



A nanofiltration–coagulation integrated system for separation and stabilization of arsenic from groundwater



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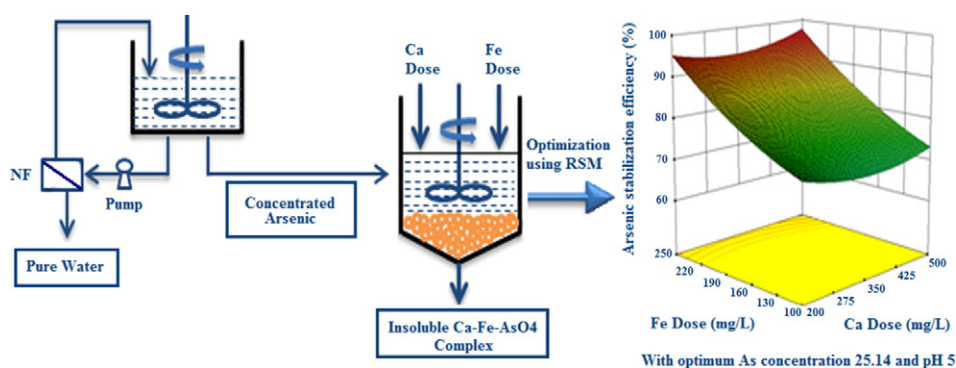
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HIGHLIGHTS

- A total solution scheme devised and tested for arsenic removal and stabilization.
- Stabilization of arsenic rejects done under response surface optimized conditions.
- Statistical model of RSM successfully predicts the system performance ($R^2=0.985$).
- The system is characterized by high flux, high rejection and low price.

GRAPHICAL ABSTRACT



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ABSTRACT

A membrane-integrated hybrid treatment system has been developed for continuous removal of arsenic from contaminated groundwater with simultaneous stabilization of arsenic rejects for safe disposal. Both trivalent and pentavalent arsenic could be removed by cross flow nanofiltration following a chemical pre-oxidation step for conversion of trivalent arsenic into pentavalent form. The very choice of the membrane module and its judicious integration with upstream oxidation and downstream stabilization resulted in continuous removal of more than 98% arsenic from water that contained around 190 mg L^{-1} of total suspended solid, 205 mg L^{-1} of total dissolved solid, 0.18 mg L^{-1} of arsenic and 4.8 mg L^{-1} of iron at a pH of 7.2. The used flat sheet cross flow membrane module yielded a high flux of $144\text{--}145 \text{ L m}^{-2} \text{ h}^{-1}$ at a transmembrane pressure of only $16 \text{ kgf} \cdot \text{cm}^{-2}$ without the need for frequent replacement of the membranes. Transmembrane pressure, cross flow rate through the membrane module and oxidant dose were found to have pronounced effects on arsenic rejection and pure water flux. For the first time, an effective scheme for protection of the total environment has been ensured in this context where arsenic separated with high degree of efficiency has been stabilized in a solid matrix of iron and calcium under response surface optimized conditions. The study culminated in a total and sustainable solution to the problem of arsenic contamination of groundwater by offering arsenic-free water at a reasonably low price of only $1.41 \text{ \$} \cdot \text{m}^{-3}$.

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1. Introduction

Contamination of groundwater by leached out arsenic is a worldwide problem. Millions of people of several countries around the world are at a risk of this arsenic poisoning (Yuan et al., 2003). Along with quite a few South-East Asian countries, the Bengal-Delta basins

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in India and Bangladesh have been the prime arsenic-prone regions in the world (Bhattacharyya et al., 2003; Wickramasinghe et al., 2004; Violante et al., 2006; Pal et al., 2009). With lowering of maximum permissible limit of arsenic in drinking water to $10 \mu\text{g}\cdot\text{L}^{-1}$ by WHO (WHO, 1993), treatment of arsenic contaminated water became further challenging as underground aquifers in many regions produced water with levels of arsenic concentration as high as $3000 \mu\text{g}\cdot\text{L}^{-1}$.

A variety of treatment options have been documented by the researchers world-wide for removal of arsenic from the contaminated groundwater and these include chemical coagulation–precipitation, adsorption, ion exchange, membrane filtration, membrane distillation, photo-catalytic oxidation and ultrafiltration (Lin and Wu, 2001; Zouboulis and Katsoyiannis, 2002; Gholami et al., 2006; Pal et al., 2007; Hsieh et al., 2008; Nguyen et al., 2008; Pal and Manna, 2010). From the quality point of view, adsorption-based process is not self-monitored as it continues to produce water even when the adsorption bed gets exhausted and the consumers often continue to collect water which is really not arsenic-free. Disposal of spent adsorbent bed is often a problem. Regeneration of spent adsorbent is energy-intensive, often incomplete and involves high cost. Membrane distillation produces water with the highest purity but suffers from the disadvantage of low volumetric flux. Physico-chemical treatment can produce treated water in large volumes but degree of purification cannot be expected to be high or even comparable to those obtained in membrane separation. Disposal of huge sludge is another problem. Membrane-based processes are known for high degree of separation and low flux over long time operation when fouling is significant. Widely studied modules like hollow fiber, spiral-wound, plate and frame and tubular types are largely fouling-prone necessitating frequent replacement of membranes. Among the membrane-based processes, nano-membrane based flat sheet cross flow filtration module stands out to be one of the most promising as it has the potential of ensuring high rejection of arsenic and high flux of pure water in a largely fouling-free membrane module at a reasonably low transmembrane pressure compared to that required in reverse osmosis (RO). The toxic compounds can be separated by nanofiltration membranes exploiting both steric (sieving) and Donnan (electrical) mechanisms depending on the characteristics of such compounds and the membranes involved.

Arsenic occurs in groundwater in two major inorganic forms namely, trivalent arsenite [As (III)] and pentavalent arsenate [As (V)]. Under normal pH conditions, trivalent arsenite behaves as a neutral molecule

which can be separated only by the steric effects of the nanofiltration membrane whereas pentavalent arsenate being negatively charged can be separated by both the Donnan and steric mechanisms. In a previous study (Sen et al., 2010), it has reported that soluble arsenic can be efficiently removed from groundwater by NF membrane with prior oxidation of trivalent arsenic to pentavalent state. However, the issue of disposal of arsenic rejects was not addressed. The continuous scheme of arsenic removal from groundwater and its subsequent stabilization is yet to be evolved. Due to limited options for disposal of highly concentrated arsenic rejects, many arsenic removal plants generally dump the rejects into the environment which leaves potential risk of recontamination of underground aquifers through natural percolation process. A possible solution to this disposal problem may be traced in stabilization of arsenic in some solid matrix. Coagulation and co-precipitation with other minerals eventually binding arsenic in an insoluble form with such co-precipitators can lead to a viable solution. Quite a few studies on such coagulation–precipitation involving salts of iron, aluminum and calcium have been reported in the literature (Camacho et al., 2000; Roberts et al., 2004; Song et al., 2006; Baskan and Pala, 2009, 2010; Sullivan and Tyrer, 2010; Wang et al., 2011; Randall, 2012). In such studies, pH, molar ratio and mineral combinations have been found to influence stabilization process but the stabilization parameters were never properly optimized eliminating the mutual interaction effects. It has been observed that ferric arsenate is not thermodynamically stable at higher ranges of pH (>8) as it formed a solid layer of ferric hydroxide on the ferric arsenate precipitate and thus reduced the precipitation efficiency (Robins, 1990). Bluteau et al. (2009) showed that ferric arsenate released arsenic at the rate of 5 mg L^{-1} at neutral or near neutral pH and concluded that it might not be appropriate for long-term storage. Attempts have been made (Leist et al., 2003; Mollah et al., 2004) to improve the effectiveness of stabilization of different arsenic compounds using some solidification binders like Portland cement. But the excess lime present in cement provides an alkaline surface on the cement matrix enhancing instability of the ferric arsenate and involving high cost of stabilization. The disposal volume also increases sharply inviting further problems in transportation. Thus the problem of instability of such solid precipitates still remains to be solved. Moreover, in removal of arsenic from contaminated water, a continuous treatment scheme with a sustainable solution to the problem of disposal is yet to emerge. The present study is thus an attempt to fill this technology gap. This novel approach

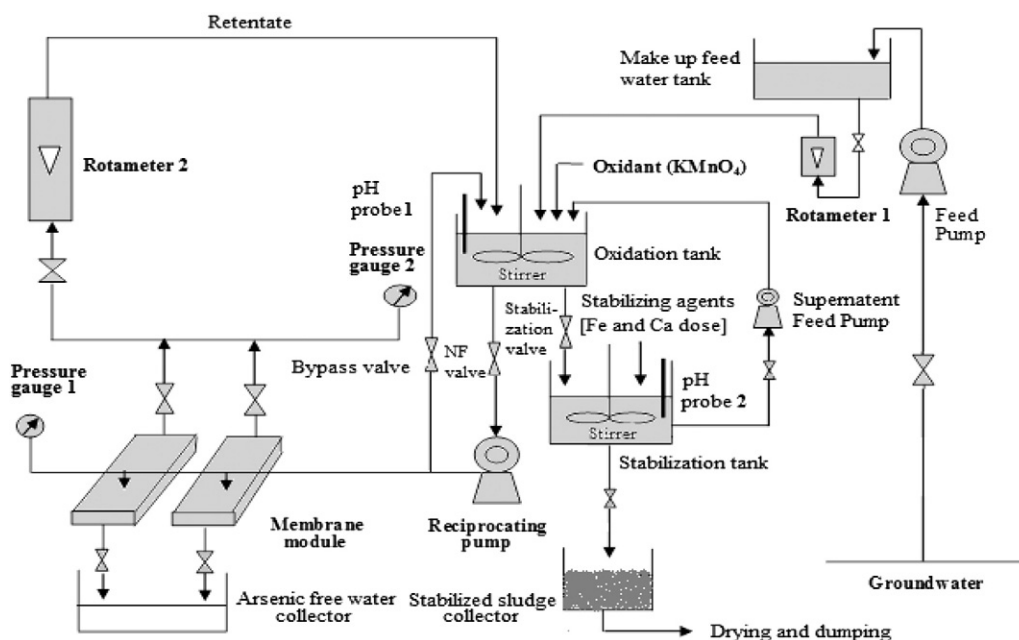


Fig. 1. Schematic diagram of the membrane-integrated hybrid process for removal of arsenic from water and its stabilization in solid matrix.

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