Use of drinking water treatment solids for arsenate removal from desalination concentrate
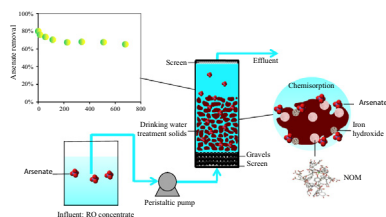
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GRAPHICAL ABSTRACT

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ABSTRACT

Desalination of impaired water can be hindered by the limited options for concentrate disposal. Selective removal of specific contaminants using inexpensive adsorbents is an attractive option to address the challenges of concentrate management. In this study, two types of ferric-based drinking water treatment solids (DWTS) were examined for arsenate removal from reverse osmosis concentrate during continuous-flow once-through column experiments. Arsenate sorption was investigated under different operating conditions including pH, arsenate concentration, hydraulic retention time, loading rate, temperature, and moisture content of the DWTS. Arsenate removal by the DWTS was affected primarily by surface complexation, electrostatic interactions, and arsenate speciation. Results indicated that arsenate sorption was highly dependent on initial pH and initial arsenate concentration. Acidic conditions enhanced arsenate sorption as a result of weaker electrostatic repulsion between predominantly monovalent $H_2AsO_4^-$ and negatively charged particles in the DWTS. High initial arsenate concentration increased the driving force for arsenate sorption to the DWTS surface. Tests revealed that the potential risks associated with the use of DWTS include the leaching of organic contaminants and ammonia, which can be alleviated by using wet DWTS or discarding the initially treated effluent that contains high organic concentration.

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1. Introduction

1.1. Reverse osmosis (RO) concentrate treatment

Desalination of seawater, brackish water, and reclaimed water has been a viable solution to providing alternative water supplies. Reverse osmosis (RO), nanofiltration, electrodialysis, and thermal distillation are well-established desalination technologies, producing fresh water for industrial, domestic, and agricultural uses [1–4]. However, with increasing water recovery (ratio of product to feed water) during desalination, the concentrations of dissolved constituents in the concentrate stream increase. Consequently, proper disposal of brines, and particularly those containing elevated concentrations of toxic contaminants, including heavy metals, is one of the primary impediments for implementation of desalination technologies [5].

Ocean discharge is widely used by desalination facilities in coastal areas, but concentrate disposal remains a major challenge

for inland plants where disposal options are limited by the quantity and quality of concentrate, regulations, and geographical and geological constrains. A recent review of technologies for concentrate treatment was published, focusing on concentrate volume minimization and beneficial uses [6]. However, most of these technologies are often costly and energy intensive. Selective removal of specific contaminants (e.g., arsenic and heavy metals) using inexpensive materials could be an alternative to meeting disposal requirements or water quality criteria for beneficial use applications [5].

1.2. Arsenic removal technologies

If present in water and consumed in large quantities, arsenic, a metalloid abundant in nature in the form of organic and inorganic compounds, can cause a variety of pathological conditions, including cutaneous and visceral malignancies [7]. The acute minimal lethal dose of arsenic in adults is 70–200 mg per day, or 1 mg per kg weight per day [8]. The United States Environmental Protection Agency (USEPA) has set an enforceable regulatory limit for arsenic in drinking water at 0.01 mg L^{-1} (10 ppb).

A variety of physicochemical techniques are being utilized for arsenic removal, including coagulation [9], ion exchange [10], RO [11], liquid–liquid extraction [12], and sorption [13]. Specifically, sorption using ferric-based sorbents is an effective treatment technology because of the high sorption capacity of iron for arsenic and simple operation. Table 1 summarizes the arsenic sorption capacity of different sorbents. Studies have shown that these materials have a strong affinity for arsenic under natural pH conditions compared to commonly used activated alumina. For sorption processes, alternative sorbents that meet the requirement of cost-effectiveness and eco-friendly disposal are needed. A low-cost and potentially effective substitute for arsenic sorbents could be the solid residuals from coagulation/flocculation water treatment processes, which often utilize ferric-based coagulants [14].

1.3. Drinking water treatment solids (DWTS)

Drinking water treatment solids (DWTS) are the residuals produced during water treatment using iron or aluminum salts

as primary coagulants. Laboratory studies have demonstrated that DWTS have strong affinity to sorb contaminants such as phosphorus [15,16], hydrogen sulfide [17], metals [18,19], fluoride [20], and arsenic [5,21]. Laboratory batch equilibration studies have shown that both ferric-based and aluminum-based DWTS have a high affinity for arsenite [As(III)] and arsenate [As(V)] species [22]. Gibbons and Gagnon examined the sorption of arsenic from groundwater in batch and column experiments using the solids from different water treatment plants [23]. The results showed that ferric and lime solids were effective sorbents for arsenic removal [23]. The arsenic removal mechanism by DWTS involves mainly inner-sphere complexation with ferric hydroxides [24], which is affected by the ferric mass in the DWTS [25,26].

The factors influencing arsenic sorption include pH, loading rate, initial arsenic concentration, competing ligands or complexing metals, temperature, and specific physicochemical properties of the adsorbing solids (e.g., specific surface area, total carbon content, porosity) [22,27,28]. X-ray absorption spectroscopy demonstrated that strong, inner-sphere complexes are formed between sorbed arsenic and the Al/Fe-hydroxide components of the solids [29,30]. Arsenic leaching tests indicated that 50–60% of the total arsenic in DWTS was bound strongly on amorphous iron hydroxides [31]. Maintaining non-alkaline and high redox potential conditions is critical for minimal arsenic mobilization in DWTS [31]. Thermodynamic studies indicate that arsenic sorption increases with increasing water temperature [32].

Although previous studies have demonstrated the feasibility of arsenic removal using DWTS (Table 1), there is lack of knowledge on the removal efficiency of arsenic from saline water, such as desalination concentrate, in which competition with co-existing ions and high ionic strength may affect the selective sorption of arsenic. Therefore, this study focused on investigating sorption of arsenate from RO concentrate using different types of DWTS during continuous-flow operation. The impacts of operating conditions on arsenate removal were investigated by performing experiments at different pH, initial arsenate concentration in RO concentrate, hydraulic retention time, temperature, and loading rate using different types of DWTS.

Table 1
Summary of results from arsenic sorption studies using various sorbents under different testing conditions.

Adsorbent	Type of water	pH	As concentration	Temp. (°C)	Operating condition and models for calculating adsorption capacity	Adsorption capacity (mg/g solids)		Refs.
						As(III)	As(V)	
Char carbon	Synthetic 0.1 M NaCl solution	2–3	157-737 $\mu g\,L^{-1}$ for As(V); 193-992 $\mu g\,L^{-1}$ for As(III)	25	Batch	89.0	34.46	[33]
Activated carbon	Synthetic 0.1 M NaCl solution	6.4–7.5	157-737 $\mu g\,L^{-1}$ for As(V); 193-992 $\mu g\,L^{-1}$ for As(III)	25	Batch	29.9	30.48	[33]
Iron oxide coated sand	Drinking water	7.6	100 $\mu g\,L^{-1}$	22 ± 2	Batch & Langmuir	0.041	0.043	[34]
Ferric-based water treatment residual	Ground water	8.0–8.2	38.8-47.2 $\mu g\,L^{-1}$	22	Batch & Langmuir	–	2.23	[35]
Lime-based water treatment residual	Ground water	8.0–8.2	38.8-47.2 $\mu g\,L^{-1}$	22	Batch & Langmuir	–	0.16	[35]
Activated aluminum	Drinking water	7.6	1 mg L^{-1}	25	Batch & Langmuir	0.180	–	[36]
MnO$_2$	Drinking water	7.9	<1 mg L^{-1}	25	Column & Langmuir	–	0.172	[37]
Hydrous ferric oxide (HFO)	Drinking water	9.0	0-60 mg L^{-1}	22	Batch	28.0	7.0	[38]
Zirconium-loaded activated carbon (Zr-AC)	Drinking water	8–9	5-100 mg L^{-1}	25	Column	–	2.8	[39]
Granular ferric hydroxide (GFH)	Drinking water	8–9	5-100 mg L^{-1}	25	Column	–	2.3	[39]
Activated alumina grains	Drinking water	5.2 for As(V); 7.0 for As(III)	2.85-11.5 mg L^{-1} for As(V); 0.79-4.90 mg L^{-1} for As(III)	25	Batch & Langmuir	3.48	15.9	[40]
FeCl$_3$ treated tea fungal biomass	Ground water	7.20	0.9 mg L^{-1} for As(V); 1.3 mg L^{-1} for As(III)	30	Batch & Freundlich	5.4	10.26	[41]