



Arsenate adsorption onto iron oxide amended rice husk char



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HIGHLIGHTS

- Produced rice husk char (RHC) and measured its specific surface area (SSA).
- Amended RHC with iron oxide (IO) and measured As(V) adsorption isotherms.
- Compared amended RHC with IO-amended sand and IO-based material.
- Evaluated SSA of RHC as function of charring temperature.
- Evaluated As(V) adsorption on IO-amended RHC with higher SSA than above RHC medium.

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ABSTRACT

In this study, rice husks were charred at 550 °C in a partially sealed ceramic vessel for 30 minutes to create a high specific surface area (SSA) rice husk char (RHC). The RHC was then amended with iron oxides using dissolved ferric nitrate, $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, to provide a surface chemistry conducive to arsenic adsorption. The 550 °C iron oxide amended rice husk char's (550 IOA-RHC's) SSA was nearly 2.5 orders of magnitude higher and the arsenate adsorptive level was nearly 2 orders of magnitude higher than those reported for iron oxide amended sand, thus indicating a positive relationship between post-amendment SSA and arsenate adsorptive levels.

Rice husks were then charred at temperatures ranging from 450 °C to 1050 °C to create an even higher SSA material, which might further increase arsenate adsorptive levels. The 950 °C RHC was chosen for amendment due to its high SSA and feasibility of being produced in the field. Once amended, the 950 °C iron oxide amended rice husk char (950 IOA-RHC) improved the arsenate adsorption capacity by thus confirming a positive relationship, though not a linear relationship, between post-amendment SSA and arsenic adsorptive capacity. Further study demonstrated that post-amendment mesoporous volume and mesoporous surface area appear to be better indicators of arsenic adsorptive capacity than SSA or iron content.

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1. Introduction

1.1. Background

The World Health Organization (WHO) has recommended an allowable drinking water arsenic concentration of 10 µg/L (World Health Organization, 2011). Symptoms of arsenicosis, the chronic results of consuming excessive levels of arsenic, include skin lesions, black foot disease, diabetes, and nervous, hepatic, haematological, and renal damage (Hughes, 2002). Numerous regions throughout the world have groundwater with naturally occurring arsenic concentrations

exceeding the WHO's recommended allowance, with more than 140 million people worldwide exposed to water exceeding the WHO's allowable concentration (Ravenscroft et al., 2009). For countries with excessive arsenic concentrations and limited financial resources, an interim concentration of 50 µg/L is often adopted (World Health Organization, 2001). Still more than 50 million people are presently or have recently been drinking water with arsenic concentrations exceeding this interim allowance (Ravenscroft et al., 2009).

Current arsenic treatment approaches include reverse osmosis, chemical coagulation followed by filtration, and adsorptive media (Crittenden et al., 2005). The advantages of arsenic adsorptive media include potentially lower energy requirements, chemical requirements, capital requirements (for smaller systems), and environmental impacts than other methods. Common arsenic adsorptive media include activated alumina and iron-oxide-based media. As an example of deploying adsorptive systems in developing countries, SenGupta's group has over a decade of experience in implementing activated alumina based systems in West Bengal (Cumbal and SenGupta, 2005; Sarkar et al., 2005, 2010).

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While several options exist for arsenic remediation, many are cost-prohibitive for those living in developing communities (in terms of capital costs, operations, and/or maintenance costs). Low-cost adsorptive media are thus needed to achieve sustainable solutions in these low-income settings. Low-cost adsorptive media would provide the potential for lower energy and water requirements than reverse osmosis, allow a more decentralized infrastructure relative to chemical coagulation and filtration, and produce a lower maintenance (media replacement) cost than many existing arsenic adsorptive media. Furthermore, locally and/or domestically produced low-cost adsorbents have the potential advantage of promoting local businesses in developing countries.

1.2. Literature review

As(III) and As(V) are the two most common valences of arsenic found in potable groundwater. As(V) (arsenate) predominately exists in the form of H_2AsO_4^- or HAsO_4^{2-} in arsenic contaminated groundwater with oxidizing conditions and potable pH values (Wang and Mulligan, 2006). As(III) (arsenite), which is found in arsenic contaminated groundwater with reducing conditions, is more toxic than As(V) (Hughes, 2002) and predominately exists as H_3AsO_3 (uncharged) for potable pH values less than ~ 9.2 (Wang and Mulligan, 2006). Unfortunately, As(III) can be harder to remove, although certain systems have proven effective at doing so (e.g., Berg et al., 2006). For many other systems, the lower As(III) adsorption affinity versus As(V) can be attributed to As(III)'s uncharged nature (Kartinen and Martin, 1995). Where elevated As(III) concentrations exist, removal by adsorption can be improved by a pre-oxidation step to convert As(III) to As(V) (Crittenden et al., 2005). Conversely, when As(III) exists and a pre-oxidation step is not employed, there is some suggestion in the literature that the iron oxide surfaces themselves can oxidize As(III) to As(V) (Devitre et al., 1991; Sun and Doner, 1998), thereby improving the overall adsorptive removal efficiency but likely rendering the initial iron oxide zone an oxidation region rather than an adsorptive zone.

Many commercial and low-cost arsenic adsorbents have been reviewed elsewhere (Mohan and Pittman, 2007; Bhatnagar and Sillanpaa, 2010). Iron-based adsorptive media, for example, are common commercial adsorbents used in arsenic mitigation and have been thoroughly studied for their arsenic adsorptive properties. Granular ferric hydroxide (GFH), an amorphous β -FeOOH, has proven capable of adsorbing significant quantities of dissolved arsenate and arsenite, but has shown limitations under higher loading rates (pressurized systems) due to compaction (Driehaus et al., 1998; Badruzzaman et al., 2004). Bayoxide E33, another iron-based adsorbent comprised of 60% α -FeOOH and Fe_2O_3 (Lin et al., 2006), has also been chosen for several EPA arsenic mitigation demonstration sites due to its ability to adsorb arsenic (Severn Trent Services, 2004).

Due to the potential price limitations of commercially available iron based adsorptive media in developing region contexts, others have considered locally available materials, such as iron oxide amended sand (Scheidegger et al., 1993; Mlilo et al., 2010). Scheidegger et al. (1993) and Mlilo et al. (2010) evaluated iron oxide amended sand, but found the adsorptive levels to be inadequate for domestic arsenic drinking water treatment. However, Mlilo et al. (2010) found that, when normalized for specific surface area, the arsenic adsorptive levels of iron oxide amended sands were comparable to iron-matrix-based media on a per unit surface area basis, demonstrating that while the iron oxide amended sand has favorable surface chemistry, the iron oxide amended sand suffers from inadequate surface area. This would lead one to consider other locally available, high surface area materials, such as biochars, that could be amended with iron oxides. Our approach of amending locally available materials is consistent with the work of others (Petruševski et al., 2002, 2007, 2008).

Several researchers have amended non-iron-oxide-based media with arsenic adsorbent materials, such as oxyhydroxides and

titanium di-oxide, as alternatives to iron-oxide-matrix-based media. Researchers have begun evaluating the arsenic adsorbent properties of TiO_2 -impregnated chitosan beads with promising results (Miller and Zimmerman, 2010). Furthermore, amending commercially available granular activated carbon (GAC) with iron has garnered considerable experimental investigation for arsenic removal (Gu et al., 2005; Chen et al., 2007; Jang et al., 2008; Chang et al., 2010) and for other uses, such as Fenton-driven methyl tert-butanol ethylene granulated activated carbon regeneration (Huling et al., 2007, 2009; Kan and Huling, 2009; Huling and Hwang, 2010). While this work supports the concept of amending higher surface area materials with iron oxides, at present many arsenic afflicted countries may not be able to access or afford commercially manufactured GAC.

There are several oxyanions common in ground water that will compete with arsenic for adsorption sites on an iron oxide surface. To examine the versatility of the iron oxide amended rice husk char (IOA-RHC) it is very important to consider competitive ions, which include sulfate, phosphate, silicate, bicarbonate, chloride, and humic acid (Meng et al., 2002; Hsu et al., 2008). Personal communication with the non-governmental organization Resource Development International in Cambodia (RDIC) has emphasized the competitive ions: phosphate, silicate, and sulfate.

1.3. Goals, hypothesis, and objectives

Given the surface area limitations of iron oxide amended sand and the potential limited availability and/or cost prohibitive nature of commercially-available media substrates at the community level, this research focused on charring locally available biomaterials, such as rice husks, for iron oxide amendment. Rice husks are ubiquitous by-products throughout much of the developing world including countries most afflicted with arsenic contamination (e.g. Bangladesh, India, and Cambodia). Studies have shown that rice husks can be carbonized and activated to $290 \text{ m}^2/\text{g}$ using physical (thermal) carbonization and activation (Deiana et al., 2008), a specific surface area approaching that of commercially-available GAC (see Gu et al., 2005). Thus, this study combines locally-producible rice husk char with iron oxide amendment and evaluates the arsenate adsorption potential of the resulting materials.

The goal of this research was to develop an arsenic adsorptive media based on iron oxide amendment of a widely available biomaterial in much of the developing world to achieve arsenic adsorptive capacities higher than iron oxide amended sand and approaching that of commercially available media (e.g. GFH and Bayoxide E33). This research builds on two hypotheses.

1. Iron oxide amended rice husk char will increase the arsenate adsorptive levels relative to iron oxide amended sand by combining appropriate surface chemistry with an elevated surface area medium for efficient arsenic removal.
2. Increasing the surface area by adjusting the charring temperature prior to iron oxide amendment will increase the arsenate adsorptive capacity of the resulting media.

Pursuing the goal and hypotheses of this research was accomplished in two steps:

1. Produced rice husk char (RHC), measured its specific surface area (SSA) before and after amending it with iron oxide, measured arsenate adsorption isotherms, and compared results to iron oxide amended sand.
2. Evaluated the SSA of RHC as a function of charring temperature, and evaluated arsenate adsorption onto an iron oxide amended rice husk char with a higher SSA than the above RHC medium.

From an implementation perspective, the effect of competing oxyanions is important to evaluate. A secondary objective of this

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