific pollutants with high stability in air such as alcohols, ketones, alkanes, benzene derivations and chlorinated compounds are being done and remarkable results have been achieved (Kim and Sugano, 2000). Most of the researchers have used microbial consortium for treatment which is in compost of edible mushrooms. In this microbial complex after microbial adaptation for decomposing the pollutant, the removal efficiency will be over 90% (Shojasadati and Elyasi, 1999). In this investigation purified *Thiobacillus thioparus* bacteria is used. After adaptation period according to microbial reactions the environment will become acidic, the filling substance (media) is high-porous seashells (63%), therefore the calcareous bed keeps the environment neutral which is effective for removal efficiency.

MATERIALS AND METHODS

This research was done in 2006 at Environmental Health Engineering lab of Shahid Beheshti University of Medical Sciences. Biofilter cylinders are 40 cm each and made of Plexiglas. Each cylinder is filled with tiny seashells up to the height of 27.5 cm. Seashell porosity is 63%. On top of each cylinder, 4 tubes are installed. These tubes are used for air and pollutant gas injection, reading output sample from each cylinder, nutritious substances injection and culture medium. There are 4 cylinders installed in series and considering the equal condition in each cylinder all results will be the average of six readings per time unit. To have even inlet gas flow, 20 small openings -150 mm in diameter each-are created at the bottom of each cylinder. In order to protect the employees from probable emission of the pollutant gas, at the outlet of each cylinder there is a column of activated carbon, a scrubber containing Ferric chloride and finally flame.

To make the pollutant gas flow in biofilter cylinders a compressed air compressor with working pressure of 6-8 bars and the capacity of 150 L of compressed air is used. The outlet flow rate of gas is controlled by a needle valve. Also in order to prevent oil particles to enter the cylinders, fiber glass filters are used in series at the outlet of the gas. These filters are 50 cm in length and 10 cm in diameter.

To produce H_2S gas, a tank made of hard aluminum is used and it is equipped with a barometer and a steel needle valve with the diameter of 10 mm. Tank's height is 310 mm and its diameter is 320 mm. Three hundred and fifty gram of Na_2S is dissolved in distilled water and is mixed with 300 cc of phosphoric acid. Acid container is placed in the middle of Na_2S solution. After closing the tank's lid it is shaken very hard. The reaction takes place in closed environment and H_2S gas is released.

$$3Na_2S + 2H_3PO_4 \rightarrow 2Na_3PO_4 + 3H_2S\uparrow$$

The barometer will show 1.5 bars.

To provide moisture for microorganisms to grow, first the air flow from the compressor enters a diffuser and then in form of bubbles passes through water in a closed tank. An electric heater is available to heat the water to the desired temperature if needed. Finally, the humid gas, whether warm or cold, passes through a flow meter and with a steady flow enters the cylinders.

The flow rate of the pollutant gas is adjusted by a manometer. In order to control the amount of pollutant gas at the inlet and outlet of the cylinders, an H_2S gas measuring device that is made in Micropac Plus Co., is used and the measuring unit is mg m^{-3}. Also to check the humidity a digital hygrometer made in Danfouse Co. Germany, with ±0.01 precision is used. A digital thermometer made in Sumone, Korea, with ±1 precision is also used. The flowchart of the constructed pilot plant is shown in Fig. 1.

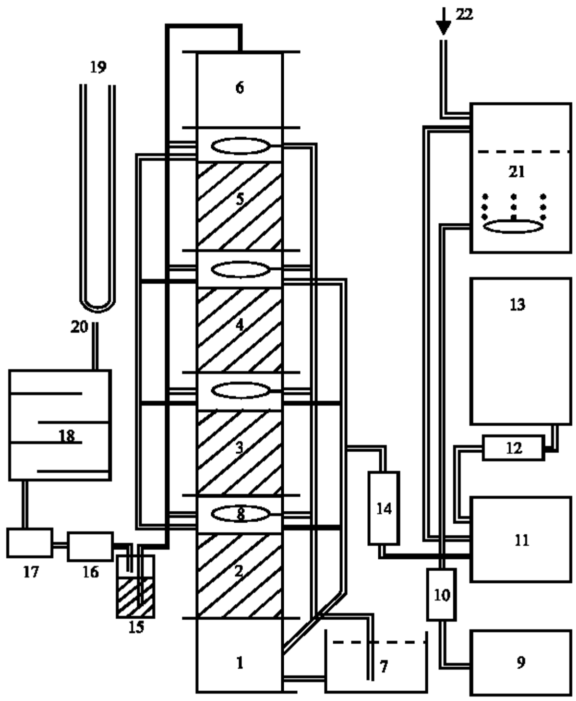

Fig. 1: Biofilter schematic. 1 = Waste Storage, 2 = Biofilter cylinder pack of seashell, 3 = Biofilter cylinder pack of seashell, 4 = Biofilter cylinder pack of seashell, 5 = Biofilter cylinder pack of seashell, 6 = Gas collection, 7 = Irrigation pack, 8 = Watering-pot, 9 = Air compressor, 10 = Fiberglass filter, 11 = Mix tank, 12 = H_2S gas adjustment 13 = H_2S producer, 14 = Flow meter, 15 = Humidity meter, 16 = Concentration of the outlet H_2S pollutant mg m^{-3}, 17 = Vacuum pump, 18 = Scrubber, 19 = Manometer, 20 = Out flow clean air, 21 = Electric heater is available to heat the water, 22 = Inflow water

RESULTS AND DISCUSSION

Since there are different variables such as; air inflow, moisture content, degree of heat and concentration of pollutant gas, in each series of experiment one of these variables changes and the rest are kept comparatively in a steady condition. Then by mixing common instances the best condition is revealed.

During 15 weeks, the concentration of inlet pollutant gas has been changed to different levels in six groups. The highest efficiency during this time is shown in Table 1.

According to the results in Table 1, the highest removal efficiency of H_2S gas in 15 weeks has been selected and the 550 samplings are summarized in Table 1. Also, the carrier gas inflow in 6 groups during 15 weeks is measured on seashell bed. The average flow rates are; less than 2, 2-4, 4-6, 6-8, 8-10 and more than 10 L min^{-1}. The highest removal efficiency in 15 weeks for different groups is shown in Table 2.

According to the process of changes in concentration of pollutant gas and carrier gas flow rate, the outcome of these changes is shown in Fig. 2-4.

According to the results from Table 1 which indicate the analysis of 550 sample readings in different situations in 15 weeks it is that the best removal efficiency is approximately between 11th to 15th week. Also the results shown in Table 1 indicate that the concentration of the pollutant gas does not have an important effect on gas treatment and the removal efficiency goes up to 78-88% when the micro-organisms are in desirable condition. If moisture is 77 to 93% and temperature is between 20.5 to 30°C their effect on removal efficiency, growth and reproduction of the *Thiobacillus thioparus* bacteria will be the same. Considering the seashell porosity and calculating of real retention time of gas in seashell bed, the real retention time is changeable between 0.3-2 min depending on the carrier gas flow.

By studying the experiment results and considering the cylinders' dimensions and the porosity of the bed, the best real retention time is 0.4 min. Also the pores in seashell bed won't clog because of bacteria reproduction. This process can be monitored by measuring head loss at the inlet and outlet of the biofilter cylinders by a manometer. As shown in Fig. 2, removal efficiency in all various concentrations increases after seven weeks passed since microbial adaptation with environmental conditions. This ascending procedure continues to 15th week. Also by comparing various H2S concentrations in Fig. 3, it specifies that the concentration of emitted gas decreases up to the 15th week if stable conditions of pH controlling and aeration rate are established.

Finally, by studying Fig. 4 in which the relation of inflow and outflow on time parameter is assessed, with the passing of time, fraction of inflow/outflow remains the same up to week eight and after that the increase of pollutant concentration inflow into the system, the outflow rate remains the same and standard. This mathematical model is used to calculate the real retention time in the bed:

$$\tau = \frac{V_f \times \theta}{Q}$$

Where:

τ = Real retention time (min)

Table 1: Comparing the best removal efficiency on seashell bed within 15 weeks with different concentration of pollutant gas

Row	Time (week)	Inflow concentration (mg m^{-3})	Average flow rate (L min^{-1})**	Moisture average (%)	Heat average (°C)	Real retention time in bed (min)	Removal efficiency (%)*	Mass loading per volume (g m^{-3} min^{-1})	Biomass average treatment capacity (g m^{-3} min^{-1})
1	11th	<20	8.200	77	23.0	0.4073	77.77	0.0278	2.1654
2	14th	20-40	6.500	90	30.0	0.5160	82.55	0.0380	3.1970
3	15th	40-60	7.500	86	28.0	0.4450	88.13	0.0830	7.3560
4	14th	60-80	8.000	86	30.0	0.4450	81.37	0.1030	8.1110
5	13th	80-100	7.125	88	29.2	0.4980	85.29	0.1250	10.6680
6	15th	100<	9.660	85	30.0	0.3510	82.47	0.2170	17.9130

*No. of samples was 550 and for each different concentration there were at least 6 readings at different times. **Average flow rate measured at different times

Table 2: Comparing the best removal efficiency on seashell bed within 15 weeks with different carrier gas inflow

Row	Time (week)	Average flow rate (L min^{-1})	Inflow concentration (mg m^{-3})**	Outflow concentration (mg m^{-3})	Inflow/ outflow ratio	Moisture average (%)	Heat average (°C)	Real retention time in bed (min)	Removal efficiency (%)*	Mass loading per volume (g m^{-3} min^{-1})	Biomass average treatment capacity (gm^{-3}min^{-1})
1	9th	<2	60.00	12.33	4.86	86	20.5	2.163	79.44	0.019	1.442
2	10th	2-4	62.40	13.05	4.78	93	21.8	1.058	79.08	0.041	3.196
3	13th	4-6	99.66	12.23	8.14	92	26.0	0.748	87.72	0.082	7.296
4	14th	6-8	76.60	7.95	9.63	92	30.3	0.487	89.62	0.103	9.436
5	14th	8-10	100.90	17.28	5.83	90	30.7	0.410	82.87	0.155	12.706
6	15th	10<	98.80	23.29	4.24	87	29.6	0.316	76.42	0.197	15.207

*No. of samples was 550 and for each different concentration there were at least 6 readings at different times. **Average flow rate measured at different times

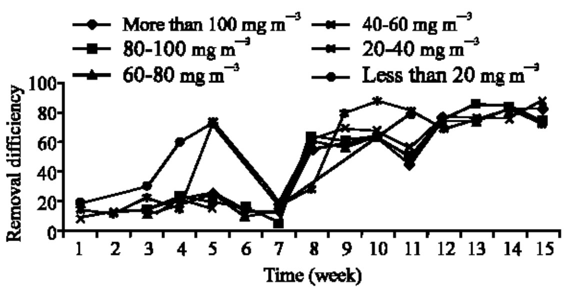

Fig. 2: The relation between time (week) and H_2S removal efficiency on seashell beds for the concentrations of less than 20 mg m^{-3} to more than 100 mg m^{-3} of pollutant gas

V_f = Biofilter volume (m^3)
θ = Porosity, or dividing the volume of empty cylinder on total volume
Q = Air flow intensity

To calculate mass loading per volume (volumetric) which is the amount of destroyed pollutant mass per volume of biofilter bed per time unit, the following formula is used:

Mass loading per volume (volumetric) = Biomass average treatment capacity × Removal efficiency

$$\text{Mass loading (volumetric)} = \frac{Q \times C_i}{V_f}$$

Where:
C_i = Inflow gas concentration (g m^{-1})

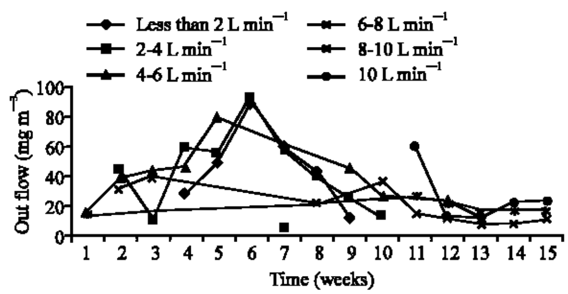

Fig. 3: The relation between time (week) and concentration of outflow pollutant gas from biofilter cylinder on seashell bed with different flow rate of less than 2 L min^{-1} to more than 10 L min^{-3}

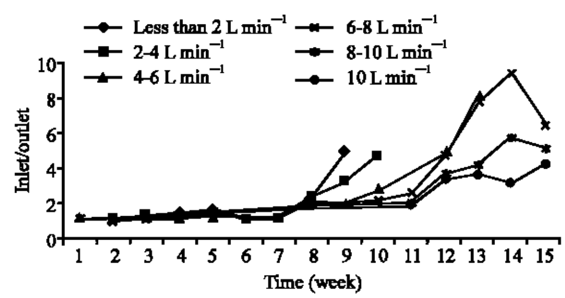

Fig. 4: The relation between time (week) and the ratio of (inflow/outflow) pollutant gas concentration from seashell bed biofilter

By comparing the results from Table 1 and 2 it shows that if the amount of mass loading is between 0.1 and 0.2 g m^3 min^{-1}, the most removal efficiency is reached.

Also the biomass average treatment capacity with numbers 12 to 17 gives the best removal efficiency.

In Melvin's research (2003) a mixture of compost and fiberglass was used instead of seashell bed and after 64 days, efficiency decreased because the porous spaces were clogged. Therefore using seashell bed seems more useful.

Also in Guillermo *et al.* (2005) investigations coconut shells were used to fill the cylinders and with the efficiency of 90% they could remove up to 280 mg L^{-1} of H_2S gas in 40 days. In similar researches done by Smet *et al.* (2000) and Jiafa *et al.* (1997), the filling material for cylinders were ceramics and compost. The removal efficiency was over 90%, but after almost 2 months it decreased because of accumulation of sulfide crystals on the bed surface. Therefore the filling material in the cylinders should be mixed constantly which is expensive in industrial planning.

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