



Microbial selenium sulfide reduction for selenium recovery from wastewater



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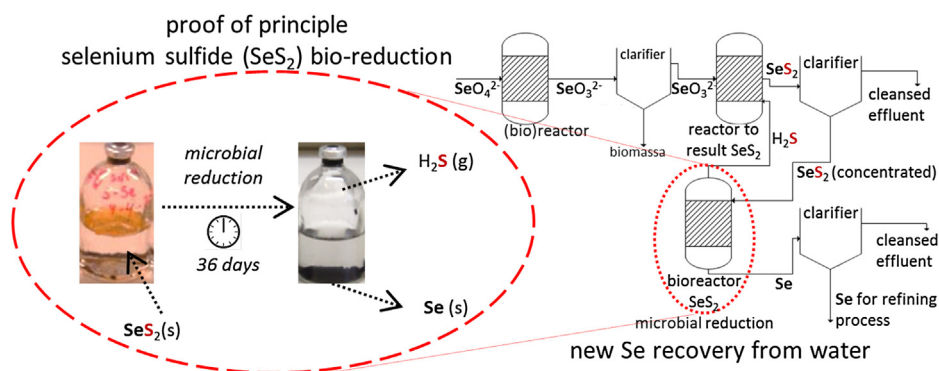
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HIGHLIGHTS

- Selenium sulfide is bio-reduced to selenium and sulfide by mixed cultures.
- Selenium particles have a high selenium purity and are hexagonal crystalline.
- Se particles are outside the biomass and this is beneficial to recover Se from water.
- Produced sulfide is recycled in the selenium recovery system.
- Se₂ bio-reduction and Se₂ precipitation conditions can be altered independently.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 27 July 2016

Received in revised form

28 December 2016

Accepted 30 December 2016

Available online 31 December 2016

Keywords:

Selenium removal and recovery

Microbial reduction

Wastewater

Selenium sulfide

Biocrystallization

ABSTRACT

Microbial reduction of selenium sulfide (SeS₂) is a key step in a new treatment process to recover selenium from selenate and selenite streams. In this process, selenate is first reduced to selenite, and subsequently selenite is reduced by sulfide and precipitates from the solution as SeS₂. The latter is bio-reduced to elemental selenium and sulfide. Two anaerobic granular sludges (Eerbeek and Emmtec) were tested for their efficiency to reduce commercial crystalline SeS₂. Emmtec sludge had the highest reducing capacity with commercial SeS₂ and was therefore also used for the bio-reduction of laboratory synthesized amorphous SeS₂. Synthesized SeS₂ was formed mixing a sulfide solution and effluent containing selenite. With both SeS₂ solids (commercial and synthesized SeS₂), Emmtec sludge produced sulfide and a solid consisting of hexagonal elemental selenium. The crystalline hexagonal structure suggests the absence of biomolecules, which stabilize amorphous selenium bio-particles under comparable process conditions (T = 30 °C and a pH between 6 and 7). Selenium particles were not attached to the biomass, suggesting an extracellular formation. The results support the feasibility of the bio-reduction process using sulfur for recovering selenium from water.

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1. Introduction

The bio-reduction of selenium sulfide (SeS₂) offers opportunities for the bio-recovery of selenium from wastewater streams. Selenate and selenite removal from such streams is sometimes essential for environmental safety [1–3]. The recovery of selenium is of its high value for industries [2,3]. An advantage of biological processes is its selectivity for substrates. Selenate can be bio-reduced to selenite [4], which can then be precipitated with sulfide to form SeS₂ [5,6]. If SeS₂ is further bio-reduced a new biological selenium removal and recovery process can be designed (see graphical abstract or for more details Supplementary information Fig. A1).

Abiotic selenite reduction with sulfide results in the formation of Se–S bonds in SeS₂. In SeS₂ both elemental redox states are zero [5] and SeS₂ consists of selenium sulfide heterocycles [6–8]. The S:Se ratio in SeS₂ depends on the precipitation conditions; for example, variation in pH may result in S:Se ratios ranging between 1.7–2.3 [5].

Selenite reduction with (bio)molecules containing thiol-groups such as glutathione [9,10] and cysteine [11] also resulted in Se–S bonds. However, the Se–S bonds thus produced were further

(bio)processed resulting to produce elemental selenium among other products [9]. In a previous study, abiotic leaching tests of SeS₂ resulted in a stable product [5]. Thus raising the question as to whether SeS₂ can also be further bio-processed into elemental selenium.

While in principle both selenium and sulfur can serve as the electron acceptor in SeS₂, sulfur is thermodynamically the more favorable electron acceptor (compare the Gibbs energy yields of reactions 1 and 2 in Table 1) [7,12]. Thus, in mixed microbial communities reduction of SeS₂ will result in the production of elemental selenium plus sulfide. As methanogenesis (reaction 6 in Table 1) generates comparable Gibbs free energy per electron as SeS₂ reduction, methane production should be avoided by choosing adequate process conditions, e.g. by removing CO₂.

We postulate that the selenium can be purified and recovered from SeS₂ by bio-reducing the sulfur in SeS₂ to sulfide. Besides commercially pure SeS₂, synthesized SeS₂ produced with selenite effluent from a selenate to selenite converting bioreactor [4] was also tested for bio-reduction. The latter SeS₂ may contain impurities that affect the biological reduction.

Table 1
Standard Gibbs free energy change of selenium reactions and methane formation at 30 °C and pH = 7.

Number	Reaction	ΔG° per reaction (kJ/mol)	ΔG° Per electron (kJ/electron mol)
(1)	$2\text{H}_2(\text{aq}) + \text{SeS}_2^{\text{a}} \rightarrow \text{Se} + 2\text{HS}^- + 2\text{H}^+$	–93.1	–23.3
(2)	$\text{H}_2(\text{aq}) + \text{SeS}_2^{\text{a}} \rightarrow \text{HSe}^- + 2\text{S} + \text{H}^+$	–14.9	–7.5
(3)	$2\text{HSe}^- + \text{SeS}_2^{\text{a}} \rightarrow 3\text{Se} + 2\text{HS}^-$	–64.6	–16.1
(4)	$\text{HS}^- + \text{SeS}_2^{\text{a}} \rightarrow 3\text{S} + \text{HSe}^-$	31.3	15.6
(5) ^a	$\text{SeS}_2^{\text{a}} \rightarrow \text{Se} + 2\text{S}$	–0.7	not applicable
(6)	$\text{CO}_2(\text{aq}) + 4\text{H}_2(\text{aq}) \rightarrow \text{CH}_4(\text{aq}) + 2\text{H}_2\text{O}(\text{l})$	–193.1	–24.1
(7)	$\text{HSeO}_3^-(\text{aq}) + 2\text{HS}^-(\text{aq}) \rightarrow \text{SeS}_2^{\text{a}} + 3\text{HO}^-$	–204.5	–51.1
(8)	$\text{HSe}^- + \text{S} \rightarrow \text{HS}^- + \text{Se}$	–32.0	–16.0

Data calculated from Amend and Shock [12].

^a ΔG° , in line 5 is estimated using the average energy change of Se–S bonds into S–S and Se–Se bonds obtained from Taavitsainen et al. [7]. Average bonds in (SeS₂): 1/9 Se–Se; 4/9 Se–S; 4/9 S–S (See Supplementary material).

Table 2
Overview of the SeS₂ reduction experiments.

2A commercial crystalline SeS ₂ bioreduction (sequence 1)							
bottle no.	Label	Lactate 100 mM	Eerbeek	Emmtec	Initial commercial SeS ₂ added & concentration		Total Volume
		(mL)	(g ww)	(g ww)	(mg)	(mM Se)	(mL)
#1–1	Control SeS ₂	5	–	–	(71.5)	(9.1)	50
#1–2	Control Eerbeek	5	0.5	–	(0.0)	(0.0)	55.5
#1–3	Control Emmtec	5	–	0.5	(0.0)	(0.0)	55.5
#1–4	Eerbeek & SeS ₂	5	0.5	–	(72.0)	(9.1)	55.5
#1–5	Eerbeek & SeS ₂ (duplicate #1–4)	5	0.5	–	(73.6)	(9.3)	55.5
#1–6	Emmtec & SeS ₂	5	–	0.5	(70.6)	(8.9)	55.5
#1–7	Emmtec & SeS ₂ (duplicate #1–6)	5	–	0.5	(73.0)	(9.2)	55.5
#1–8	Emmtec & Eerbeek & SeS ₂	5	0.25	0.25	(706.8)	(89.1)	55.5
2B synthesized amorphous SeS ₂ bioreduction (sequence 2)							
bottle no.	Label	Lactate 100 mM	Emmtec	old medium with SeS ₂	synthesized SeS ₂	Fresh medium	Total Volume
		(mL)	(g ww)	(mL)	(mM Se)	(mL)	(mL)
#2–1	synthesized SeS ₂ (control)	5	–	50	9.6 ^a	–	55.0
#2–2	Emmtec (control)	5	0.5	–	0.0	50	55.5
#2–3	Emmtec & synthesized SeS ₂	5	0.5	50	9.5 ^a	–	55.5
#2–4	Emmtec & synthesized SeS ₂ (duplicate #2)	5	0.5	50	9.5 ^a	–	55.5

^a Selenium content (mM Se) in synthesized Se–S solution was measured by ICP and corrected for the dilution 50 mL/total volume.

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