Biological technologies for the removal of sulfur containing compounds from waste streams: bioreactors and microbial characteristics

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Abstract Waste gases containing sulfur compounds, such as hydrogen sulfide, sulfur dioxide, thioethers, and mercaptan, produced and emitted from industrial processes, wastewater treatment, and landfill waste may cause undesirable issues in adjacent areas and contribute to atmospheric pollution. Their control has been an area of concern and research for many years. As alternative to conventional physicochemical air pollution control technologies, biological treatment processes which can transform sulfur compounds to harmless products by microbial activity, have gained in popularity due to their efficiency, cost-effectiveness and environmental acceptability. This paper provides an overview of the current biological techniques used for the treatment of air streams contaminated with sulfur compounds as well as the advances made in the past year. The discussion focuses on bioreactor configuration and design, mechanism of operation, insights into the overall biological treatment process, and the characterization of the microbial species present in bioreactors, their populations and their interactions with the environment. Some bioreactor case studies are also introduced. Finally, the perspectives on future research and development needs in this research area were also highlighted.

Keywords Air pollution control · Bioreactors · Sulfur containing compounds · Microbial characterization · Desulfurization bacteria

Sulfur-containing off-gases are usually generated and emitted from industrial processes, wastewater treatment, and waste disposal in landfills (Zhang et al. 2015). Hydrogen sulfide (H₂S), methyl mercaptan, dimethyl sulfide, dimethyl disulfide, carbonyl sulfide and carbon disulfide are frequently present in exhaust gas, which creates ecological and health hazards when discharged into the environment (Smith et al. 2001). Several desulfurization techniques for effluent gases have been developed to control the pollution generated by sulfur-containing compounds in order to meet emission standards. These techniques include gas-phase conversion, liquid absorption, and solid adsorbent adsorption (Atanes et al. 2012). Compared with conventional physicochemical methods, biological technologies are more widely applied to purify waste gases with large flow rates and moderate concentrations due to their high efficiency, cost effectiveness, convenient operation, fewer by-products or residuals and environmental acceptability (Fig. 1) (Estrada et al. 2012; Ralebitso-Senior et al. 2012).

This paper provides an overview of the current biological techniques used for the treatment of air streams containing sulfur compounds as well as the novel techniques being developed at the moment. The discussion focuses on bioreactor characteristics and application area, along with microbial characterization of the species present, their
interactions with the environment, and insights into the overall biological treatment process. Some bioreactor case studies are also introduced.

**Bioreactors for sulfur compounds removal**

There are three categories of traditional air phase bioreactors: biofilters, bioscrubbers, and biotrickling filters (Deshusses 1997). To date, newly developed methods have focused on coping with hydrophobic or less biodegradable compounds and attenuation of fluctuating loads. Among them, fungal bioreactors, thermophilic bioreactors, combined-bioreactors, and bioreactors integrated with physicochemical techniques have been reported for the elimination of sulfur compounds from waste streams (Table 1). Most of practical experiences involve full scale traditional bioreactors, while new type bioreactors seem to be less popular in sulfur compounds removal.

**Traditional bioreactors**

**Biofilters**

Biofiltration is the oldest biotechnology used for removal of undesired off-gas components. In the early 1920s, biofilters had already been applied to control hydrogen sulfide emissions from wastewater treatment plants (Van Groenestijn and Hesselink 1993). Biofilters are reactors that usually consist of a gas pre-humidity facility and a filter bed packed with porous materials, on which a mixed culture of pollutant-degrading microorganisms is immobilized as biofilm. The gases to be treated are forced through the packed bed after pre-humidification (Fig. 2). Biodegradable compounds are transported from the gaseous phase to the microbial biofilm through a liquid or moisture phase and subsequently biologically oxidized into less harmful substances. Bench and pilot scale studies have shown that a wide variety of pollutants in industrial and municipal exhaust streams that contain organic and inorganic sulfur compounds, such as hydrogen sulfide (Morales et al. 2012), sulfur dioxide (Philip and Deshusses 2003; Zhang et al. 2015), and volatile organic sulfur (Ho et al. 2008), can be successfully treated with biofiltration (Devinny et al. 1999).

**Bioscrubbers**

A bioscrubber consists of two separate units: an absorption unit with inert packing materials and a bioreactor unit with suspended microbial strains in the aqueous phase (Fig. 3, Delhomenie and Heitz 2005). Nutrients essential for microorganism growth and maintenance are supplied to the liquid periodically. The contact between input gaseous contaminants and water is achieved in the absorption unit, resulting in absorption and transfer into the liquid phase. Currently, gas and liquid phases were counter-flow within the absorption unit. The purified gases are released at the top of the unit, while the liquid with the dissolved target compounds is subsequently pumped to and treated in the bioreactor unit. The transfer surface between the sulfur compounds and the aqueous phase can be increased via the addition of inert packing materials in the absorption unit (Van Groenestijn and Hesselink 1993). Part of the treated solution in the bioreactor can be recycled to the spraying tower for sulfur compound absorption. The liquid can be discharged or refreshed to remove undesired products. While it is easier to monitor and control temperature, pH and ionic strength in bioscrubbers, a lower specific gas/liquid surface area is the drawback of this kind of bioreactor. This restricts the field of bioscrubber application to sulfur dioxide or hydrogen sulfide with low Henry coefficients in order to avoid high spray columns and large water flows (Potivichayanon et al. 2006; Tosati and Jinsiriwanit 2013).

**Biotrickling filters**

Biotrickling filters can be regarded as an intermediate between biofilters and bioscrubbers. Here, waste gas is carried through a column packed with inert materials covered with biofilm, which is continuously irrigated and recirculated with an aqueous solution containing essential nutrients required by the biological system (Fig. 4). Without a separate bioreactor for regeneration, absorption and biodegradation of the target compounds are combined in one column, allowing for easier control of reaction conditions. Biotrickling filters have a lower specific surface
Table 1: Bioreactors for sulfur containing compounds removal

| Bioreactor type | Target pollutant | Source | Scale | C_{in}/loading | RE/EC | EBCT (s) | References |
|-----------------|------------------|--------|-------|----------------|-------|---------|------------|
| Biofilter       | H₂S              | Lab-scale | 303–6071 mg m⁻³ | >95 % | 99 % | 29–152 | Rattanapan et al. (2010) |
|                 | H₂S              | Lab-scale | 227–1136 mg m⁻³ | 99 % | 99 % | 60 | Omri et al. (2011) |
|                 | H₂S              | Pilot-scale | 3.2 g m⁻³ h⁻¹ | >99 % | 50 | Van Groenestijn and Hesselink (1993) |
|                 | H₂S              | Pilot-scale | 300 m³ h⁻¹ | >99 % | 60 | Lebrero et al. (2010) |
|                 | EtSH; DMDS; DMS | Lab-scale | 3.8–7.6 mg m⁻³ | 82 % (H₂S); 90–97 % (DMDS and EtSH) | 99 % | 384 | Jaber et al. (2014) |
|                 | EtSH; DMS; DMS  | Lab-scale | 0.18 mg m⁻³ (H₂S), 0.85 mg m⁻³ (EtSH), 0.22 mg m⁻³ (DMS) | 1.49 × 10⁻² g m⁻³ h⁻¹ (H₂S), 7.09 × 10⁻³ g m⁻³ h⁻¹ (EtSH), 1.85 × 10⁻² g m⁻³ h⁻¹ (DMS) | 60 | Li and Liu (2004, 2009) |
| Bioscrubber     | H₂S              | Pilot-scale | 0.4–2.5 g m⁻³ h⁻¹ | 100 % | 60 | Hort et al. (2009) |
|                 | H₂S, MM          | Rendering plant | 0.05–5 mg m⁻³ | 98 % | 113 | Anet et al. (2013) |
|                 | DMS              | Lab-scale | 0.484 g m⁻³ h⁻¹ | 62–74 % | 30 | Giri et al. (2010) |
|                 | DMS              | Lab-scale | 9.4 g m⁻³ h⁻¹ | 100 % | 30 | Shu and Chen (2009) |
|                 | DMS              | Pilot-scale | 9.4 g m⁻³ h⁻¹ | 100 % | 30 | Giri and Pandey (2013) |
|                 | MeH              | Lab-scale | 303 mg m⁻³ | 98 % | 12.71 | Potivichayanon et al. (2006) |
|                 | MeH              | Lab-scale | 32–326 mg m⁻³ h⁻¹ | 80 % at 37 g m⁻³ h⁻¹ | 63 | Tosati and Jinsiriwanit (2013) |
| Biotrickling filter | H₂S          | Lab-scale | 3.8–27.3 mg m⁻³ | 98 % | 600 | Baspinara et al. (2011) |
|                 | H₂S              | Pilot-scale | 83–916 g m⁻³ h⁻¹ | 95 % | 60 | Baspinara et al. (2011) |
|                 | H₂S              | Lab-scale | 30–238 mg m⁻³ | 97.7 ± 0.3 % | 16 | Solcia et al. (2014) |
|                 | H₂S              | Lab-scale | 84 g m⁻³ h⁻¹ | 97.7 ± 0.3 % | 120 | Fortuny et al. (2011) |
|                 | H₂S              | Pilot-scale | 0.59–5 g m⁻³ h⁻¹ | >99 % | 25 | Chen et al. (2014) |
|                 | TAS              | Lab-scale | 1.2 × 10⁻³ mg m⁻³ | 100 % | 66 | Li et al. (2015a, b) |
|                 | EtSH             | Lab-scale | 1.05 × 10⁻³ mg m⁻³ | 100 % | 110 | An et al. (2010) |
|                 | MM               | Lab-scale | 160–192 mg m⁻³ | 1.8 g m⁻³ h⁻¹ | 180 | Montebello et al. (2012) |
|                 | DMS              | Lab-scale | 407 mg m⁻³ | 90 %/8.3 g m⁻³ h⁻¹ | 120 | Sercu et al. (2005) |
|                 | SO₂              | Lab-scale | 857–2857 mg m⁻³ | 100 % | 6 | Philip and Deshusses (2003) |
area (100–300 m² m⁻³) than biofilters, which makes them unfit for the treatment of hydrophobic compounds.

Table 2 demonstrates the advantages, disadvantages and application areas of the three basic gas bioreactors. The selection of bioreactor type relies on the load and kind of pollutant to be treated. Table 3 exhibits the physical and chemical properties of common sulfur compounds. Generally, biofilters are suitable for dimethyl sulfide, ethanethiol, and volatile organic sulfur control, while bioscrubbers and biotrickling filters are preferred for the treatment of sulfur dioxide. Hydrogen sulfide can be treated effectively by the three type bioreactors.

New biotechniques for sulfur compounds removal

Fungal bioreactors

Results on fungal bioreactors for hydrogen sulfide emission control were initially reported in 1923 (Van Groenestijn and Hesselink 1993). The search for efficient fungal biocatalysts has mainly been for the biofiltration of waste gases containing hydrophobic compounds, such as mercaptan (Zhu and Liu 2004), bis(2-chloroethyl) sulfide (Itoh et al. 1997), and dibenzothiophene (Elmi et al. 2015). Hydrophobic compounds are poorly absorbed by bacterial biofilms due to their low solubility in water. This results in decreased elimination of hydrophobic compounds in conventional biofilters, which are based on bacterial activity. In addition, biofilter operational stability is often hampered by acidification and drying out of the filter bed (Van Groenestijn and Liu 2002). Bioreactors with fungi attached to inert packing material were developed to overcome these problems (Kennes and Veiga 2004). Zhu and Liu (2004) used a fungal vapor-phase biofilter packed with cubed polyurethane foam to treat a gas stream contaminated with ethyl mercaptan, which achieved a maximum elimination capacity of 26 g m⁻³ h⁻¹ and removal efficiency of over 98 %. The components of a styrene, alpha-pinene, and sulfur compound mixture were completely degraded (>99 %) by a fungal biofilter inoculated with the species P. ostreatus (Braun-Lüllemann et al. 1997). This system withstood fluctuations in concentration and efficiency was not affected by shut down periods. Fungal biofilters can be an effective alternative to conventional abatement technologies for treating off-gases containing hydrophobic compounds, even under discontinuous loading conditions (Moe and Qi 2004).

Combined-bioreactors

Bacteria and fungi are the two dominant microbial groups in bioreactors. Bacteria are most likely to dominate under favorable conditions and have shown some advantages in
the removal of hydrophilic compounds, while fungi can take up hydrophobic compounds faster than the surface of flat aqueous bacterial biofilms (Devinnay et al. 1999). Gaseous effluents from industrial operations or waste treatment processes are often charged with a complex mixture of pollutants, including hydrophilic and hydrophobic compounds. Complex compounds may require many metabolic steps in the transformation from their original form to carbon dioxide and water, and different species may specialize in different parts of the process. It is difficult to degrade all contaminants at the same time using one type of microorganism. The combined-bioreactor was developed to cope with gases containing compounds with different water solubility. This bioreactor consists of two reaction zones, one with suspended growth bacteria and another packed with material for attached growth of fungi. Bacteria generally dominate in the suspended zone and fungi dominate in the immobilized zone, which is achieved by controlling the pH and relative humidity in the individual zones. Contaminated gases first enter the suspended zone. The water-soluble compounds are transferred from the gas phase to the liquid phase swiftly, and then absorbed and subsequently biodegraded by bacteria. The residual gases of the poor water-soluble contaminants then flow into the immobilized zone and are removed by fungi attached to packing material. As a result of the biodegradation of pollutants by bacteria and fungi in the individual bioreactor zones, hydrophilic and hydrophobic compounds, e.g., hydrogen sulfide, ethyl mercaptan, diethyl sulfide, ammonia, acetic acid and styrene, can be efficiently removed. Li and Liu (2004, 2009) indicated that the removal of different compounds differs considerably in the individual zones. A combined-bioreactor can be optimally designed according to the characteristics of compounds in the gaseous effluents. Moreover, gases can be simultaneously humidified when passing through the suspended zone. Humidification of this zone is sound and optimum moisture in the immobilized zone can be maintained for a relatively long period, without any additional pre-humidification system. Investment and operational costs should be reduced when these conditions are applied to a scaled-up commercial process (Li and Liu 2006).

**Thermophilic bioreactors**

In general, biofiltration techniques are limited to operation under mild temperature conditions (25–35 °C) in which mesophilic and oxidizing microbial populations form biofilms. However, emissions generated from boiler combustion, petroleum refinery, smelting, and composting facilities contain sulfur compounds at temperatures higher than 50 °C (Morales et al. 2012). Precooling the gas stream
Table 2 Application area, advantages and disadvantages of the traditional and some new type biotechnologies for the removal of waste gases (Van Groenestijn and Hesselink 1993; Devinny et al. 1999; Kennes and Veiga 2004)

| Bioreactor type                  | Advantages                                                                 | Disadvantages                                                                 | Application area                                                                 |
|----------------------------------|-----------------------------------------------------------------------------|-------------------------------------------------------------------------------|--------------------------------------------------------------------------------|
| Biofilter                        | High gas/liquid surface area, Easy operation and start-up, Low operation cost | Poor control of reaction conditions, Slow adaptation to fluctuating concentrations in gas, Large area required | Concentration target compounds $<1.0 \text{ g m}^{-3}$, Henry coefficient $<10$ |
| Bioscrubber                      | Better control of reaction conditions (pH, nutrients), Possibilities to avoid accumulation of products, Low pressure drop | Low surface area for mass transfer, Wash out of slow growing microorganisms, Complicated start-up procedure, High investment, maintenance and operational costs | Concentration target compounds $<5 \text{ g m}^{-3}$, Henry coefficient $<0.01$ |
| Biotrickling filter              | Comparable to bioscrubbing, Better retention of slow growing microorganisms, Single reactor | Low surface area for mass transfer, Disposal of excess sludge, Complicated start-up procedure, Higher operational costs | Concentration target compounds $<0.5 \text{ g m}^{-3}$, Henry coefficient $<1$ |
| Fungal bioreactor                | High substrate/microbiota contact surface area, High elimination capacity (especially for hydrophobic compounds), Resistant to low pH value, water content and limited nutrient concentrations | Fast clogging, Potential pathogenicity of some fungal strains, Metabolize limited substrates | Waste stream with hydrophobic compounds |
| Thermophilic bioreactor          | No additional cooling equipment, Low operation cost, Higher degradation kinetics | Poor control of reaction conditions (moisture), Long adaptation and biomass build-up period | Waste stream with high temperature |
| Bioreactor integrated with physicochemical techniques | Compact and mobile, Possibilities to be installed in series with the bioreactor, High removal efficiency, Less sensitivity to inlet load surge, Possibilities to minimize bioreactor volume | Poor control of reaction conditions, Complicated start-up procedure, High investment, maintenance and operational costs | A wide range of feed stream flow rates and compositions |

Table 3 The physical and chemical properties of sulfur compounds

| Name of the compounds          | Molecular formula | OTCs (ppm) | Henry’s law constants | Solubility in water (g/L) |
|--------------------------------|-------------------|------------|-----------------------|--------------------------|
| Hydrogen sulfide               | H$_2$S            | 0.00041    | $5.5 \times 10^2$     | 4.12                     |
| Sulfur dioxide                 | SO$_2$            | 0.3        | 40.67                 | 94                       |
| Carbon disulfide               | CS$_2$            | 0.21       | $5.82 \times 10^3$    | 2                        |
| Methyl sulfide                 | (CH$_3$)$_2$S     | 0.0001     | $1.12 \times 10^2$    | Insoluble                |
| Methyl mercaptan               | CH$_3$SH          | 0.0001     | $1.08 \times 10^2$    | Insoluble                |
| Ethyl mercaptan                | CH$_3$CH$_2$SH    | 0.0000087  | $4.5 \times 10^3$     | 6.76                     |
| Diethyl sulfide                | (CH$_3$CH$_2$)$_2$S | $3.3 \times 10^{-5}$ | –                      | Insoluble                |
| Dimethyl disulfide             | (CH$_3$)$_2$S$_2$ | 0.0003     | –                     | Insoluble                |
to appropriate temperatures is an effective preprocessing method (Shanchayan et al. 2006), but increases the investment and operational costs of the treatment. Alternatively, thermophilic biofiltration, which utilizes the metabolic activity of thermophilic organisms, does not require precooling treatment. Thermophilic biofilters have the advantages of low cost and high microbial metabolism rate (Devlin et al. 1999). Elimination capacity can be increased using thermophilic bacteria instead of mesophilic bacteria. Furthermore, thermophilic bioreactors exhibit low biomass accumulation, thus alleviating the risk of clogging (Kong et al. 2001; Mohammad et al. 2007). Sulfur dioxide and other sulfur compounds have been successfully purified through thermophilic biofiltration (Morales et al. 2012; Zhang et al. 2015).

**Bioreactors integrated with physicochemical techniques**

Biological treatment processes used to remove and degrade volatile organic compounds (VOCs) from contaminated gases emitted by industrial operations or waste treatment are always subjected to transient loading conditions because of the inherently unsteady-state of contaminant generating processes. Biofiltration for VOC control in waste gas streams is best operated at steady contaminant loadings. However, a biofiltration system needs a large volume bioreactor, which is slow to adapt to fluctuating concentrations in waste gas. To provide long-term stable operation of a bioreactor under adverse contaminant feeding conditions, bioreactors integrated with various physicochemical techniques have been developed for waste gas stream treatment.

**Bioreactors integrated with an adsorption unit** Single- and double-activated carbon adsorption columns have been used as buffer units placed before a biofilter for VOC emission control with unstable pollutant loads. A biofilter followed by an adsorption column packed with granule active carbon (GAC) was shown to be an effective alternative to conventional biotechniques for the attenuation of fluctuating loads of waste gas streams. Using such a system, Li et al. (2008a, b) increased the elimination capacity and achieved a total removal efficiency of 98.6 % in experiments with fluctuating o-xylene concentration over 1000 mg m$^{-3}$ and empty bed contact time of 72 s. The combination of adsorption and microbial processes not only led to high and stable removal efficiency, but also improved the capacity of resisting shock loads. Ottengraf et al. (1986) applied activated carbon filters as a buffer in combination with biofilters for plants operating discontinuously. In that system, exhaust gases from a night soil treatment plant were treated by a pilot-scale integrated deodorization system consisting of an activated carbon fabric reactor (placed before the biofilter) and a peat biofilter. Stable removal efficiency of dimethyl sulfide and dimethyl disulfide in this integrated system continued for longer than that in a peat biofilter alone. Furthermore, integrated systems more easily achieved microbial activity than that of single-stage peat biofilters due to water supply requirements and peat stabilization (Park et al. 1993).

By testing various adsorbents, Weber and Hartmans (1992) concluded that selection of the most suitable type of adsorbent for a specific application depended on the nature of the contaminant and on the magnitude of the concentration fluctuations in the waste gas. Adsorption on GAC can be a physical or chemical process. When adsorbed on the carbon surface, molecules of organic pollutants are strongly captured by adsorption forces. The texture and surface chemistry of activated carbons, which are responsible for their adsorption capacity, can be modified according to the characteristics of the compounds to be treated (Li et al. 2011a).

**Membrane bioreactors** Another biotechnique for the treatment of volatile sulfur compounds is the use of membranes integrated with bioreactors. Two different approaches have been reported: submerged membrane bioreactors (Fig. 5) and circulated membrane bioreactors (Fig. 6). Both membranes separate the gas phase and the liquid phase, which contains microorganisms, but permit the transfer of target compounds between the two phases.

**Submerged membrane bioreactors** In submerged membrane bioreactors, the surface of the membrane forms the contact area, which is larger than that in biofilters or biotrickling filters (Reij et al. 1998). The microorganisms are attached to the liquid side of the membrane or are in suspension. The concentration difference between the gas phase and the biofilm phase provides the driving force for diffusion across the membrane. The driving force relies mainly on the air–water partition coefficient and the concentration in the liquid phase of the diffusing component. This bioreactor is usually used for the treatment of hydrophobic volatile sulfur compounds, e.g. dimethyl sulfide (Luvsanjamba et al. 2008; Kumar et al. 2010), because the liquid phase is situated at the opposite side of the biofilm and does not form a barrier for mass transfer of the poor water-soluble compounds. Kumar et al. (2010) conducted dimethyl sulfide removal from waste air using a membrane biofilm reactor under continuous feeding conditions, and observed a maximum elimination capacity of 258.3 g m$^{-3}$ h$^{-1}$ under optimum conditions, which is higher than that reported for biofilters or biotrickling filters previously. Volckaert et al. (2014) suggested that mixtures
of hydrophilic and hydrophobic compounds should be treated by a combination of membrane bioreactor and biofilter due to their ability to be operated in a complementary manner. Usually, polydimethylsiloxane membranes have been selected for dimethyl sulfide removal (De Bo et al. 2003; Kumar et al. 2010).

**Circulated membrane bioreactors** In circulated membrane bioreactors, target compounds can be separated from the gas stream when they pass through the membrane modules. Unlike the submerged membrane bioreactors, a pressure difference between the two sides of the membrane is necessary. A large volume of air flows on one side of the membrane at the feed pressure during the removal and recovery of target compounds from air streams at a certain pressure by membrane separation. When a vacuum is applied to the other side to create a partial pressure driving force, the target compounds selectively permeate through the membrane in preference to air. Separation occurs because different compounds transport across the membrane at different rates. Using single or several membrane modules as separation units before a biofilter was first achieved in the control for mixtures of hydrogen sulfide and methane emission (Chinese Patent ZL201210238668.0). In gas membrane modules, permeation of hydrogen sulfide through membranes is much faster than that of methane in the off-gas. Therefore, the permeated stream becomes enriched in hydrogen sulfide, while the retentate stream turns to hydrogen sulfide depleted air. The enriched-hydrogen sulfide in the permeated stream is charged to the biofilter and the concentration of sulfur compounds in the retentate stream is then low enough for venting into the atmosphere. An integrated bioreactor system with a gas separation membrane module installed after a biofilter was proposed for styrene treatment to provide long-term stable operation of a biofilter under adverse contaminant feeding conditions (Li et al. 2012). Total removal efficiency of over 96 % was achieved when the biofiltration column faced fluctuating loads and the maximum elimination capacity of the integrated bioreactor system was 93.8 g m$^{-3}$ h$^{-1}$, which was higher than that obtained with the biofiltration column alone. The combination of gas separation and microbial processes resulted in increased removal efficiency, less sensitivity to inlet load surge and minimized bioreactor volume (Li et al. 2012).
To date, all investigations carried out on membrane reactors have been laboratory scale experiments. The main reasons restricting their application are service life, cost of membrane materials, and membrane fouling during the application process.

**Bioreactors integrated with non-thermal plasma** Non-thermal plasma (NTP) is a promising technology for waste gas treatment (Durme et al. 2008). Previous research using a non-thermal dielectric barrier discharge plasma reactor filled with ceramic Raschig rings exhibited good hydrogen sulfide removal performance (Liang et al. 2011). Another bench scale system integrated with a NTP and biotrickling filtration unit was investigated for the treatment of gases containing dimethyl sulfide (Wei et al. 2013). The integrated system achieved nearly 96 % dimethyl sulfide removal efficiency, and NTP oxidized dimethyl sulfide to methanol and carbonyl sulfide. Subsequently, the intermediate organic products were further oxidized to sulfate, carbon dioxide, and water vapor by biological degradation. The addition of ozone from NTP increased the complexity of the microbial community in the biotrickling filtration unit and its activity in dimethyl sulfide removal, demonstrating that an NTP-integrated bioreactor is achievable and could be applied to sulfur compound removal from gaseous effluent.

The type of bioreactor used for abatement has a direct consequence on the efficiency of the treatment process. An understanding of the bioreactors used for the treatment of sulfur compounds, their design and configuration, as well as necessary parameters for their operation, will not only help increase the efficiency of the treatment process but also provide insight into the development of newer, improved, and more robust treatment techniques.

**Microbial characteristics**

A wide variety of microorganisms capable of degrading sulfur containing compounds have been isolated and identified from the bioreactors. In general, the efficiencies of bioreactors can be improved by inoculating specific microorganism with enhanced metabolic capabilities. Performance data of bioreactors containing bacteria, fungi or mixed populations as dominant microorganisms are demonstrated in Table 4. These data indicate that good results have been achieved both with bacterial and fungal biocatalysts.

**Sulfur-oxidizing bacteria**

*Thiobacillus sp.*

Within bioreactors, microorganisms supported on media or maintained in suspension may produce enzymes to biodegrade odor into carbon dioxide, water and less hazardous products. Bacteria and fungi are two dominant microorganism groups used in bioreactors. Bacteria are most likely present in conventional biofilters under favorable conditions due to their rapid substrate uptake and growth. Various sulfur-oxidizing bacteria, including autotrophic and heterotrophic bacteria, have been used for sulfur-containing malodorous gas control for many years. In 1970, for example, *Thiobacillus* sp. was isolated from bioreactors for treating hydrogen sulfide ( Moriarty and Nicholas 1970), and was shown to grow on methanethiol, dimethyl sulfide, dimethyl disulfide and sulfur dioxide, which can be utilized as electron acceptors (Cho et al. 1991).

Under aerobic conditions, sulfur-oxidizing bacteria oxidize sulfur-containing compounds to produce sulfuric acid [Eqs. (1–4)]. Acid formation causes the pH of the bioreactor to fall, which results in substantial reductions in treatment success (Furusawa et al. 1984; Yang and Allen 1994). The addition of alkalis in packing material or irrigation water is an effective way to solve this problem, and alkaline biotrickling filters have been used for the effective treatment of hydrogen sulfide odor (González-Sánchez et al. 2008). The gas contact time ranged from 1 to 6 s, and H2S inlet concentrations were from 3.8 to 27.3 mg m⁻³. H2S removal exceeded 98 % when the loading was less than 30 g m⁻³ h⁻¹.

An alternative method is to operate the bioreactor at low pH. *Thiobacillus* species, e.g. *Thiobacillus thioparus* and *Acidithiobacillus thiooxidans*, often dominate in bioreactors and are capable of rapidly oxidizing hydrogen sulfide (Liu et al. 2008), methanethiol, dimethyl sulfide, and dimethyl disulfide (Ramírez et al. 2011) in environments with pH values below 3. In addition, *Thiobacillus thioparus* is accompanied by acidiphilic heterotrophs, which consume by-products (usually low molecular organics) of *T. thioparus*. Therefore, a sulfide biofilter can treat more than just sulfide. Studies have shown that low pH bioreactors are effective for simultaneous treatment of hydrogen sulfide and toluene (Cox and Deshusses 2002; Gao et al. 2011).

\[
\begin{align*}
H_2S & \xrightleftharpoons{O_2} S^0 + 2H^+ O_2 SO_4^{2-} + 2H^+ \quad (1) \\
SO_2 + H_2O & \rightarrow HSO_3^- + H^+ O_2 SO_4^{2-} + H^+ \quad (2) \\
CH_3SH_{\text{microorganism}} & \rightarrow CH_4 + H_2S O_2 CO_2 + H_2O + SO_4^{2-} \\
& + 2H^+ \quad (3) \\
CH_3OSCH_3_{\text{microorganism}} & \rightarrow CH_3SCH_3 O_2 CH_3SH O_2 H_2S O_2 \\
& \rightarrow H_2SO_4 \quad (4)
\end{align*}
\]

*Pseudomonas sp.*

*Pseudomonas* sp. secrete oxidative enzymes that can degrade sulfur compounds. Acting as sulfide oxidizers,
| Microbiota type | Target pollutant | Bioreactor | Removal | References |
|----------------|-----------------|------------|---------|------------|
| *Thiobacillus concretivoros* | H₂S | Biofilter | C_{in}: 176–239 mg m⁻³  
RE: 95–98 % | Moriarty and Nicholas (1970) |
| *Thiobacillus thioparus, Thiobacillus denitrificans* | H₂S | Biofilter | C_{in}: 250 mg m⁻³  
RE: 100 % (180 days) | Degorce-Dumas et al. (1997) |
| *Thiobacillus novellus* | H₂S | Bioreactor (30 °C) | 303 mg m⁻¹ H₂S was removed in 2.0 h, 107 mg m⁻³ MM in 2.6 h, 83 mg m⁻³ DMS in 7.0 h, and 126 mg m⁻³ DMDS in 6.0 h in a fermenter containing 8.3 × 10⁹ cells | Cha et al. (1999) |
| *Thiobacillus thiooxidans* | H₂S | 25 °C | Oxidized the H₂S at a maximum oxidation rate of 0.84 mol g⁻¹ cell⁻¹ min⁻¹ | Hirano et al. (1996) |
| *Thiobacillus sp.* | H₂S | | | Cox and Deshusses (2002) |
| *Acidithiobacillus thiooxidans* | H₂S | Biotrickling filter | Load: 4.2–102 g m⁻³ h⁻¹  
EC: 57.5 g m⁻³ h⁻¹  
RE: 92 %  
EBRT: 59 s | Ramírez et al. (2011) |
| *Pseudomonas sp.* | H₂S, MM, DMDS, DMS from swine wastewater treatment system | Field scale biofilter | Load: 0.03–0.48 g m⁻³ h⁻¹ (H₂S); 0.07–2.49 g m⁻³ h⁻¹ (MM); 0.26–5.03 g m⁻³ h⁻¹ (DMDS)  
RE: 96–100 %  
EBCT: 13–30 s | Ho et al. (2008) |
| *Pseudomonas putida* | H₂S | Biofilter | C_{in}: 15–227 mg m⁻³  
RE: >96 %  
Flow rate: 3.6 × 10⁻² m³ h⁻¹ | Chung et al. (1996) |
| *Pseudomonas sp.* | H₂S, MM | | Remove 1.5 mg m⁻³ of H₂S gas or 1.0 mg m⁻³ MM gas in 2 h | Honma and Akino (1998) |
| *Lysinibacillus sp.* | H₂S from landfill site | Full-scale biofilter | C_{in}: 10.5 g m⁻³ h⁻¹  
EC: 9.1 g m⁻³ h⁻¹  
EBCT: 60 s | Li et al. (2012) |
| *Aquanicrobium defluvii,* *Stenotrophomonas sp.* | H₂S from a landfill site | Full-scale biofilter | C_{in}: 38.3 mg m⁻³  
EC: 2.1 g m⁻³ h⁻¹  
EBCT: 60 s | Li et al. (2013) |
| *Ochroacterium anthropi* | DMS | Lab-scale biofilter | Load: 9.4 g m⁻³ h⁻¹  
RE: 100 %  
EBCT: 30 s | Shu and Chen (2009) |
| *Microbacterium sp.* | DMS | | | |
| *Pseudomonas putida* | H₂S, DMS | Biotrickling filter | C_{in}: 407 mg m⁻³  
RE: 90 %  
EBCT: 120 s | Sercu et al. (2005) |
| *Hyphomicrobiurn sp.* | H₂S, DMS | | | |
| Engineered *E. coli BL21* | MP and parathion | Biofilter | C_{in}: 0.04–0.25 mg m⁻³ (MP); 0.04–0.33 mg m⁻³ (parathion)  
RE: 98.6 % (MP); 95.2 % (parathion)  
EBCT: 34 s | Li et al. (2011b) |
they are frequently applied in desulphurization bioreactors (Honma and Akino 1998). *Pseudomonas* sp. remained the predominant community (56–70 %) after long-term evaluation of a biofilter in the elimination of volatile-sulfur compounds emitted from a swine wastewater treatment system. Hydrogen sulfide, methanethiol, dimethyl disulfide, and dimethyl sulfide were effectively reduced to 96–100 % at gas residence times of 13–30 s, and elemental sulfur and sulfate were their primary oxidation metabolites (Ho et al. 2008). *Pseudomonas fluorescens* was shown to grow on dimethyl disulfide and produce H$_2$SO$_4$, whereas dimethyl sulfide was utilized as an electron acceptor by *Pseudomonas fluorescens* (Ito et al. 2007).

**Table 4 continued**

| Microbiota type                  | Target pollutant | Bioreactor | Removal | References |
|----------------------------------|------------------|------------|---------|------------|
| Engineered *E. coli* SD2         | parathion        |            | 0.146 mg m$^{-3}$ of parathion was hydrolyzed effectively by the co-culture | Gilbert et al. (2003) |
| *Pseudomonas putida*             |                  |            |         |            |
| Engineered *E. coli* BL21        | MP               | Biofilter  | C$_{in}$: 5.11 mg m$^{-3}$ | Li et al. (2008a, b) |
|                                  |                  |            | EC: 950 mg m$^{-3}$ h$^{-1}$ |            |
| *Ochrobactrum* sp.               |                  |            | EBCT: 35 s |            |
| *Paenibacillus* sp. and *Pseudomonas* spp. | SO$_2$ | Biofilter (60 °C) | C$_{in}$: 100–200 mg m$^{-3}$ | Zhang et al. (2015) |
|                                  |                  |            | EC: 50.67 g m$^{-3}$ h$^{-1}$ |            |
|                                  |                  |            | EBCT: 18 s |            |
| *Bacillus* sp.                   | H$_2$S           | Biofilter  | C$_{in}$: 1442 mg m$^{-3}$ | Ryu et al. (2009) |
|                                  |                  | (60 °C)    | RE: >95 % |            |
|                                  |                  |            | EBCT: 1.2 min |            |
| *Sulfolobus metallicus*          | H$_2$S           | Biofilter  | C$_{in}$: 0–1.0 × 10$^2$ mg m$^{-3}$ | Morales et al. (2012) |
|                                  |                  | (50 °C)    | EC: 37.1 ± 1.7 g m$^{-3}$ h$^{-1}$ |            |
|                                  |                  |            | EBCT: 120 s |            |
| *Thiomonas* sp.                  | H$_2$S           | Bioreactor | C$_{in}$: 151 mg m$^{-3}$ | Asano et al. (2012) |
|                                  |                  | (45 °C)    | RE: 99 % |            |
|                                  |                  |            | EBCT: 300 s |            |
| *Unidentified basidiomycete*      | H$_2$S, MM,      | Removal rate: 3477 mg m$^{-3}$ g$^{-1}$ dry cell mass h$^{-1}$ (H$_2$S); 240 mg m$^{-3}$ g$^{-1}$ dry cell mass h$^{-1}$ (MM); 1212 mg m$^{-3}$ g$^{-1}$ dry cell mass h$^{-1}$ (DMS); 235 mg m$^{-3}$ g$^{-1}$ dry cell mass h$^{-1}$ (DMDS) | Phae and Shoda (1991) |
|                                  | DMS, DMSDS       |            | Load: 45 g m$^{-3}$ h$^{-1}$ |            |
| *Cephalosporium*                 | H$_2$S, MM,      | Removal rate: 0.27 mg m$^{-3}$ g$^{-1}$ dry cell mass h$^{-1}$ (H$_2$S); 15.6 mg m$^{-3}$ g$^{-1}$ dry cell mass h$^{-1}$ (MM); 29.2 mg m$^{-3}$ g$^{-1}$ dry cell mass h$^{-1}$ (DMS) | Ishikawa et al. (1980) |
|                                  | DMS, DMDS        |            | EBCT: 58 s |            |
| *Penicillium frequens*, *Paecilomyces variotii*, *Aspergillus wentii*, *Cephalosporium acremonium* | EtSH | Biofilter | C$_{in}$: 0.18 mg m$^{-3}$ (H$_2$S); 0.85 mg m$^{-3}$ (EtDH); 0.22 mg m$^{-3}$ (DMS) | Zhu and Liu (2004) |
| *Aspergillus niger*, *Trichoderma koningi*, *Penicillium frequentans* | H$_2$S, EtSH, DMS | Removal rate: 1.49 × 10$^{-2}$ g m$^{-3}$ h$^{-1}$ (H$_2$S); 7.09 × 10$^{-2}$ g m$^{-3}$ h$^{-1}$ (EtSH); 1.85 × 10$^{-2}$ g m$^{-3}$ h$^{-1}$ (DMS) | Li and Liu (2009) |
|                                  |                  |            | EBCT: 60 s |            |

*C$_{in}$ Inlet concentration, RE removal efficiency, EC elimination capacity, EBCT empty bed contact time, H$_2$S hydrogen sulfide, MM methyl mercaptan, DMS dimethyl sulfide, DMDS dimethyl disulfide, EtSH ethanethiol, SO$_2$ sulfur dioxide, MP methyl parathion*

**Considered microbial consortium**

Considerable progress has been made in the field of biological degradation of pollutants by constructed microbial consortia, in particular volatile organic sulfur compounds. A laboratory-scale biofilter inoculated with *Escherichia coli* BL21 over-expressing methyl parathion hydrolase (Yang et al. 2006) was operated for treating an air stream.
containing parathion and methyl parathion (MP) so as to test the feasibility of genetically engineered microorganism application. Due to the highly effective biodegradation by genetically engineered bacteria, complete hydrolysis of MP and parathion occurred from the third day of operation and no MP or parathion was detected in the outlet gas. Compared with conventional biofilters, the biofilter inoculated with the engineered *E. coli* BL21 was far more effective, especially in the initial stages (Li et al. 2011b). In another study, a consortium composed of two engineered strains (*Escherichia coli* SD2 and *Pseudomonas putida* KT2440 pSB337) was assembled and resulted in the complete mineralization of parathion. 146 mg L⁻¹ of parathion could be hydrolyzed by *E. coli* SD2 and the hydrolysis product p-nitrophenol (PNP) was mineralized by *P. putida* KT2440 pSB337 (Gilbert et al. 2003). A consortium comprised of an engineered *E. coli* BL21 enabling the over expression of methyl parathion hydrolase and a natural PNP degrader (*Ochrobactrum* sp. strain LL-1) was assembled for complete mineralization of MP (Li et al. 2008a, b). The co-culture effectively hydrolyzed MP and prevented the accumulation of PNP in the suspended culture. The bioreactor containing the dual-species consortium maintained an average MP removal efficiency of more than 98 % over a 75 day period. The maximum MP elimination capacity was 950 mg m⁻³ h⁻¹, corresponding to empty bed contact time of 35 s and inlet gas concentration of 5.11 mg m⁻³ (Li et al. 2008a, b).

**Thermophilic desulfurization bacteria**

Most biochemical transformations occur more rapidly at high temperature. The development of immobilized thermophilic desulfurization bacteria in bioreactors for thermophilic operation creates more rapid and economical treatment processes (Asano et al. 2012). To treat hot gases containing hydrogen sulfide, Ryu et al. (2009) used a biofilter filled with porous particles inoculated with *Bacillus* sp. TSO3, a culture collected from high-temperature compost. The biofilter achieved excellent performance, removing 56.6 g m⁻³ h⁻¹ of hydrogen sulfide.

Bacteria are adapted to living in water or humid environments. Cell surfaces are often covered with a layer of liquid film. The pollutant to be treated is initially absorbed by the aqueous film that surrounds the biofilm, and then biodegradation takes place within the biofilm. Therefore, bacteria have high removal capacity in the treatment of hydrophilic compounds. They have difficulty, however, in removing hydrophobic compounds because of their low solubility in water. Moreover, maintaining suitable pH and moisture conditions in the bioore are also challenging problems.

**Fungi**

Biodegradation of sulfur-compounds by fungi is also possible, although it is apparently much less common among eukaryotes than among bacteria. The aerial mycelium of fungi may grow in air space. Their hydrophobic surface remains dry and offers a very large surface area in direct contact with off-gases flowing through the bioreactor. As a result, fungi tend to display greater metabolic diversity, attacking hydrophobic compounds beyond the capabilities of bacteria. They are also more tolerant towards drying and able to remain active when water is less available. Zhu and Liu (2004) reported a successful bench-scale biofilter operated at low water content for the treatment of ethanethiol by fungi. Although the pH was within 3–5 and the relative humidity of the off-gas was below 85 %, fungi growth could be observed and the bioreactor presented high and constant activity. A biofilter populated by a mixed culture of *Coriolius versicolor* and *Tyromyces palastris* was investigated for the potential ability of Basidiomycetes to degrade bis (2-chloroethyl) sulfide. Bis (2-chloroethyl) sulfide was degraded very rapidly by both fungi species. Fungi present in gas-phase biofilters allowed the bioreactors to reach high elimination capacities in the removal of hydrogen sulfide, methanethiol, dimethyl sulfide and dimethyl disulfide, and were resistant to low pH values and reduced moisture content (Phae and Shoda 1991). The optimal pH for degradation activity was between 5.6 and 6.9. The hydrogen sulfide was oxidized to sulfate via sulfite and dimethyl sulfide was stoichiometrically converted to dimethyl sulfoxide. Usually, the packing material used in gas-phase bioreactors is a porous substance with a large specific surface area and a coarse fraction. The hyphae grown by fungi extend into the crevices and pores of the support materials and other parts collect contaminants for degradation at the surface. The aerial mycelia not only improve absorption of hydrophobic compounds in gases, but also act as supporters for bacteria attached growth. *Bacillus* sp. was shown to accumulate on the mycelium of fungi, thereby expanding the contact area for the uptake of substrates and oxygen in the gas phase (Li and Liu 2009). Most fungi for degrading sulfur-compounds have been isolated from mesophilic environment. Fungal biofiltration under thermophilic condition is a seldom explored research area (Kennes and Veiga 2004).

Various factors influence the growth kinetics of participating microorganisms, including substrate concentration, moisture content, temperature, oxygen transfer, and pH value (Ottengraf et al. 1986). The species present, their population diversities, metabolic transformations, and interactions with the environment and each other are fundamental to biofilter operation. Molecular biological methods, such as polymerase chain reaction-denaturing
Conclusions and future trends

Sulfur-containing compounds in waste streams are sulfurous pollutants with environmental health implications, and need to be treated prior to discharge into the environment. The first research devoted to the biofiltration of sulfurous pollutants was published about 100 years ago. The implementation of biological techniques for waste stream abatement has continued to increase and has become an alternative to traditional physicochemical treatments. The type of bioreactor used for abatement has a direct consequence on the efficiency of the treatment process. Future research should define how to maintain high performance for long periods under stable conditions and should focus on the development of eco-friendly and techno-economically viable treatment processes. In addition, treatment improvement must target biofilter microbial communities and operational factors e.g. loading, temperature, moisture content, pH and oxygen transfer.

The potential of various bacteria to metabolize sulfur compounds effectively has been reported previously. Among them, *Thiobacillus* sp. and *Pseudomonas* sp. are most widely used in laboratory and pilot scale systems. Future research should focus on the isolation and identification of new microorganisms with a broader substrate spectrum as well as the construction of microbial consortia for high selectivity and efficiency removal.

The potential pathogenicity of bioaerosols with bacterial cells and fungal spores emitted from bioreactors is another key issue that needs to be investigated in biological waste air treatment studies, but has been somewhat overlooked during bioreactor design.

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