



Sorption of metals and metalloids from reverse osmosis concentrate on drinking water treatment solids



Lu Lin^a, Xuesong Xu^a, Charalambos Papelis^a, Tzahi Y. Cath^b, Pei Xu^{a,*}

^a Department of Civil Engineering, New Mexico State University, 3035 S Espina Street, Las Cruces, NM 88003, USA

^b Department of Civil and Environmental Engineering, Colorado School of Mines, 1500 Illinois Street, Golden, CO 80401, USA

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ABSTRACT

Drinking water treatment solids (DWTS) from coagulation processes could be a low-cost sorbent for selective removal of certain metals and metalloids from brine. In this study, continuous-flow sorption processes and factors affecting sorption of boron (III), copper (II), chromium (VI), lead (II), and selenium (VI) from reverse osmosis (RO) concentrate were investigated using different types of iron-derived DWTS. DWTS was demonstrated to be very effective in removal of copper and lead from RO concentrate, attributed to formation of strong inner-sphere complexes between the metal cations and Fe/Al oxide surface sites in the DWTS, as well as attractive electrostatic interactions with the negatively charged DWTS surface. Sorption of boron and selenium on the DWTS was lower. Boron exists primarily as nonionic boric acid at operating pH of 5.5–7.8, while selenate is a weakly bonding anion typically assumed to form outer-sphere surface complexes with Fe/Al oxides, both resulting in low removal. Sorption of the metals and metalloids was affected by their speciation and electrostatic interactions with DWTS. Higher influent pH increased boron and copper sorption but inhibited removal of chromium. Water temperature, solid moisture content, and loading rate did not have significant impact on sorption to the DWTS.

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1. Introduction

The presence of metals and metalloids such as chromium (Cr), copper (Cu), lead (Pb), boron (B), and selenium (Se) in water can adversely impact microorganisms, plants, and animals [1]. More importantly, these contaminants bio-accumulate through food chains, leading to sub-lethal toxicity or death of humans and other animals [1]. Although trace amounts of these substances are ubiquitous in the environment, anthropogenic activities may release additional, harmful quantities of materials to the environment through industrial activities, mining, waste disposal, and agricultural use of pesticides and herbicides [1].

Because most metals and metalloids cannot be degraded in natural environment, it is important to reduce the quantity of these contaminants to a safe level prior to water use or discharge. Common treatment technologies include chemical precipitation [2], ion exchange [3], reverse osmosis (RO) [4], liquid–liquid extraction [5], and sorption [6]. Sorption is an effective method to transfer metals and metalloids from an aqueous solution to a solid phase. There is a growing effort to develop inexpensive sorbents for removal of metals and metalloids from water and wastewater, such as red

mud and steel slag [1], natural and modified kaolinite and montmorillonite [7,8], agricultural waste material [9], chemically modified plant wastes [10], clino-pyrrhotite [11], lignite [12], aragonite shells [13], kaolinite [14], peat [15], natural zeolites [16], lignin [17,18], diatomite [19], and clay [20]. The removal of metals and metalloids by different types of low-cost sorbents is summarized in Table S1 in the Supporting Information. Red mud, activated slag, and iron slag exhibited remarkable capacities for sorption of copper (II) and lead (II). Dry aluminum-derived water treatment solids also achieved good removal of chromium (VI) and selenium (VI), but sorption of boron (III) was low.

One of the most important factors controlling the sorption of metals and metalloids on amphoteric surfaces is pH [21]. Sorption of cations on amphoteric surfaces increases with increasing pH, whereas sorption of anions is typically a mirror image of the characteristic cation sorption behavior and decreases with increasing pH. The affinity of cations for amphoteric surfaces is correlated with the first hydrolysis constant of the metal cation, whereas the affinity of anions is related to the basicity constant of the anion considered as a base; stronger bases tend to have higher affinity for amphoteric surfaces [21].

The essential constituents of these sorbents are amorphous and crystalline iron and aluminum oxides. Electrolyte ions can form surface complexes of different strength and type on hydrous iron,

* Corresponding author. Tel.: +1 575 646 5870.

E-mail address: pxu@nmsu.edu (P. Xu).

manganese, and aluminum oxide surfaces, including outer-sphere, ion pair complexes, as well as inner-sphere, covalent coordination complexes [22–27]. The high reactivity of hydrous metal oxides results from the fact that hydroxyl groups of the active surface sites form “bridges” for sorption of cation and anion metals and metalloids [27]. The OH–OH distances of the hydrous oxides match well with the geometry of the coordination polyhedral of metals and metalloids [27].

Drinking water treatment solids (DWTS) are produced during coagulation processes using iron or aluminum salts. It is reported that approximately 60% and 90% of total iron and aluminum, respectively, exist in the form of amorphous instead of crystalline oxides in DWTS [28]. These iron and aluminum precipitates can be active substances for sorption of cationic and anionic contaminants. Previous researches have shown that DWTS could remove phosphate [28,29], hydrogen sulfide [28], fluoride [30], and metals and metalloids, including arsenic, selenium, lead, chromium, and mercury from water [1,31–39]. DWTS has also been used for soil remediation to reduce the bioavailability and bioaccessibility of arsenic, barium, cadmium, chromium, copper, nickel, lead, and zinc in soil [31,33,34,37,38,40,41].

Although previous studies have demonstrated the potential use of DWTS to sorb metallic and metalloid contaminants from water, the majority of experiments were conducted in bench-scale, batch operations or using synthetic water (Table S1). More research is needed to understand sorption capacities and breakthrough volumes of DWTS under different operating conditions. Toxicity Characteristic Leaching Procedure (TCLP) tests indicate that DWTS is non-hazardous waste [42]; therefore, using DWTS as sorbent might be a suitable option to reduce disposal costs and an inexpensive method for removal of metals and metalloids from impaired waters. However, DWTS is laden with organic and inorganic substances sorbed or precipitated from surface water or groundwater. Therefore, desorption or leaching of these substances, even if non-toxic, may contaminate the treated water during reuse of DWTS. This concern can be alleviated if DWTS is used to treat another waste stream, such as desalination concentrate, when the production of high quality water is not critical.

In recent years, desalination technologies, such as RO, have been increasingly used to produce water from impaired streams such as brackish water, seawater, surface water, and wastewater [43–46]. While desalination processes generate high quality product water, dissolved salts and other contaminants are retained in the concentrate stream. Depending on water recovery, the concentration of dissolved solids in the retentate can be two to ten times higher than in the feed water. Therefore, naturally occurring minerals and anthropogenic contaminants such as boron, arsenic, chromium, selenium, lead, and copper, may be concentrated in the brine, exceed the discharge limits, and negatively impact the environment if they are not treated properly. Concentrate management and disposal is currently one of the most challenging issues associated with water desalination [47,48]. Conventional technologies for concentrate treatment are often energy intensive and cost-prohibitive, mainly because of high salinity and near saturation level of sparingly soluble salts.

Selective removal of metallic and metalloid contaminants from desalination concentrate using DWTS may offer a low-cost alternative to a major problem. Because high ionic strength and salt concentration could have a significant effect on sorption [24,25], the sorption capacity of DWTS for constituents found in desalination concentrate requires further investigation. Therefore, the objective of this study was to evaluate the sorption capacity of DWTS for various metal and metalloid contaminants, including boron (III), copper (II), chromium (VI), lead (II), and selenium (VI) from desalination concentrate in continuous-flow conditions. The impact of operating conditions on sorption, including pH, feed concentration,

hydraulic and mass loading rates, and temperature were investigated. Two different types of DWTS were studied to determine their sorption capacity.

2. Materials and methods

2.1. RO concentrate and spiked metals and metalloids

RO concentrate was collected from the Kay Bailey Hutchison Desalination Plant in El Paso, Texas, that treats brackish groundwater. The total dissolved solids (TDS) concentration of the RO concentrate was approximately $10 \pm 2.3 \text{ g L}^{-1}$ with electrical conductivity of $18 \pm 2.1 \text{ mS cm}^{-1}$. The major ions in the RO concentrate included Na^+ (2660 mg L^{-1}), Ca^{2+} (670 mg L^{-1}), Mg^{2+} (168 mg L^{-1}), K^+ (69 mg L^{-1}), Cl^- (4990 mg L^{-1}) and SO_4^{2-} (1272 mg L^{-1}) while minor elements included arsenic, boron, chromium, copper, iron, manganese, and selenium. The pH was 7.8 with alkalinity of 388 mg L^{-1} as CaCO_3 . The total organic carbon (TOC) concentration in the RO concentrate varied between 0.5 and 7.7 mg L^{-1} . The physicochemical properties of the RO concentrate and the DWTS characteristics are summarized in Table 1.

To investigate the selective removal of metals and metalloids from desalination concentrate, metal salts of boron (III), copper (II), chromium (VI), lead (II), and selenium (VI) were spiked into the RO concentrate simultaneously (Table 2). These contaminants were selected because of their frequent occurrence in natural and industrially contaminated water, and their potential adverse impact on agricultural irrigation and human health [49–51]. The spiking concentrations of the contaminants were 2–6 times higher than the Maximum Contaminant Levels (MCLs) of the United States National Primary Drinking Water Regulations [52], or determined based on the recommended maximum concentrations of contaminants in irrigation water [53]. The spiked metal salt concentrations are within the typical concentration range of real RO concentrates. The metal salts spiked in the RO concentrate were of ACS grade or higher from Fisher Scientific Co. (Fair Lawn, NJ), J. T. Baker Chemical, Co. (Phillipsburg, NJ), and Sigma–Aldrich (St. Louis, MO).

Electrical conductivity and pH of the water samples were measured using a conductivity and pH meter (Model 431-61, Cole-Parmer, Vernon Hills, IL). TOC was quantified using a carbon analyzer (Shimadzu TOC-L, Kyoto, Japan). Common anions including chloride, phosphate, and sulfate were measured using an ion chromatograph (IC, ICS-2100, Dionex, Sunnyvale, CA, USA), and the concentrations of aluminum, arsenic, boron, calcium, chromium, copper, iron, potassium, magnesium, manganese, sodium, lead, and selenium were quantified using an inductively coupled plasma mass spectrometry (ICP-MS, Elan DRC-e, PerkinElmer, Waltham, MA, USA). Alkalinity was measured using a digital titrator (Hach, Colorado, USA) and 1.6 N sulfuric acid standard solutions to an endpoint pH of 4.6. The TDS concentration was measured following evaporation method at 180°C after filtering the RO concentrate sample using a $0.45 \mu\text{m}$ cellulose acetate membrane filter (Toyo Roshi Kaisha, Ltd., Japan).

2.2. DWTS and characterization

Two types of dewatered DWTS were studied to compare their selective sorption capacities for metals and metalloids in RO concentrate (Table 1). One DWTS (referred to as SDWTS) was obtained from a conventional surface water treatment plant in Colorado, USA, which uses coagulation, flocculation, sedimentation, and greensand filtration to treat the water diverted from a creek to produce potable water. Ferric sulfate is the coagulant used to remove turbidity, natural organic matter, and manganese from the surface

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