



Mathematical modeling of biological sulfide removal in a fed batch bioreactor

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ABSTRACT

In this study, biological sulfide removal is investigated in a fed batch bioreactor. In this process, sulfide is converted into elemental sulfur particles as an intermediate in the oxidation of hydrogen sulfide to sulfate. The main product is sulfur at low dissolved oxygen or at high sulfide concentrations and also more sulfates are produced at high dissolved oxygen. According to the carried out reactions, a mathematical model is developed. The model parameters are estimated and the model is validated by comparing with some experimental data. The results show that, the proposed model is in a good agreement with experimental data. According to the experimental result and mathematical model, sulfate and sulfur selectivity are sensitive to the concentration of dissolved oxygen. For sulfide concentration 0.2 (mM) in the bioreactor and dissolved oxygen of 0.5 ppm, only 10% of sulfide load is converted to sulfate, while it is 60% at the same sulfide concentration and dissolved oxygen of 4.5 ppm. At high sulfide load to the bioreactor, the concentration of uneliminated sulfide increases; it leads to more sulfur particle selectivity and consequently, less sulfate selectivity.

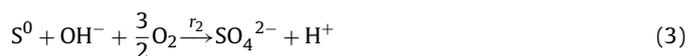
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1. Introduction

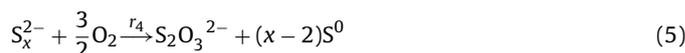
Hydrogen sulfide is emitted by many industries such as petroleum refining, natural gas and petrochemical plants [1]. It is an extremely toxic gas and has potential for injuring developing central nervous systems at low-dose exposures [2]. The threshold limit value for air 0.5–10 ppbv [3], natural gas 4 ppmv [4] and for fresh or salty water fish is 0.5 ppm [5]. Sulfide in wastewater is also corrosive and has a very unpleasant odor. H₂S is frequently the main component of most observable odorous emissions [6]. Several microorganisms are capable of oxidizing H₂S at ambient temperatures and pressures. A review on bacteria of the sulphur cycle was discussed by Tang et al. [7]. Also, a review on removal of H₂S from gas streams using biological processes was discussed by Sayed et al. [8]. There is a wide variety of sulfur compounds oxidizing bacteria (SOB), each of the bacteria has its own specific properties, such as the size, shape, the source of energy, carbon, or hydrogen, the sulfide-oxidizing pathway, and the location of storing intermediate sulfur [9]. The product of the sulfide oxidation at low dissolved oxygen is sulfur and at high dissolved oxygen is a sulfate ion (Eqs. (1) and (2)) [10]:



SOB produce elemental sulfur as an intermediate in the oxidation of hydrogen sulfide to sulfate [11–13]. On the other word, SOB oxidize sulfide to sulfur particles, and then the produced sulfur particles are biologically oxidized to sulfate. According to this fact, sulfur and sulfate production are consecutive reactions. Thus, Eq. (2) can be reformed as Eq. (3).



In this system, in addition to the biological oxidation of sulfide to sulfur and sulfate (Eqs. (1) and (3)), undesirable abiotic reactions occur in the bioreactor as shown in Eqs. (4)–(6):



Dissolved sulfide can react with S⁰ to produce S_x²⁻ ions, according to Eq. (4), and S_x²⁻ ions are abiotically oxidized to S⁰ and S₂O₃²⁻ (Eq. (5)) [14]. The rate of Eq. (6) is slow at the ambient temperatures, thus, it can be ignored relative to other reactions [15]. Distinguishing between polysulfide ions (S_x²⁻) of different chain length (x) is difficult, however at neutral to alkaline pH values, concentrations of S₆²⁻, S₅²⁻ and S₄²⁻ are dominant in the solution [16]. Teder

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Nomenclature

A	constant of extended Debye–Hückel equation (0.509) ($\text{mol}^{-1/2} \text{L}^{1/2}$)
a	radius of the ion (m)
C_B	optical density of bacteria
F_{in}	input flow rate (ml h^{-1})
I	the ionic strength (mmol L^{-1})
k_1	reaction rate constant ($\text{mmol L}^{-1} \text{h}^{-1}$)
k_2	reaction rate constant (mmol L^{-1})
k_3	reaction rate constant (mmol L^{-1})
k_4	reaction rate constant ($\text{mmol L}^{-1} \text{h}^{-1}$)
k_5	reaction rate constant (mmol L^{-1})
k_6	reaction rate constant (mmol L^{-1})
k_7	reaction rate constant (mmol L^{-1})
k_8	reaction rate constant ($\text{mmol}^{-0.59} \text{L}^{-0.591} \text{h}^{-1}$)
pK_x	the acid dissociation constant
r	reaction rate ($\text{mmol L}^{-1} \text{h}^{-1}$)
S	sample volume (ml)
T	toluene volume (ml)
t	time (h)
V	volume of the broth in the bioreactor (L)
X	chain length of polysulfide ion
Z	the ionic charge of ion
β	constant of extended Debye–Hückel equation (0.328×10^8) ($\text{mol}^{-1/2} \text{L}^{1/2} \text{m}^{-1}$)
γ	activity coefficient

[17] has shown that the chain length of polysulfide (S_x^{2-}) increases with increase of temperature ($x=5.0$ at 25°C and $x=6.5$ at 80°C). At moderate alkaline conditions, the average chain length of polysulfide (x) varies from 4.6 to 5.5 [14,17–20]. In the present study, the average chain length x was chosen equal to 5.

2. Materials and methods

2.1. Strain and media

In this study, *Thiobacillus thioparus* (DSMZ 5368) was used as sulfur oxidizing bacteria for producing elemental sulfur as the product in the oxidation of hydrogen sulfide. The medium for growth is shown in Table 1.

After addition of all compounds, the pH of the medium was 7.1 ± 0.1 at 30°C . Then the strain (100 cm^3 of the medium culture containing bacteria with an OD_{600} of about 0.5) was added to a flask containing 2000 cm^3 of the medium. After that, the medium was kept in a shaker incubator with 200 rpm and temperature 30°C for five days to get enough concentration of bacteria.

2.2. Experimental set-up and procedure

A reactor with total volume of 3.8 L operated under fed batch conditions, as shown in Fig. 1 was constructed. During each experiment, the temperature, dissolved oxygen, sulfide load and pH were controlled. The reactor was placed in a water bath for temperature control within 0.5°C of the set point (30°C). The pH was controlled using 1N HCl and 1N NaOH solutions between 7.8 and 8.2. The dissolved oxygen (DO) was changed between 0.5 and 6 ppm and controlled using nitrogen and oxygen injection. The original inoculum of this reactor was the free thiosulfate and bromocresol purple main medium (Table 1). The bioreactor was charged with 2000 cm^3 of the medium and inoculated with centrifuged biomass. After temperature stabilization, feeding of sulfide to the bioreactor was started. Sulfide solution was prepared by solving sodium

sulfide ($\text{Na}_2\text{S} \cdot 9\text{H}_2\text{O}$) in deionized water. The concentration of prepared sulfide solution was 500 mM. The concentration of this solution was standardized based on methylene blue method described by Trüper using a spectrophotometer (Zeiss) at 665 nm [22]. The solution was added to the bioreactor by an infusion pump (JMS OT-701) after calibration of the pump. During the experiments, the flow rate of the sulfide solution was changed between 1.5 and 23 mL h^{-1} , thus the HS^- load was changed between 0.3 and $5.7 \text{ mmol L}^{-1} \text{h}^{-1}$. The flow rate of recirculating gas was maintained at 15 L min^{-1} and was spread by a diffuser; this caused a good mixing of the broth. A schematic diagram of the experimental setup is shown in Fig. 1.

The reactor was equipped with sensors for temperature, pH (Metrohm LL-Unitrode Pt 1000), dissolved oxygen (AZ 8403) and oxidation reduction potential (ORP Ag/AgCl electrode AZ 8501) electrode. ORP electrode is a measure of the tendency of a chemical species to acquire electrons and thereby be reduced. Reduction potential is measured in millivolts. The total concentrations of sulfide (HS^- and polysulfide) were determined based on methylene blue method described by Truper using a spectrophotometer at 665 nm [22]. The concentration of polysulfide was determined based on Teder method [17]. The concentration of sulfide (HS^-) is the difference between total sulfide concentrations and polysulfide concentration. The concentration of sulfate was determined via the turbidimetry method at 420 nm [23]. While the thiosulfate concentration was determined via methylene blue method at 760 nm [24].

A method is proposed for simultaneous analysis of the bacteria and sulfur particles (not published yet) which is discussed in supplementary data file.

An UV/VIS spectrophotometer (Optima, SP 3000 plus) was used for measurement of all components.

3. Mathematical modeling

Model equations of fed batch reactors allow for continuous feeding, absence of outflow (except of sampling) from the bioreactor and increase in volume (accumulation of total mass) in the bioreactor, as schematically shown in Fig. 1. Simulation of fed batch bioreactors can be used to demonstrate the important characteristics of quasi-steady state, and use of alternative feed strategies.

In the proposed mathematical model, the density of broth was assumed to be constant during the process. The following model equations are obtained by making material balances around the bioreactor and considering Eqs. (1, 3–5):

$$\frac{d[\text{HS}^-]}{dt} = \frac{F_{in}([\text{HS}^-]_{in} - [\text{HS}^-])}{V} - r_1 - r_3 + r_{-3} \quad (7)$$

$$\frac{dS^0}{dt} = \frac{-F_{in}S^0}{V} + r_1 - r_2 - (x-1)r_3 + (x-1)r_{-3} + (x-2)r_4 \quad (8)$$

$$\frac{d[\text{SO}_4^{2-}]}{dt} = \frac{-F_{in}[\text{SO}_4^{2-}]}{V} + r_2 \quad (9)$$

$$\frac{d[S_x^{2-}]}{dt} = \frac{-F_{in}[S_x^{2-}]}{V} + r_3 - r_{-3} - r_4 \quad (10)$$

$$\frac{d[S_2O_3^{2-}]}{dt} = \frac{-F_{in}[S_2O_3^{2-}]}{V} + r_4 \quad (11)$$

$$\frac{dV}{dt} = F_{in} - F_{out} \quad (12)$$

The reaction of HS^- with sulfur to produce polysulfide (Eq. (4)) was studied by Kleinjan et al. [16]. This reaction is almost quick; thus, HS^- can be assumed in equilibrium with S_x^{2-} during the

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