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Selenium removal using a bacteria cultured reactor packed with alum-impregnated activated alumina

Yuxia Ji[1], Yi-tin Wang[1]
[1]University of Kentucky
Department of Civil Engineering, University of Kentucky,
Lexington, KY, USA
yuxia.ji@uky.edu; ywang@uky.edu

Abstract **–** Selenium is a primary pollutant that brings a number of threats to the aquatic lives as well as human beings. Bacterial selenium reduction was observed as an effective way for selenium remediation. However, Se(IV) accumulation may occur during Se(VI) reduction due to the slow reduction rate of Se(IV). In this study, an alum-impregnated activated alumina packed bioreactor cultured with *Shigella fergusonii* strain TB42616 was applied in order to remove Se(IV) during Se(VI) reduction. Approximately 70% selenium was removed under an hydraulic retention time of 3.1 days with no Se(IV) accumulation observed, indicating that Se(IV) removal was enhanced by the adsorption of the alum-impregnated activated alumina. A 10% more selenium removal was also observed by employing alum-impregnated activated alumina instead of unmodified ones.

Keywords**:** Adsorption, Selenium, Reduction, Packed-bed reactor.

1. Introduction

Selenium becomes one of the concerning pollutants in recent years. Anthropogenic activities are one of the major sources contributing up to 40% global selenium emission, including metal refinery and processing, fossil fuel combustion, mining, agricultural irrigation as well as pigment manufacturing [1]. Although beneficial in trace amount, selenium at high doses can cause health adverse effects such as dermal diseases and dermatitis, hair loss, nail abnormalities, neurological impairment, etc., due to its toxicity [2]. Selenium at high concentrations also brings huge risks to the aquatic lives, especially to the egg-laying vertebrates, and causes reproductive impairment, mutagenicity and potential cancers [3]. As a result, USEPA regulated selenium concentration as 3.1 μg/L in lotic fresh water, 2 μg/L in lentic fresh water and 50 μg/L in effluent discharge [4].

Se(VI) and Se(IV) are the two predominant species in water due to their high solubility and low adsorption by soil and sediments[5]. Selenium removal technologies used recently include bacterial reduction, adsorption using activated alumina and ferrihydrite, membrane process, ion-exchange, etc. [6]. Bacterial selenium reduction is one of the most efficient as well as economic ways for selenium removal among all these technologies as it is capable for both Se(VI) and Se(IV) reduction. Bacterial species involved in selenium reduction have been studied for years including *Pseudomonas stutzeri* [7], *Bacillus subtilis* [8], *Enterobacter cloacae* [9], *Bacillus selenitireducens* [10], *Escherichia coli* [11], etc. However, it was reported that Se(IV) accumulation may potentially occurred during Se(VI) reduction due to the relatively slower Se(IV) reduction rate than Se(VI) reduction[11, 12]. Considering that Se(IV) is even more toxic than Se(VI) [13, 14], the total toxicity may correspondingly increase.

In this study, the activated alumina was modified with alum first and then packed as the bacterial attachment media in order to remove Se(IV) accumulation during bacterial Se(VI) reduction. The objective of this study is to evaluate the overall selenium removal by a packed-bed reactor using alum-impregnated activated alumina with a Se(VI)-reducing strain, *Shigella fergusonii strain* TB42616.

2. Methods

2.1. Bacterial strain

A Se(VI)-reducing strain, *Shigella fergusonii strain* TB42616, was used in this study. The bacterial strain was isolated from sludge samples collected from the aeration tank at Town Branch Wastewater Treatment Plant located in Lexington, Kentucky as described by Ji and Wang [15]. The strain was identified as *Shigella fergusonii* using 16s rRna sequencing with

99% similarity after purification through the streak method. The purified strain was preserved at 4 °C and transferred to a fresh nutrient agar plate every three weeks to prevent mutation.

2.2. Alum-impregnated activated alumina

The alum-impregnated activated alumina was prepared as described by Tripathy et al. [16] by adding 200 mL of 1M $Al_2(SO_4)_3 \cdot 18H_2O$ and 200 mL of 5% $NaHCO_3$ to 100 g activated alumina with pH adjusted to 3.4 ± 0.1 using 0.1M HCl. The activated alumina was immersed in the solution for 16 hours for reaching equilibrium and then subsequently washed thoroughly with deionized water. The washed pellets were dried in a biological cabinet for 5 days at ambient temperature and then preserved in a reagent bottle at ambient temperature.

2.3. Growth medium

A modified chemically defined medium (MCDM) used as the feed growth medium in this study consisted of 300 mg/L $(NH_4)_2SO_4$, 200 mg/L $CaCl_4 \cdot 2H_2O$, 70 mg/L $MgSO_4$, 5850 mg/L NaCl, 0.6 mg/L H_3BO_4, 0.08 mg/L $CoSO_4$, 0.08 mg/L $CuSO_4$, 0.63 mg/L $MnCl_2$ and 0.22 mg/L $ZnCl_2$ supplemented with a pH buffer of 3000 mg/L $NaHCO_3$ and 1000 mg/L glucose as carbon source. 50 mg/L Se(VI) or Se(IV) was added to feed medium. The medium was autoclaved at 121°C for 15 minutes and preserved at 4°C until use.

2.4. Reactor configuration and startup

A Pyrex glass column with a height of 12 cm and a diameter of 2.5 cm was used as the reactor packed with 40 g alumina pellets of 3 mm in diameter (Figure 1). The total volume of reactor was measured as 74 cm^3, while the cross-section area was measured as 76 cm^2. All the connected tubings and the reactor parts were assembled under a biological cabinet after autoclaved at 121°C for 15 minutes. A calibrated peristaltic pump was used to feed MCDM into the reactor throughout the study.

Three reactors were run simultaneously including two non-bacteria seeded reactors and one bacterial seeded reactor. The non-bacterial seeded reactors were fed with the MCDM without glucose supplemented, respectively. While, bacterial strains were inoculated in the bacteria seeded reactor after harvesting overnight-grown cells using a centrifuge at 5000 rpm and then washed three times with 0.85% NaCl.

2.5. Analytical method

Se(VI) and Se(IV) were measured using the colorimetric method after formation of florescent piazselenol compound according to Section 3500 C of the Standard Methods for the Examination of Water and Wastewater [17]. The pH was measured by a pH meter, while the cell density was measured by a cell counter.

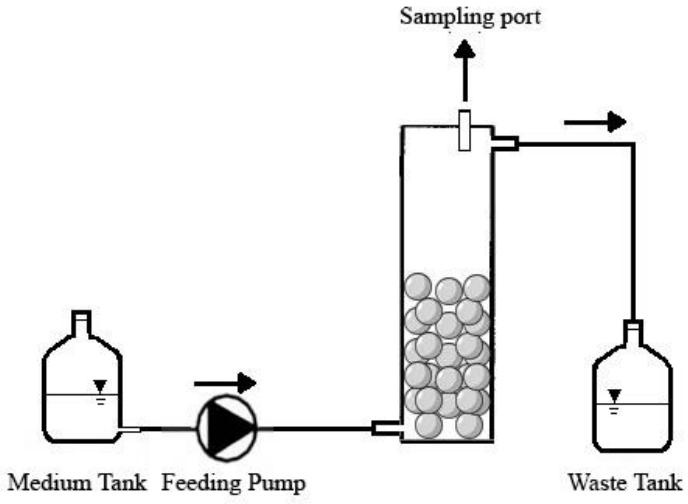

Fig. 1: The configuration of the packed-bed reactor.

3. Results and conclusion

A batch study was carried out first to evaluate Se(VI) reduction by the pure culture of *Shigella fergusonii* strain TB42616. A two-stage process of Se(VI) reduction was observed. The data in Figure 2 show that Se(VI) was reduced to Se(IV) and then to Se(0). Se(IV) accumulation was also observed in this study with a highest concentration of 6.6 mg/L at the 2nd day. This observation was consistent with recent studies that Se(IV) was accumulated during Se(VI) reduction by a pure culture [11, 15]. In this study, up to 33% Se(IV) was accumulated at an initial Se(VI) concentration of 20 mg/L, while such ratio may increase with the initial Se(VI) concentration. A previous study reported that Se(IV) reduction completely ceased at a certain level at an initial Se(VI) concentration of 400 mg/L after 6 days incubation with *E.Coli* culture [12].

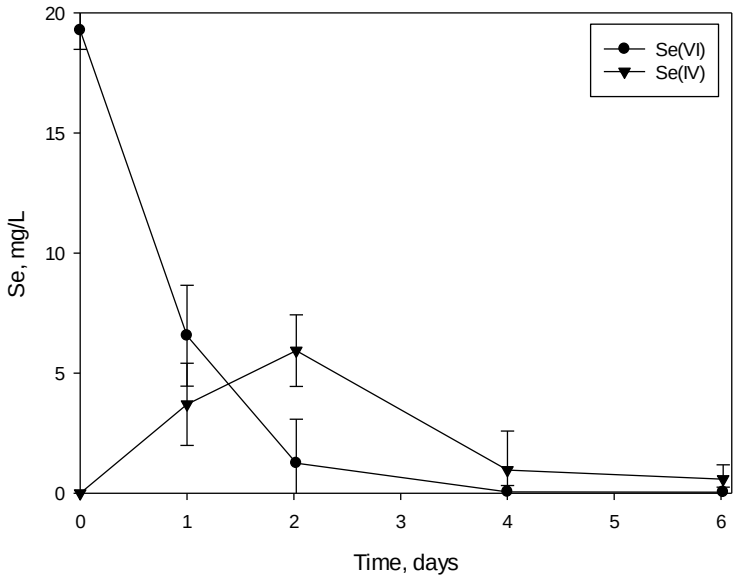

Fig. 2: Se(VI) reduction by a pure batch culture of *Shigella fergusonii* strain TB42616.

Activated alumina has already been proved able to adsorb Se(IV) in many studies. A previous study reported that activated alumina was effective for Se(IV) adsorption but not for Se(IV) [18]. Thus, activated alumina had a substantially higher adsorption capacity of Se(IV) than Se(VI). In this study, a modified activated alumina, alum-impregnated activated alumina, was used as it was reported to adsorb fluoride and As(V) more efficiently than unmodified ones [16, 19]. As a result, the alum-impregnated activated alumina was used in this study.

Selenium removal was investigated with a continuous-flow reactor packed with alum-impregnated activated alumina under a hydraulic retention time of 3.1 days at 30°C and pH 7. Se(VI) and Se(IV) at 50 mg/L were fed into the reactors by a peristaltic pump, respectively. As shown in Figure 3, the breakthrough of Se(VI) in the non-bacteria seeded reactor occurred after the 6th day and became saturated at day 16, while the breakthrough of Se(IV) did not begin at 37th day. This observation further proved that Se(IV) adsorption capacity of alum-impregnated activated alumina was significantly higher than Se(VI). However, Se(IV) accumulation was not observed in the reactor even after 37 days in the bacteria seeded reactor evidenced by relatively low Se(IV) concentration (<1 mg/L) throughout the experiment. Consequently, Se(IV) accumulation was significantly reduced by the alum-impregnated activated alumina. However, Se(VI) concentration of 17 mg/L still was detected in the effluent representing approximate 70% removal, indicating that the hydraulic retention time may not be sufficient to reduce Se(VI) completely.

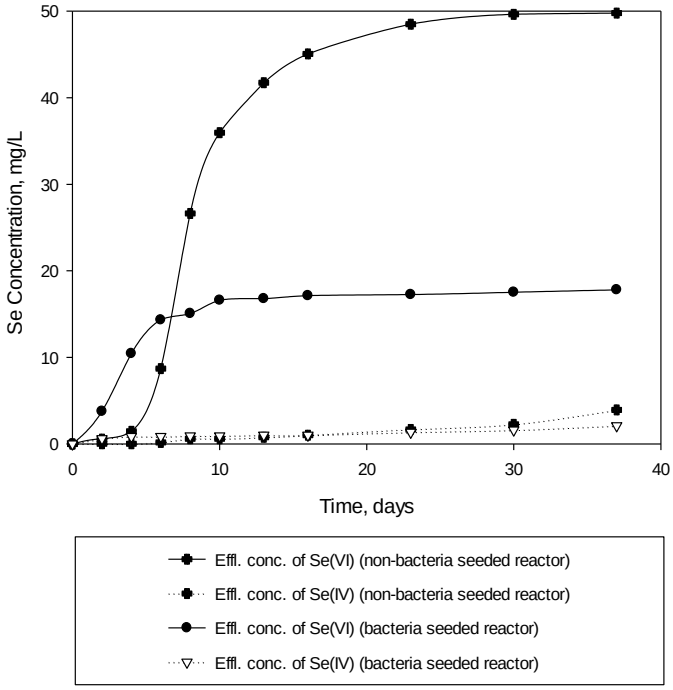

Fig. 3: Effluent concentrations of Se(VI) and Se(IV) from alum-impregnated activated alumina packed reactors.

The mass balance of selenium in the reactor is shown in Figure 4. It clearly indicated that the output of selenium mass equalled the input mass after the 16 days (exhaustive phase) in the non-bacteria seeded reactor. Significant difference between the input selenium and output of the soluble selenium was observed, indicating that selenium was substantially removed. The amount of Se(VI) reduced by *Shigella fergusonii* strain TB42616 was revealed by the difference between the Se(VI) output from the non-bacteria seeded reactor and from the bacteria seeded reactor. The increasing amount reduced Se(VI) indicated that the cultured Se(VI)-reducing strain was effective in reducing Se(VI) to Se(IV) which was absorbed subsequently by the activated alumina. The application of alum-impregnated activated alumina the increased selenium removal by more than 10% compared to the unmodified ones as reported by a previous study [20]. The alum-impregnation of activated alumina expanded both Se(VI) and Se(IV) adsorption capacity, thus extending the lifetime of the activated alumina.

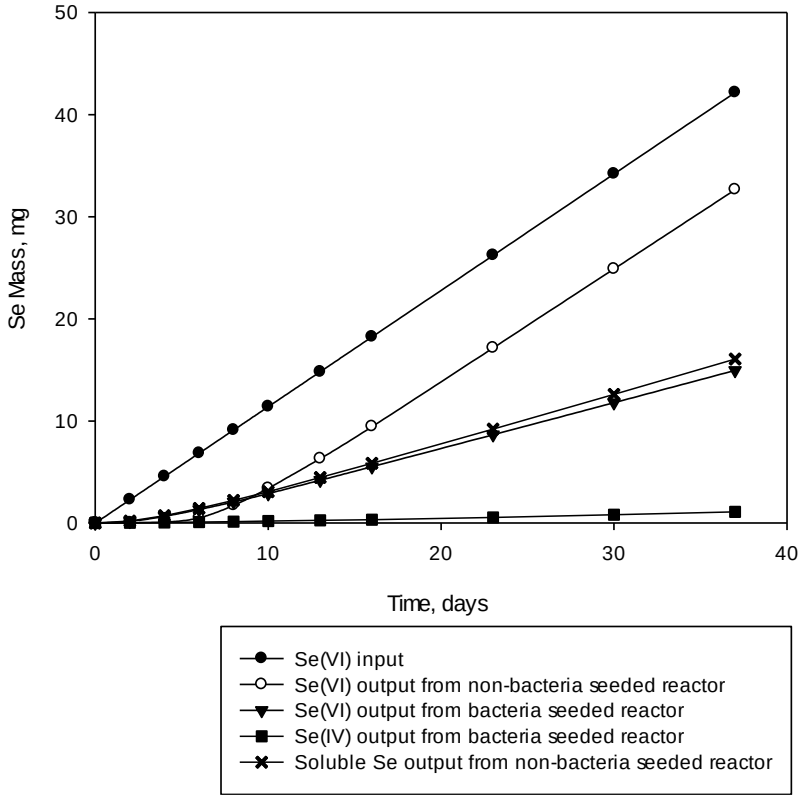

Fig. 4: Cumulative selenium input as well as out from the alum-impregnated activated alumina packed reactors.

4. Conclusion

In this study, selenium removal was investigated using alum-impregnated activated alumina packed reactor cultured with *Shigella fergusonii* strain TB42616. Se(IV) was observed a more favourable adsorbate for alum-impregnated activated alumina than Se(VI). Approximately 70% selenium was removed under a hydraulic retention time of 3.1 day. No Se(IV) accumulation was observed throughout this study, indicating that Se(IV) removal was enhanced by adding alum-impregnated activated alumina compared to the single bacterial selenium reduction process. In addition, the removal efficiency was enhanced by 10% as alum-impregnated activated alumina rather than nonmodified ones was used.

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