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# A novel MD-ZVI integrated approach for high arsenic groundwater decontamination and effluent immobilization

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## ABSTRACT

A highly effective novel system of direct contact membrane distillation (DCMD) integrated with acid-purged zero-valent iron (APZ) technology has been developed. Compared to conventional processes of arsenic removal which reconstitute secondary contamination at disposal site, this system proves capable of simultaneous removal and immobilization of arsenic from contaminated water with great efficiency and improved water flux. Using composite microporous membranes of polytetrafluoroethylene (PTFE) and polypropylene (PP) integrated with APZ in a DCMD-APZ configuration, varying residual arsenic concentrations were injected anoxically into  $\leq 2$  g acid-washed Fuchen (XK 13-201) reduced Fe powder at a flow rate of 0.33 mL/min and pH  $6 \pm 1.0$  at 60 °C. Results from this unique configuration show advantages including maximum distillate flux production of 55.5 kg/m<sup>2</sup> h with greater than 95% arsenic rejection efficiency using PTFE/PP composite membrane, fast adsorption and immobilization of rejected arsenic on APZ at  $t_{1/2} \leq 30$  min, and electrochemical reduction of As(V) and/or As(III) to sparsely soluble As(0) as confirmed by macroscopic wet chemistry, adsorption kinetic model and X-ray photoelectron spectroscopic (XPS) analysis conducted within 5 days of experimental period. Since the arsenic adsorption/reduction process is a thermodynamically assisted phenomenon, integrated configuration of the DCMD-APZ technology stands out as a promising technique to mitigate the unresolved challenges of arsenic contamination and re-dissolution in groundwater.

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## 1. Introduction

Arsenic (As), a well-known carcinogen found in groundwater or localized environments at high concentrations due to natural geo-accumulation processes or as a result of specific release from metal smelting operations, chemical spills and microbial mediated processes, has generated severe health concerns around the globe. As connoted from the Greek word

arsenikon, arsenic has been attributed to being the “king of poison” (Vahidnia et al., 2007; Wang et al., 2012). Therefore, long-term exposure to high levels of As in drinking water has been associated with various skin lesions, cardiovascular diseases, diabetes, neuropathies, as well as cancers of several organs (Mazumder et al., 1998; Balakumar and Kaur, 2009; Navas-Acien et al., 2008; Mukherjee et al., 2003). Arsenic contamination in drinking water continues to stir concern

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internationally, due to its widespread distribution. This problem is particularly prevalent in Datong Basin, China where new endemic areas are constantly emerging.

Thus, it is highly desirable to develop approaches that can significantly improve the remediation of this class of contaminants in the environment; as such, integrated use of membrane purification technologies hold promises (Yang et al., 2014). Polymer and inorganic membranes have been essentially utilized for various filtration processes including chlorinated compounds and heavy metals removal, such as arsenic (Wang and Chung, 2013; Yarlagadda et al., 2011). Studies on As removal using membrane distillation (MD), a thermally driven process involving movement of vapor across a microporous hydrophobic membrane, have been well researched (Pal and Manna, 2010; Yarlagadda et al., 2011; Jadhav et al., 2015). Among the different kinds of MD process, direct contact membrane distillation (DCMD) is the most efficient and easy to utilize MD technique. Advances in this field include fabrication of hybrid membrane with higher rate of contaminants rejection, longevity and less fouling capabilities (Wang and Chung, 2015; Wang et al., 2015). Yet, there is a huge knowledge gap between the emergence and application of membrane distillation technique for arsenic groundwater remediation and lack of an adequate understanding of the process enhancement still exists.

Furthermore, recent research on high performance membranes based on carbon nanotubes reveals a distinctive 3D nanostructure that efficiently adsorbs arsenic from contaminated groundwater due to protected agglomeration, increased surface-to-volume ratio, and water pathway of 3D nanostructures (Vadahanambi et al., 2013). However, the commercial viability of this new class of thin-film composite membranes for water re-use hinges on the development of inexpensive coatings, chemistries and scalable processing methods that can reproducibly achieve the desired membrane structure and yield reasonable fluxes (Shannon et al., 2008). While these advances are plausible, treatment of residual arsenic currently lags behind in all available membrane applications for arsenic containment.

Increasing research efforts have focused on using various adsorbents such as Fe binary oxides (Kong et al., 2014) and zero valent iron (ZVI) to remove As (Farrell et al., 2001; Kanel et al., 2005; Ramos et al., 2009; Li et al., 2015; Baikousi et al., 2015), and have demonstrated that ZVI is an effective sorbent which may be used to immobilize As from groundwater via surface adsorption/complexation, reduction, surface precipitation or co-precipitation (Mak et al., 2009; Noubactep, 2008). However, there are limitations in the use of ZVI to remediate As in water. These include competition from other oxyanions such as phosphate and silicate for sorption sites at corroded iron surfaces and possible later release of sequestered As due to subsequent mineralogical transformation of initially formed corrosion products (Su and Puls, 2004). Controversies about the mechanisms for removal of arsenic especially regarding redox reactions are an ongoing issue. The rate and extent of As(V) reduction by ZVI may depend on the prevailing experimental conditions. For instance, some authors have argued that the contaminants enmeshed in the matrix of iron corrosion products are stable for long time under environmental conditions whether they are chemically transformed or not (Noubactep, 2006; Lackovic et al., 2000). The current concept on the microbial reduction of ferrihydrite or iron corrosion products in situ/ex situ, resulting in the release of adsorbed species (e.g. arsenic) raises questions about the immobilization status

of adsorbed contaminants (Yadav et al., 2015; Huang et al., 2015).

Considering the foregoing, treatment of arsenic contaminated water by employing the efficiency of membrane filtration processes coupled with residual arsenic containment technologies could be a hot direction for future research. In view of this, the objectives of this study were to (1) introduce a highly effective novel system of direct contact membrane distillation (DCMD) integrated with acid-purged zero-valent iron (APZ) technology, (2) evaluate the different mechanisms essential for optimization of the DCMD process in the context of high arsenic groundwater, and (3) investigate the efficiency of DCMD-APZ technique for simultaneous high feed-As rejection and residual arsenic containment respectively. Most studies have utilized ZVI and modified reduced iron for adsorptive removal of arsenic under aerobic and anoxic conditions but little is known about the mechanism of arsenic removal in pump-and-treat method under simulated rigorous anoxic conditions. To the best of our knowledge, no study has reported the containment of residual arsenic generated from membrane treatment processes which are potential sources of secondary pollution. Hence, this study sheds light on the advantage the DCMD-APZ technology offers for efficient arsenic contaminant removal and immobilization.

## 2. Experimental

### 2.1. Materials and methods

Acrylonitrile butadiene styrene (ABS), a terpolymer of acrylonitrile, butadiene and styrene, was purchased from Saipwell (Shanghai, China) and used for the design of a lab-made module with dimension 28 cm × 19.5 cm × 6.5 cm, in the DCMD set-up. We obtained three hydrophobic flat-sheet membranes from Membrane Solutions LLC (Shanghai, China) and they were put to use without further treatment. Properties of each membrane are summarized in Table 1. Properly preserved groundwater samples obtained from Datong Basin in Shanxi Province, China (groundwater characteristics are presented in Table 2) and arsenic simulated solutions prepared using analytical reagent grade of Na<sub>2</sub>AsO<sub>4</sub>·7H<sub>2</sub>O (Sinopharm Chemical Reagent Co. Ltd., China) for As(V) and NaAsO<sub>2</sub> (Sigma-Aldrich) for As(III) were used in the experiments. All chemical reagents utilized including HCl, NaOH, and NaCl are of analytical reagent grade. Deoxygenated ultra-pure water purge with high purity nitrogen gas at 70 °C for 4 h was employed in the APZ reaction process. This was done to simulate rigorous anoxic conditions found in groundwater environments and to eliminate dissolved O<sub>2</sub>, a potential oxidant in these solutions.

### 2.2. Acid-purged ZVI synthesis

Zero valent iron (XK 13-201) obtained from Tianjin Fuchen Reagent Co. Ltd., China, with bulk iron composition of Fe–98%,

**Table 1 – Membrane characteristics.**

Membrane type	Active layer	Substrate layer	Nominal pore size (μm)	Porosity (%)	Thickness (μm)
PTFE0221	PTFE	PP	0.22	82	170
PTFE0221B	PTFE	PET	0.22	80	160
MSPP270045	PP	PET	0.45	75	160

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