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Removal of Hydrogen Sulfide Gas using Biofiltration - a Review

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Abstract

Hydrogen sulfide (H_2S) is extremely toxic to living organisms and plants. H_2S gas contamination may be treated by both chemical and physical methods but they have high capital costs, demand large energy inputs and result in the generation of secondary hazardous wastes. Biofiltration, a biological technique, has significant economic advantages over other air pollution control technologies. Biofiltration is a process by which contaminated gases pass through the biofilter and pollutants are transported into the biofilm where they are utilized by microbes as a carbon source, an energy source. *Thiobacillus* sp. is the most frequently used microbial species in H_2S biofiltration and can degrade H_2S for energy and produce sulfate or sulfuric acid. Moreover, media selection for biofiltration (combing both natural and synthetic media) is an important step towards the development of a successful biofiltration operation. In addition, the optimization parameters of a biofiltration operation are found. First, optimal moisture content may vary from 20 to 60 wt%. Second, most microbial growths occur near neutral pH and wide deviation from these levels will impact the efficiency of the biofiltration. Third, the optimum temperature of biofiltration is near the optimum temperature for microbial inoculation based on removal efficiency. Finally, because nutrient supply is less critical as H_2S removal requires few nutrients, commercial fertilizer or secondary effluent from wastewater treatment plants can be used for humid and nutrient supply. Many biofiltrations are designed for H_2S control.

Keywords: Biofiltration, hydrogen sulfide, hydrogen sulfide removal

Introduction

Hydrogen sulfide (H_2S) is extremely toxic; it can cause injury to the central nervous system even at 10 ppm [1]. It is toxic to microorganisms, and also corrosive to concrete and steel [2,3]. This gas is produced by various industrial processes, including wastewater treatment, food processing, petroleum refining, drug manufacturing, paper and pulp manufacturing, and solid waste processing. These are the main causes of global environmental problems such as air pollution and acid rain. Acid-mine drainage is also a major environmental problem in terrains affected by untreated acidic waters [4].

H_2S contamination may be treated by biochemical, chemical and physical methods [5]. A number of physicochemical processes such as dry gas reduction-oxidation (redox) process, liquid redox processes and liquid adsorption process are usually employed for desulphurization of gases containing H_2S. However, they have high capital costs, demand large energy inputs and result in the generation of secondary hazardous wastes [6]. Several physical means of controlling the formation of acid-mine drainage have been developed but they have not been very successful. Therefore, efforts have been directed towards biological processes for the removal of contaminants, which are characterized by small capital costs and low energy requirements.

With regard to this biological technique, biofiltration is a process that utilizes microorganisms growing or immobilised on an organic porous support. The organic medium acts as a physical support for the active biomass and in some cases provide nutrients for growth. The contaminated gaseous stream passes through the filter bed. The bed material absorbs biodegradable volatile compounds and the microorganisms degrade them into less harmful compounds [7]. Biofiltration has significant economic advantages over other air pollution control technologies. For example, this technique requires a relatively low initial capital outlay and minimal operating costs [8-10]. Moreover, studies have shown that biofiltration can remove more than 99 % of the H_2S in the air [5]. Thus, biofiltration of H_2S is both economical and effective, prompting numerous researchers to seek optimized biofiltration methods and reagents.

Moreover, biofiltration is a biological waste gas treatment system that provides high porosity, high nutrient availability, high moisture retention capacity, and high buffering capacity to sustain microbial growth on a suitable support media [11,12]. The efficiency of any biofiltration process depends on the temperature, moisture content, pH level, flow rate, surface loading rate and the physical structure of the biofiltration [13].

Therefore, this paper reviews current biofiltration process for hydrogen sulfide removal and investigates research imperative for potential process improvement.

Mechanisms used in biofiltration

Biofiltration is a complex process with many physical, chemical, and biological phenomena [14] and has recently been recognized as one of the most popular and efficient technologies for odor treatment [15]. A typical biofiltration process consists of 2 steps. Firstly, the pollutant is transferred from the air stream into the liquid and adsorbed on a solid medium. The pollutant is then biodegraded by microbes living in the liquid phase or on the packing material [16]. A general biofiltration process may include the elements illustrated in **Figure 1**. Examples of biofiltration for the removal of H_2S include:

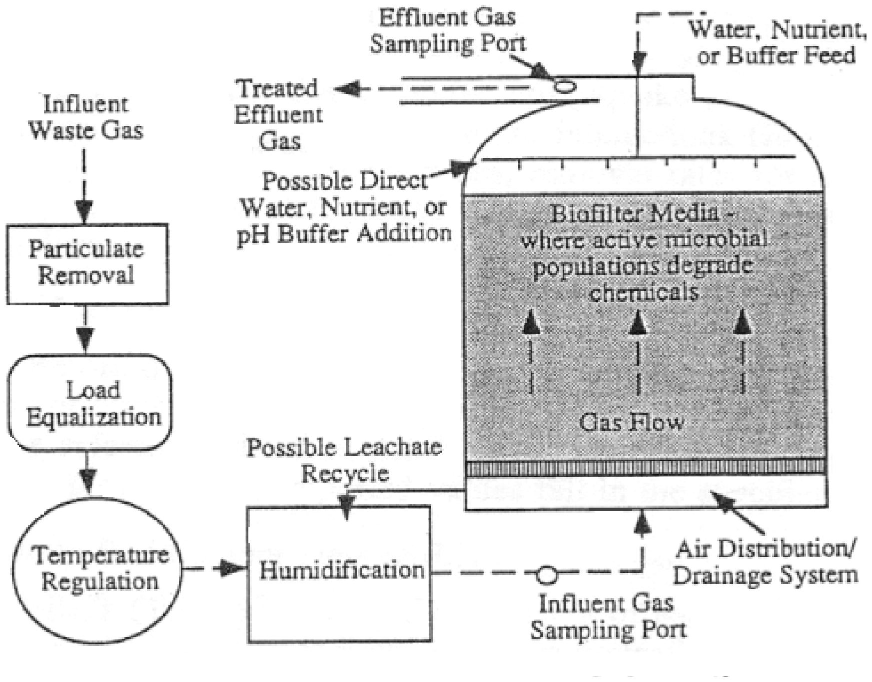

Figure 1 Biofiltration system schematic.
Source: Swanson and Loehr [36]

Chung *et al*. [17] studied a novel biofiltration system to control H_2S emission using *Thiobacillus thioporus* CHll, which were immobilized with Ca-alginate to produce pellet packing materials for a novel biofilter system that controls H_2S emission. It was found that H_2S removal efficiency was greater than 98 %.

Rattanapan *et al*. [15,18] studied a biofiltration system with a pure culture of sulfur oxidizing bacteria immobilized on granular activated carbon (GAC) for H_2S removal. The results found that the efficiency of H_2S removal was more than 98 % even at high concentrations (200 - 4,000 ppm) and the maximum elimination capacity was about 125 g H_2S/m^3 of GAC/h.

Microorganisms used in biofiltration

H_2S may be degraded by microorganisms in 3 different ways: assimilation, mineralization, and sulfur oxidation [19]. H_2S uptake rate based on the assimilation process is far too low to achieve reasonably high removal efficiencies from a highly loaded waste gas stream. The pathway for microorganisms to degrade H_2S by oxidation results in the release of energy and produces sulfuric acid. The most energy is released when sulfide is oxidized completely to sulfate as seen in Eq. (1). Sulfide oxidation often occurs in steps with elemental sulfur as an intermediate product, as seen in Eq. (2) - (4) [20], and in oxygen-limited environments, oxidation may proceed only to elemental sulfur, producing less energy. Cells can either deposit sulfur inside or outside their cell membranes. Other reduced sulfur compounds, such as thiosulfate, can also be oxidized also releasing energy as seen in Eq. (4) [21].

$$H_2S + 2O_2 \rightarrow SO_4^{2-} + 2H^+ \qquad (\Delta G^{0'} = -798.2\ kJ/rxn) \qquad (1)$$
$$HS^- + \tfrac{1}{2}O_2 + H^+ \rightarrow S^0 + H_2O \qquad (\Delta G^{0'} = -209.4\ kJ/rxn) \qquad (2)$$
$$S^0 + H_2O + 1\tfrac{1}{2}O_2 \rightarrow SO_4^{2-} + 2H^+ \qquad (\Delta G^{0'} = -587.1\ kJ/rxn) \qquad (3)$$
$$\tfrac{1}{2}S_2O_3^{2-} + \tfrac{1}{2}H_2O + O_2 \rightarrow SO_4^{2-} + H^+ \qquad (\Delta G^{0'} = -409.1\ kJ/rxn) \qquad (4)$$

The desirable bacteria to be used in a biofiltration to convert H_2S to S_0 should possess the following basic features: reliable capability of converting H_2S to S_0, minimum nutrient inputs, and easy separation of S_0 from the biomass. The chemolithotrophic sulfide oxidizers (also referred to as colorless sulfur bacteria) have diverse morphological, physiological and ecological properties, and are able to grow chemolithotrophically on reduced inorganic sulfur compounds such as sulfide, sulfur and thiosulphate and in some cases organic sulfur compounds like methanethiol, dimethylsulfide and dimethyldisulfide [22].

The sulfur bacteria encompass many genera such as *Thiobacillus*, *Acidithiobacillus*, *Achromatium*, *Beggiatoa*, *Thiothrix*, *Thioplaca*, *Thiomicrospira*, *Thiosphaera*, and *Thermothrix* to name a few. The genus *Thiobacillus*, one of the most studied groups, consists of several gram-negative and rod-shaped species which utilize oxidation of sulfide, sulfur and thiosulfate for generation of energy and growth [23]. Bacteria from the genus *Thiobacillus* seem to have better H_2S removal efficiency than other species of sulfide oxidizing bacteria because of smaller nutritional requirements, and have the ability to grow using H_2S as an energy source [4]. *Thiobacillus* species is the most frequently used bacteria in H_2S biofiltration [17,24-27]. *Thiobacillus* sp. uses only carbon dioxide as its carbon source when biodegrading H_2S. No typical carbon source has been found to have an inhibitory effect on its growth [28] and is thought to account for most sulfide oxidation, via the sulfite-oxidase pathway as described in **Figure 2**. These bacteria have an ability to grow under various environmental stress conditions such as acidic conditions. They include both acidophobic bacteria that prefer a pH near 7 and acidophilic bacteria that grow at low pH values, allowing efficient H_2S oxidation over a wide pH range. These points show that this genus seem to be better because of smaller nutritional requirements, and allow the decrease or even the elimination of the initial lag phase and the increase in the efficiency of H_2S removal, maintaining a stable operation [26].

Moreover, most research has studied H_2S removal using a pure culture immobilized on a carrier [17,29-31]. Although pure culture inoculation has drawn great attention because it can shorten the start-up time and increase the removal efficiencies [32], there are limitations on employing pure cultures for industrial applications. Therefore, some researchers have used a mixed culture or consortium from compost [33] and wastewater treatment plant sludge to remove H_2S gas [16,34]. However, there is still a lot of microbial diversity in different sources but the consortium require at least 3 weeks for acclimation [16,33-34]. In addition, various microorganisms for H_2S removal have never been reported and there is no comparison in the reported literature about the performance of biofiltration inoculated with pure culture and a consortium for the biofiltration removal of H_2S. Comparison between pure and mixed culture for H_2S removal, therefore, will be novel and interesting research.

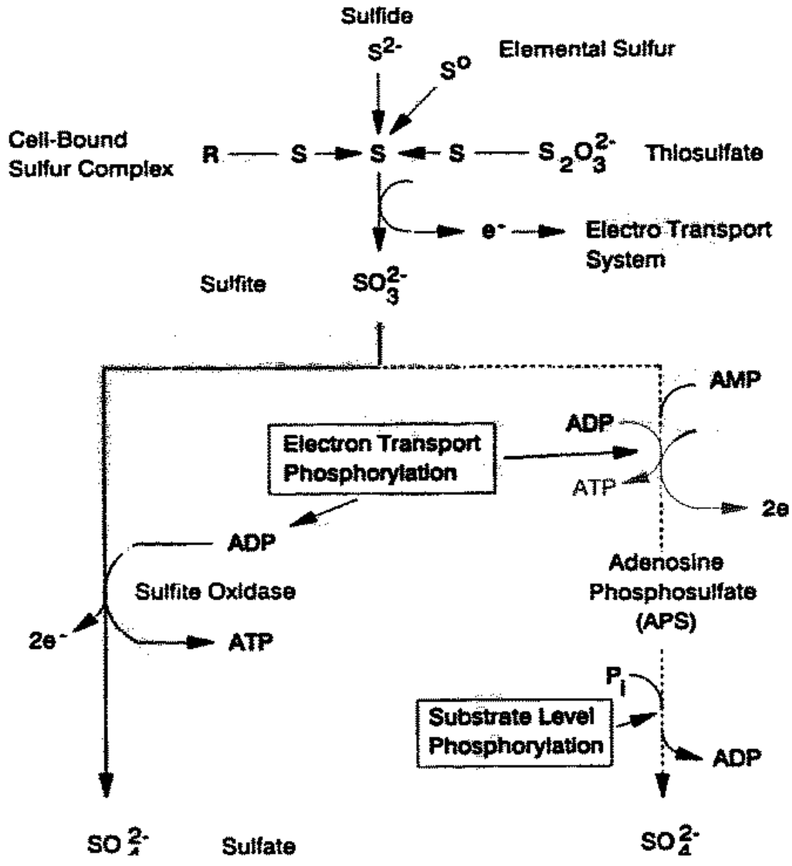

Figure 2 Steps in the oxidation of sulfur compounds by *Thiobacillus* species.
Source: Yang [20]

Packing media used in biofiltration

Selecting optimum packing media for biofiltration is an important step towards the development of successful biofiltration operations, and the quality of the biofiltration medium has been reported as one of the key factors in biofiltration performance [35]. A good filter should have a large surface area, high water retention capacity without becoming saturated, low bulk density, high porosity, structural integrity, and a buffer capacity towards acidification and high contaminant loads [36,37]. Many media used in biofiltration have included natural materials. Soil, the most commonly used medium tends to short-circuit and clog and limits its effectiveness [38]. Compost [2,39-40] has good water retention properties, a large density of microorganisms, and a suitable organic content. However, it suffers from aging effects that create short-circuiting of the biofilter and further decrease its effectiveness [41]. Peat as a packing material has been demonstrated to be preferable to soil or compost, but it is naturally hydrophobic, and moisture in the peat beads is difficult to control [26,42]. Wood bark [43], wood chips [44], lava rock [45] and synthetic materials such as ceramic saddles [46], polyethylene pall rings [47], synthetic foams [48], activated carbon [15,18,49-51], extruded diatomaceous earth pellets [52], glass beads [53] and Ca-alginate [17,54] have also been used as media for biofiltration.

Natural organic packing materials generally have an advantage over synthetic media in providing nutrients for microbial growth. Among the operational advantages of using natural organic materials are the presence of complex microbial communities capable of degrading several pollutants, a high water retention capacity and available organic matter and nutrients for microorganisms [14]. A number of different natural support media have been studied for biofiltration applications [55] and it has also been proposed that biofiltration using natural support media is one of the most cost effective treatment methods compared with other alternative technologies. Although they have been used successfully in many applications, several common problems appear over the operation time as a result of packing deterioration and biomass accumulation. The uncontrolled delivery of nutrients [56] may cause problems including medium clogging and channeling, excessive pressure loss of gas flow, and the occurrence of secondary and co-metabolic reactions. Filter beds of synthetic materials are nowadays also being used. Nevertheless, it has been found that during long-term operation of a compost biofiltration, the nutrient availability may limit biofiltration performance [56]. Nutrient limitation may also occur in biofiltration using other natural media for odor removal during long-term operation. On the other hand, the use of synthetic packing media requires proper seeding with nutrient, moisture and organisms. Hence, there is a current tendency to investigate the use of synthetic materials as support media, which are physically and chemically more stable. Material porosity is a key feature of the biofiltration bed, and the presence of a compact network of micropores was initially thought to be indispensable for a proper water holding capacity. In order to combine the advantage of both natural and synthetic media, it is common to use a mixed packing bed containing two types of material. For example, Ergas *et al.* [58] used filter media consisting of air-dried compost, perlite and crushed oyster shell. The compost consisted of 50 % digested sewage sludge and 50 % forest products. Perlite increased the porosity of the bed and the oyster shell provided calcium carbonate as a pH buffer. Shareefdeen and Baltzis [59] used peat mixed with polyurethane foam, vermiculite and perlite for biofiltration of methanol vapor because of the large surface it provides for microbial adhesion and minimal pressure drop.

Parameters affecting biofiltration

The most important parameters to control are moisture, pH, nutrients, and temperature. To ensure stable performance over long operation periods, some key operating parameters need to be carefully controlled. Typical operation conditions of biofiltration for H_2S treatment are shown in **Table 1**. More detailed descriptions of their key parameters are given below.

Table 1 Operational conditions of biofiltration for waste air treatment.

Parameters	Operational conditions
Bed height	1 ~ 1.5 m
Cross section area	1 ~ 3,000 m^2
Waste air flow	50 ~ 3 × 10^5 m^{-3}·h^{-1}
Surface loading	5 ~ 500 × 10^5 m^3·m^{-2}·h^{-1}
Bed void volume	50 %
Operational temperature	15 ~ 30 °C
Inlet air relative humidity	> 98 %
Water content of the support material	60 % by mass
pH	pH 6 ~ 8 (support material)
Typical removal efficiencies	60 ~ 100 %

Source: Denvinny *et al* [14]

Moisture is essential for the survival and metabolism of the resident microorganisms and contributes to the filters buffer capacity [60]. Williams and Miller [3] identified bed moisture content as the single most important parameter for biofiltration viability. Optimal moisture content varied from 20 to 60 wt% in their review of operational biofiltrations. Heat generated by biological activity in biofiltration may increase the temperature of the bed medium above that of the inlet gas. Even if the gas enters the biofilter saturated with water, it will become unsaturated as its temperature rises after being contacted with the bed medium. Hence it is important to supply 100 % humid air to the biofilter and/or irrigate the bed periodically to compensate for moisture loss so as to maintain the viability of the organic bed. Conversely, too much moisture leads to a slow mass transfer of odorous compounds into the biofilm and anaerobic zones, where oxygen required for biooxidation is depleted. For this reason, the capacity of the soil bed to remove odor significantly when they become too wet is reduced. Excessive moisture will also result in an increased pressure drop through the packed bed.

pH. Most microorganisms prefer a specific *pH range*. Hence, a change in pH can strongly affect their activities. Each species of microorganism is most active over a certain pH range and will be inhibited or killed if conditions deviate from this optimal range. Most biological growth occurs near a neutral pH and a wide deviation from their optimum levels will impair the efficiency of biofiltration. A notable exception is sulfide oxidizing bacteria which thrive at a low pH though its growth will be inhibited when the system pH is lower than a certain threshold. In biofiltration treating sulfur containing gases, sulfate and hydrogen ions will be produced and thus there will be a change in the system pH during long term operation. For instance, crushed oyster shells and maerl (a mineral that supports marine organics which contains 82 % calcium carbonate) were used extensively as a source of alkalinity for buffering and as a carbon source for autotrophic bacteria responsible for nitrification and sulfide oxidation [58,61]. In addition, many bacteria have their pH optimum between 6 and 8 [55,62], but H_2S can also be oxidized at acidic pH by microorganisms like *Thiobacillus* [63], *Acidothiobacillus, Beggiatoa* [64], *Sulfolous* [65].

Temperature is also one of the most important variables in determining microbial growth rates and the types of species present in a microbial community [35]. For successful operation, the temperature of a system should remain relatively constant. The temperature of biofiltration is mainly influenced by the temperature of the inlet air stream and somewhat by the exothermic biological reactions in the bed [63]. As the temperature increases, the metabolic and cell growth rates increase, but the sorption decreases [66]. However, above a certain critical temperature, inactivation of certain key proteins and an abrupt cessation of growth occur [68]. The optimal temperature for various species range widely, but most biofiltration applications operate at temperatures in the mesophilic range (20 - 45 °C), with 35 - 37 °C often noted as the optimal temperature [37]. More recently, some studies of thermophilic operations (45 - 75 °C) have also been published [69].

Nutrient supply. Carbon and energy from the degradation of contaminants and *nutrients* such as nitrogen, phosphorous, and trace elements are required for microbial growth [37]. In biofiltration, nutrients can be supplied with the humidification system or packing can be periodically soaked in a nutrient solution. Organic packing media in biofiltration such as compost have enough mineral nutrients and do no need extra nutrient supply. However, the rate of media degradation can be too slow to support effective biodegradation of the target air pollutant [70]. Media may also be depleted of nutrients during long term operation. Nutrient limitation has been reported in compost packed biofiltration treating hexane during a 3 month operation [57]. Inorganic and synthetic media, such as lava rock, plastic rings, or ceramic carriers do not have an appropriate supply of nutrients. If this type of medium is used, additional nutrients must be added to the biofiltration bed. Usually N, P and K are added in the form of commercial fertilizer [14] or secondary effluent from a wastewater treatment plant [56]. The nutrient issue in general is important for volatile organic compound control. For H_2S control, nutrient supply is less critical as H_2S requires few nutrients due to sulfide oxidizing bacteria being used to using H_2S as a source of energy [71,72]. Hence in many biofiltrations that are designed for H_2S control, secondary effluent can be used.

Conclusions

This review considers the preferred treatment method for H_2S gas. In the case of H_2S, biofiltration methods involving sulfide oxidizing bacteria provide the inherent advantage of maintaining the H_2S gas. Nevertheless, biofiltration is a complex process with many physical, chemical, and biological phenomena. The contaminated gases pass through the reactor, and pollutants are transported into the biofilm where they are utilized by microbes as a carbon source, an energy source or both. *Thiobacillus* sp. is the sulfide oxidizing bacteria and the most frequently used bacteria species in H_2S biofiltration. It occurs naturally in the sanitary sewer system and wastewater treatment plant, and quickly converts H_2S to sulfate in a low growth intensive manner.

Moreover, the choice of media for biofiltration is an important step towards the development of a successful biofiltration operation. Hitherto, media used have included natural materials, which have a general advantage over synthetic media in providing nutrients for microbial growth. The use of synthetic packing media is necessary because of low head losses due to larger interstices between packing granules or pieces, larger specific surface areas, and solid phase adsorption of contaminants. On the other hand, the use of solely inert synthetic packing media requires proper seeding with nutrient, moisture and organisms. Therefore, biofiltration with a continuous moisture and nutrient supply, is usually used when using an inert synthetic packing material. In order to combine the advantage of both natural and synthetic media, it is common to use mixed packing beds containing 2 types of material.

Finally, system moisture, pH, nutrients and temperature may all affect biofiltration operation. Optimal moisture content varies from 20 to 60 wt%. pH is specifically preferred for various microorganisms. Most biological growth also occurs near a neutral pH and a wide deviation from this level impairs the efficiency of biofiltration. The optimal temperature for biofiltration, therefore, is near the optimal temperature for microbial inoculation based on removal efficiency. Nutrients such as nitrogen, phosphorous, and trace elements are required for microbial growth. Usually N, P and K are added in the form of commercial fertilizer or secondary effluent from wastewater treatment plants because nutrient supply is less critical as H_2S requires few nutrients. Hence, in many biofiltrations that are designed for H_2S control, secondary effluent is used.

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