



Engineering metal (hydr)oxide sorbents for removal of arsenate and similar weak-acid oxyanion contaminants: A critical review with emphasis on factors governing sorption processes

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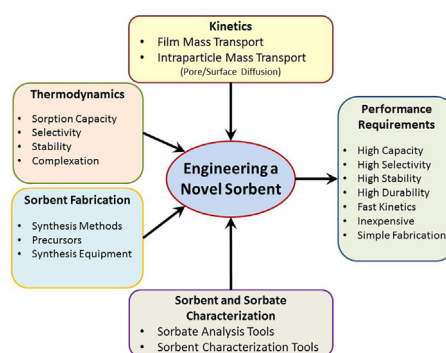
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HIGHLIGHTS

- Sorbents represent a platform for simple and inexpensive water treatment systems.
- Thermodynamics and kinetics mechanisms dictate sorbent performance.
- A conceptual framework and holistic approach in developing sorbents is paramount.
- Solution oriented platform for addressing real water problems is established.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 13 March 2017

Received in revised form 13 April 2017

Accepted 14 April 2017

Available online 23 April 2017

Editor: D. Barcelo

Keywords:

Arsenic
Oxyanions
Sorbent
Sorbate
Water treatment
Metal (hydr)oxide nanomaterial

ABSTRACT

To create an integrative foundation for engineering of the next generation inexpensive sorbent systems, this critical review addresses the existing knowledge gap in factor/performance relationships between weak-acid oxyanion contaminants and metal (hydr)oxide sorbents. In-depth understanding of fundamental thermodynamics and kinetics mechanisms, material fabrication, and analytical and characterization techniques, is necessary to engineer sorbent that exhibit high capacity, selectivity, stability, durability and mass transport of contaminants under a wide range of operating and water matrix conditions requirements. From the perspective of thermodynamics and kinetics, this critical review examines the factors affecting sorbent performances and analyzes the existing research to elucidate future directions aimed at developing novel sorbents for removal of weak-acid oxyanion contaminants from water. Only sorbents that allow construction of simple and inexpensive water treatment systems adapted to overcome fiscal and technological barriers burdening small communities could pave the road for providing inexpensive potable water to millions of people. Novel sorbents, which exhibit (1) poor performances in realistic operating and water matrix conditions and/or (2) do not comply with the purely driven economics factors of production scalability or cost expectations, are predestined to never be commercialized.

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1. Introduction

Anthropogenic pollution and extensive overuse of water for domestic, agricultural, and industrial applications threaten to deplete the

existing fresh water resources in the near future (Bouwer, 2000; Helweg, 2000; Hoekstra and Mekonnen, 2011). Providing access to potable water, which already represents a major problem for many communities in the developing world, is projected to transcend the national geo-economic boundaries and become an imperative global issue (Bogardi et al., 2012; Bundschuh et al., 2010; Cohen et al., 2004; Mutikanga et al., 2009; Ravenscroft et al., 2009; van der Bruggen and

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Borghgraef, 2010; Wescoat et al., 2007). Existing water treatment technologies, which are typically applicable to large communities, become economically infeasible when scaled-down to serve the needs of smaller communities (Berg, 2015; Litter et al., 2012; Wilcox et al., 2010). Small communities typically rely on groundwater as their potable water source and often lack financial, infrastructural, or technical resources to operate complex water treatment systems capable of simultaneously removing multiple contaminants (Brennan and McBean, 2011; Dawoud and Raouf, 2009; Foster and Chilton, 2003; Wilcox et al., 2010). These challenges are especially cumbersome in environments where geological formations promote dissolution of weak-acid oxyanion species, like arsenic, vanadium, or antimony, which cause chronic toxicity and require innovative treatment approaches (Filella et al., 2002; Gao et al., 2011; Goh and Lim, 2004; Gorny et al., 2015; Smedley and Kinniburgh, 2002; Szrameka et al., 2004). Development of simple and inexpensive groundwater treatment systems, which are (1) tailored to simultaneously remove multiple oxyanionic contaminants with specific chemistries and (2) adapted to overcome fiscal and technological barriers burdening small communities, could pave the road for providing inexpensive potable water to millions of people.

Sorbents represent an ideal platform for engineering simple and inexpensive systems capable of simultaneously removing multiple harmful weak-acid oxyanions from complex water matrices. In the last two decades, there has been a rapid expansion in development of novel sorbents for treatment of weak-acid oxyanions (Bang et al., 2011; Ippolito et al., 2011; Mohan and Pittman, 2007; Möller, 2011; Westerhoff et al., 2006). However, the majority of this work has been based on the trial and error approach, rather than on a fundamental understanding of the physico-chemical principles that govern the performances of a sorbent in a given groundwater matrix (Bleiman and Mishael, 2010; Camacho et al., 2011; Diamadopoulos et al., 1993; Duc et al., 2006; Gibbons and Gagnon, 2011; Kamei-Ishikawa et al., 2008; Maeda et al., 1990; Monteil-Rivera et al., 2000; Ramana and Sengupta, 1992; Park and Kim, 2011; Pillewan et al., 2011; Pirišä et al., 2011; Prathap and Namasivayam, 2010; Saikia et al., 2011; Sari et al., 2012; Suzuki et al., 1997; Li et al., 2011; Ma and Tu, 2011; Mariussen et al., 2012; Mohan and Pittman, 2007; Mostafa et al., 2011; Mou et al., 2011; Rovira et al., 2008; Tanboonchuy et al., 2011; Watkins et al., 2006; Wei et al., 2012; Yang et al., 2011; Zhang et al., 2009a, 2009b; Zheng et al., 2011). A comprehensive understanding of the sorbent/sorbate interactions in complex water matrices represents a cornerstone in developing the next generation of sorbents, which should exhibit high selectivity and capacity for removing toxic weak-acid oxyanions from water. Only by bridging this fundamental knowledge with existing macroscopic engineering concepts, successful development of the next generation sorbent based systems could be accomplished.

Transition metal (hydr)oxides, such as ferric (hydr)oxides or titanium dioxide, have been widely used for removal of oxyanion contaminants from water (e.g. arsenic, antimony, selenium, vanadium) (Bortun et al., 2010a; Hodi et al., 1995; Hristovski et al., 2007; Jordan et al., 2013a; Naeem et al., 2007; Nguyen et al., 2011; Peraniemi et al., 1994; Pierce and Moore, 1980, 1982; Westerhoff et al., 2006; Xu et al., 2012). Although many of these studies provide a partial description of how sorption of weak-acid oxyanions on metal (hydr)oxide surfaces is influenced by the fundamental factors, they do not address the theoretical and practical aspects of the problem in context of developing a solution oriented platform that could serve as a tool for addressing real world problems. In-depth understanding of these factors, organized in a conceptual framework that contextualizes their practical applicability, provides a foundation for engineering the next generation of high-performance sorbents for removal of toxic weak-acid oxyanion contaminants from complex water matrices.

In order to create an integrative foundation for engineering of the next generation inexpensive sorbent systems, this critical review addresses the existing knowledge gap in factor/performance relationships between weak-acid oxyanion contaminants and metal (hydr)oxide

sorbents. To accomplish the goal of this study: (1) sorbent performance requirements are summarized; (2) determinants engaged in the process of sorbent-engineering are characterized; (3) main factors controlling these determinants are systematically examined.

2. Conceptual framework for engineering the next generation of sorbents

Conceptual frameworks often represent a beneficial tool that helps contextualize the unique challenges emerging from development and implementation of novel technologies. A conceptual framework that describes the determinants engaged in the process of engineering novel sorbents is illustrated in Fig. 1.

Engineering the next generation sorbents is primarily directed by the relatively invariable performance requirements. In brief, the sorbents must (1) exhibit high selectivity and capacity for specific contaminants; (2) allow for fast mass transport of contaminants; (3) minimize operational and maintenance requirements when operated in full-scale applications; and (4) be inexpensive and simple to fabricate when compared to other water treatment technologies. Successful fulfillment of these requirements necessitates a solid understanding of thermodynamics, kinetics, material fabrication, and analytical and characterization techniques. Thermodynamics actuates the sorption capacity, selectivity, and stability of a sorbent. Change in thermodynamic conditions, such as change in sorbate concentration or chemistry, could significantly affect these key sorbent properties. Other sorbent properties, such as particle size and porosity, significantly influence kinetic parameters, like intra-particle and external (film) mass transport processes, that affect the design of a sorbent system. A comprehensive understanding of material fabrication and characterization is imperative for tailoring sorbent characteristics to fit desired performances for specific water matrices. Considering the interdependence of these factors, a holistic approach in developing the next generation sorbents becomes paramount. This holistic approach, however, mandates an in-depth understanding of the relationships among these factors, their prevalence, and the challenges stemming from them.

3. Factors governing the sorption thermodynamics

3.1. The chemistry of weak-acid oxyanion sorption onto metal (hydr)oxides

Empty d orbitals in a transitional metal act as Lewis acids for the oxyanions' electron rich oxygen atoms (Kutzelnigg, 1984; Sherman, 1985; Stair, 1982, 1991). Consequently, weak-acid oxyanion contaminants sorb onto metal (hydr)oxide surfaces by creating oxygen bridges via ligand exchange with OH⁻ or OH₂ groups located on the sorbents' surface and form stable monodentate or bidentate inner-sphere complexes, although recent evidence suggests that tridentate complexes could also be formed under specific conditions (Fig. 2) (Breynaert et al., 2008; Goldberg and Johnston, 2001; Grossl et al., 1997; Ippolito et al., 2009; Jin et al., 2007; Lange et al., 1991; Manning et al., 2002; Mitsunobu et al., 2009, 2010; Otte et al., 2013; Peak, 2006; Rakshit et al., 2011; Wang et al., 2011; Waychunas et al., 1993, 1995a).

The thermodynamic stability of formed inner-sphere complexes is strongly dependent on the number of formed oxygen bridges, the orientation and proximity of the populated Lewis acid sites, and the relative surface loading (Fendorf et al., 1997). For example, the modeled Gibbs free energy change of reaction between AsO₄ tetrahedra and FeO₆ octahedra amounts <- 58 kJ/mol for monodentate and almost doubles to <- 100 kJ/mol for bidentate surface complexes with a net charge of zero (Farrell and Chaudhary, 2013). Bidentate corner-sharing complexes (Fig. 3(a)) between ferric (hydr)oxides surfaces and weak-acid oxyanions, like vanadate and arsenate, are considered more favorable and thermodynamically more stable than a bidentate edge-sharing complexes (Fig. 3(b)) (Majzlan, 2010; Peacock and Sherman, 2004; Sherman and Randall, 2003).

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