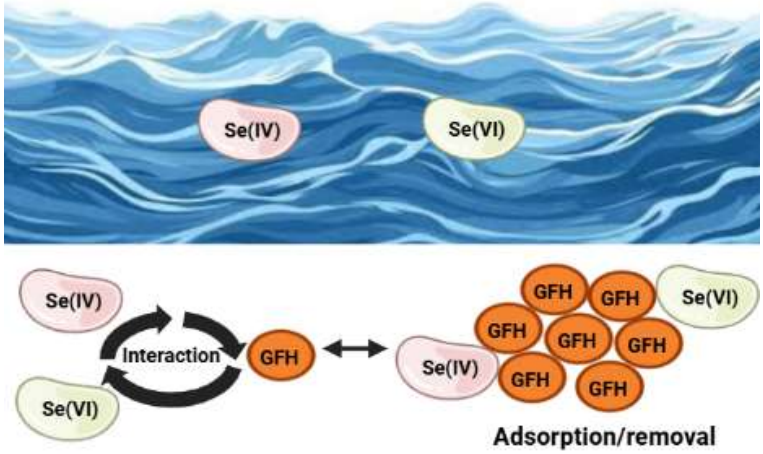
1.10 ReSeO: Removal of selenium oxyanions from drinking water by batch and continuous mode adsorption on granular ferric hydroxide



Project Duration

01.12.2024 – 30.11.2026

Funding

Alexander Von Humboldt Stiftung

Figure 4: Adsorptive removal of selenium oxyanions using granular ferric hydroxide.

Introduction

High concentrations of ionic contaminants such as selenium (Se) in groundwater pose serious risks to human and environmental health. Se oxyanions, originating from mining, petroleum refining, fossil fuel combustion, and irrigation, have been detected at extremely high levels in drinking and surface water bodies around the globe [1]. Although Se is an essential micronutrient, excessive intake (>400 µg/day) can cause severe health issues such as reproductive and developmental disorders, hair loss, muscle damage, organ failure, cancer, and even death. To ensure safe drinking water, regulatory limits for Se have been set at 10 µg/L (Germany), 40 µg/L (WHO), and 50 µg/L (U.S. EPA) [2,3]. Commercial iron-based adsorbents, particularly granular ferric hydroxide (GFH), have shown strong potential for removing various oxyanions like arsenic hate [4]. However, Se removal remains challenging due to its toxicity, solubility, and multiple oxidation states (selenate and selenite) depending on pH and redox conditions. Moreover, little is known about Se–GFH interactions, long term GFH stability, or iron release under batch and continuous flow mode operational conditions. The planned project aims to address these limitations, develop effective understanding and practical solutions for Se removal from drinking water.

Research Goals

The ongoing research project comprises the following three innovative and important objectives: (i) Unravelling Se(IV) and Se(VI) removal behaviour in aqueous matrices using commercial iron-based adsorptive material i.e. GFH in systematically designed batch-mode sorption experiments; (ii) Investigating the optimum boundary conditions for removal of selenium in small-scale adsorption filters relevant for practical applications in drinking water treatment; (iii) Conducting a techno-economic feasibility analysis of developed water filtration technology for Se removal from real source waters of drinking water supply.

Approach

At the beginning of the project, calibration curves for Se(IV) and Se(VI) were developed using ion chromatography (IC) and atomic absorption spectrophotometry (AAS) to enable accurate quantification of both species. These calibration curves with R^2 values of 0.999 allowed reliable detection and speciation of Se at concentrations as low as 2 µg/L, ensuring high analytical precision throughout the study. Batch laboratory experiments were conducted using 100 mL solution in 250 mL conical flasks with continuous shaking using a mechanical shaker to evaluate the adsorption performance of GFH for Se(IV) and Se(VI), as well as to assess potential iron leaching into the treated water. For Se(IV), all solution preparations and experiments were performed under anoxic conditions following a 5-minute nitrogen gas purge. A wide range of solution chemistries was examined, including the influence of GFH dosage, pH, contact time, Se speciation and initial concentration, and the presence of coexisting ions. These parameters were systematically investigated to better understand their impact on Se removal efficiency and GFH stability. Adsorption kinetics (Pseudo first and second order) and equilibrium isotherm models (Langmuir and Freundlich) were applied to the experimental data to gain deeper mechanistic insight into the removal behavior of Se(IV, VI) by GFH. These models helped elucidate the governing adsorption mechanisms, surface interactions, and maximum adsorption capacity of GFH for Se species in water. These experimental findings will directly support the design and optimization of a continuous-mode, small-scale GFH adsorption unit for the removal of Se oxyanions from real raw water samples.

Recent Results

The results of the batch experiments conducted across a range of GFH dosages (0–5 g/L) are summarized in Figure 2. GFH exhibited a significantly higher removal efficiency for Se(IV) than for Se(VI), even at low dosages. At 0.5 g/L GFH, Se(IV) removal reached 90.2%, successfully meeting the drinking water guideline of 10 µg/L. In contrast, only 28.5% of Se(VI) was removed under the same conditions, highlighting the critical role of Se speciation on treatment performance. Although increasing the GFH dosage to 2 g/L improved Se(VI) removal to 88.4%, this level may still pose health concerns, emphasizing the challenges associated with Se(VI) remediation. The corresponding adsorption capacities at 0.5 g/L GFH dosage further emphasize this preferential affinity for Se(IV), with GFH exhibiting a threefold higher capacity for Se(IV) (0.180 mg/g) than for Se(VI) (0.057 mg/g). At higher dosages, adsorption capacity declined, likely due to saturation of available adsorption sites, reduced Se concentration in solution, and overdosing effects. To determine whether GFH stability contributed to the observed differences in removal performance, residual iron in solution was also measured. Only negligible iron leaching was detected, up to 0.107 mg/L for Se(IV) and 0.008 mg/L for Se(VI), thus indicating that GFH stability was not responsible for the variations in removal efficiency. This confirms that differences in Se–GFH interactions and solution chemistry, rather than adsorbent dissolution, play the dominant role in governing adsorption behaviour.

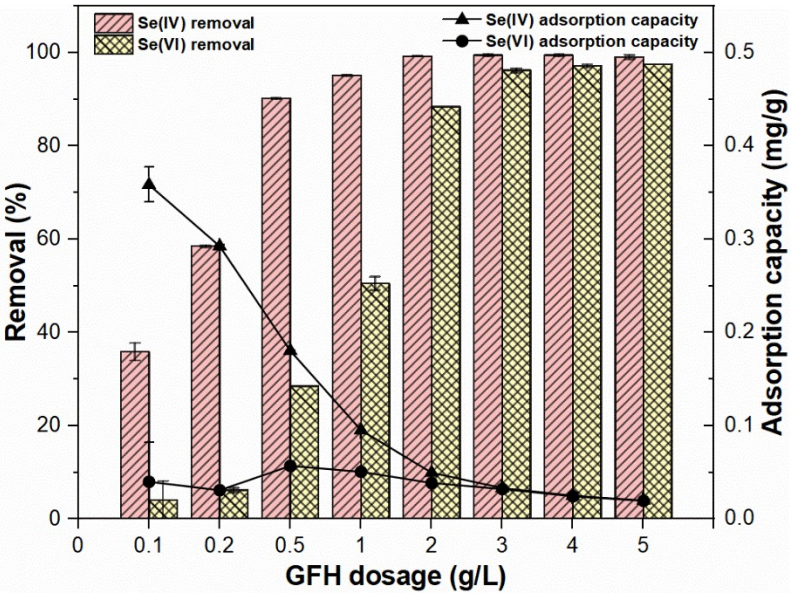


Figure 2: Influence of GFH dosage (0.1-5 g/L) on removal performance (%) and adsorption capacity (mg/g) of Se(IV, VI) species under controlled pH (7), sodium bicarbonate (1 mM), contact time (2 hours) and initial Se(IV, VI) concentrations (100 µg/L).

The kinetic and isotherm model parameters for Se(IV) and Se(VI) adsorption onto GFH are presented in **Table 1**. Kinetic experiments were conducted at neutral pH, 100 µg/L initial Se(IV) and Se(VI) concentrations, 2 g/L GFH dosage for different reaction times (0 to 48 hours). Moreover, isotherm experiments were conducted at neutral pH, 2 g/L GFH dosage, equilibrium reaction time (2 hours) for different initial Se(IV) and Se(VI) concentrations (10 µg/L to 10 mg/L). Under the given conditions, the maximum adsorption capacity (q_m) at the equilibrium reaction time (t) of 2 hours was higher for Se(IV) (7.086 mg/g) compared to Se(VI) (4.475 mg/g). The fitting parameters indicate that Se(IV) adsorption onto GFH aligns more closely with the pseudo first order (PFO) kinetic model and the Langmuir isotherm, suggesting a predominantly monolayer, homogeneous adsorption process. In contrast, Se(VI) adsorption is better described by the pseudo second order (PSO) kinetics and the Freundlich isotherm, implying a more heterogeneous and multilayer adsorption behavior. Overall, these mechanistic insights and differences underscore the need for adsorption process specifically optimized for the targeted Se(IV) and Se(VI) species present in contaminated water.

Table 1: Kinetic and isotherm model parameters for adsorption of Se(IV) and Se(VI) onto GFH.

Models	Ions	Parameters		R^2
Kinetic				
PFO		q_e (mg/g)	k_1 (1/h)	
	Se(IV)	0.049	7.260	0.992
	Se(VI)	0.042	3.864	0.964
PSO		q_e (mg/g)	k_2 (mg/g.h)	
	Se(IV)	0.050	324.877	0.985
	Se(VI)	0.044	163.223	0.975
Isotherm:				
Langmuir		q_m (mg/g)	k_L (L/mg)	
	Se(IV)	7.086	4.648	0.926
	Se(VI)	4.475	0.588	0.991
Freundlich		n	k_F [(g/mg)(L/mg)]1/n	
	Se(IV)	1.993	7.289	0.923
	Se(VI)	1.717	1.507	0.998

Conclusion and Outlook

The results indicate that GFH exhibits a higher adsorption affinity for Se(IV) than for Se(VI) under the tested conditions. Kinetic and isotherm analyses revealed that the adsorption of Se(IV) species followed the PFO and Langmuir models, while Se(VI) adsorption onto GFH was better described by PSO and Freundlich models. Notably, iron leaching remained insignificant across varying redox conditions, confirming the stability of GFH in the operational environment. Studies on the influence of coexisting ions on Se(IV) and Se(VI) removal are currently ongoing. Since these experiments were performed in synthetic batch systems, further research will assess adsorption performance in artificially prepared and real water samples using a small scale continuous GFH unit with the possibility of regeneration of GFH media. A response surface methodology will be applied to optimize operating parameters and achieve the target drinking water limit of 10 µg/L Se. Additionally, spent GFH will be characterized using advanced analytical techniques, and the economic feasibility of the treatment units will be evaluated. Collectively, these efforts will support the development of a practical and scalable Se removal system for field applications.

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