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Arsenic removal mediated by acidic pH neutralization and iron precipitation in microbial fuel cells

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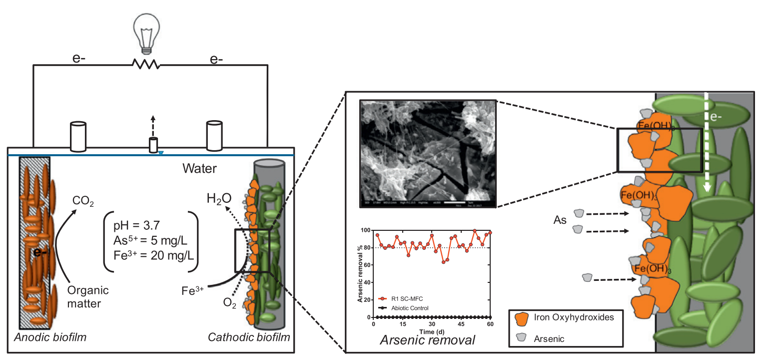
HIGHLIGHTS

- Arsenic and iron concentrations were removed in microbial fuel cell systems.
- Biofilms on electrodes were tolerant to low pH and high concentrations of metals.
- pH neutralization favored the formation of Fe minerals, and subsequent As removal.
- XRD analysis suggests the precipitation of Amorphous Fe minerals.

GRAPHICAL ABSTRACT



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ABSTRACT

High concentrations of arsenic (As) in natural waters are a growing concern worldwide. In northern Chile, fluvial systems enriched in As from natural and anthropogenic sources have been found to contain microbial communities with exoelectrogenic activity. Previous work performed with Microbial Fuel Cells (MFCs) resulted in a neutralizing microbial community developed from a consortium extracted from northern Chile. Considering that the formation of iron minerals, which have been reported as good As sorbents, would be favored by pH neutralization, the use of neutralizing MFCs could result in a sustainable alternative for Fe and As removal. In this work, we quantified the removal of As and Fe from acidic waters in air-cathode single-chamber MFCs. Our results show a removal ~80% of As and Fe and, simultaneously, a pH neutralization from ~3.7 to ~7.2. Additionally, non-MFC experiments indicate that the removal of As and Fe is dependent only on the activity of the microbial community developed during MFC operation and not on the MFC electrochemical performance. In addition, scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDS) analysis showed spatial associations between Fe and As on the surface of cathodes, suggesting the idea that iron oxyhydroxides formation would be associated with the higher oxygen concentration near the cathodes. Powder X-ray diffraction (XRD) analysis showed the dominance of iron amorphous minerals, which may be favoring the removal of As. These results indicate that acid/metal-tolerant bacteria favor pH neutralization and consequently the removal of Fe and As by processes of surface sorption and/or As-Fe co-precipitation. Furthermore, these findings expand the possible MFC applications to the simultaneous removal of Fe and As from acidic waters, enabling its use as an energetically sustainable bioremediation alternative.

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1. Introduction

Arsenic (As) in natural and drinking water is a major worldwide concern because of its great impact at very low concentrations (Kumar et al., 2004; Smedley and Kinniburgh, 2002). As is a carcinogen and has high toxic effects on human health (Cheng et al., 2011; Engel and Lamm, 2008; Farrell et al., 2001; Wu et al., 1989). For instance, in northern Chile, high concentrations of As profoundly affects the quality of water resources and creates serious health risks for the use and consumption of water (Caceres et al., 2005; Leiva et al., 2014; Pizarro et al., 2010). One possible origin of this problem is the release of As from minerals like arsenopyrite (FeAsS) and enargite (Cu_3AsS_4), which can be accelerated by natural conditions (e.g. pH, electrical conductivity, microbial activity) (Nordstrom et al., 2000; Pizarro et al., 2010; Smedley and Kinniburgh, 2002; Wang and Mulligan, 2006). Similarly, mining discharges have been shown to significantly increase the dissolved concentrations of As and iron (Fe) in rivers of northern Chile, such as the Elqui, Camarones, Loa, and Lluta rivers (Leiva et al., 2014; Oyarzun et al., 2004; Romero et al., 2003; Valenzuela et al., 2009).

In recent decades, the development of new methodologies to treat waters contaminated with As has received significant attention (Amin et al., 2006; Ballinas et al., 2004; Binbing et al., 2002; Borho and Wilderer, 1996; Johnston and Heijnen, 2001; Khan et al., 2002; Kumar et al., 2004; Leupin and Hug, 2005; Mohan and Pittman, 2007; Ning, 2002). Particularly, the use of Fe oxides to adsorb As has been studied as one of the inexpensive alternatives (Dixit and Hering, 2003; Mohan and Pittman, 2007). This is based on the association that has been observed between As and Fe minerals (e.g. oxyhydroxides) in natural environments (Belzile and Tessier, 1990; Smedley and Kinniburgh, 2002). Furthermore, it has been stated that the adsorption of As onto Fe minerals plays a key role in the control of As concentrations in the aqueous phase and in its speciation (Dixit and Hering, 2003; Vitre et al., 1991). Regarding sorption mechanisms, they are dependent on the oxidation state of As and also on the mineralogy of Fe oxyhydroxides (Bissen and Frimmel, 2003). Additionally, it has been observed that the formation of these minerals can be favored by neutralization of pH, which is commonly acidic for waters contaminated with As, Fe and other metals (Burrows et al., 2017; Seo et al., 2017).

Microbial Fuel Cell (MFC) is a promising technology for renewable energy generation (Chaudhuri and Lovley, 2003; Logan et al., 2006; Rabaey and Verstraete, 2005). This technology uses microorganisms, known as electrochemically active microorganisms (EAM), as catalysts to oxidize organic or inorganic electron donors and then, exogenously transfer electrons towards an electrode (anode), which is externally connected to another electrode (cathode), where the reduction of a final electron acceptor occurs (Chaudhuri and Lovley, 2003; Logan et al., 2006). In recent years, there have been advances in different aspects of MFC research, such as the reactor architecture (Roy et al., 2016) or electrode materials (ElMekawy et al., 2017; Aryal et al., 2017) which enabled the development of biotechnological applications for the removal

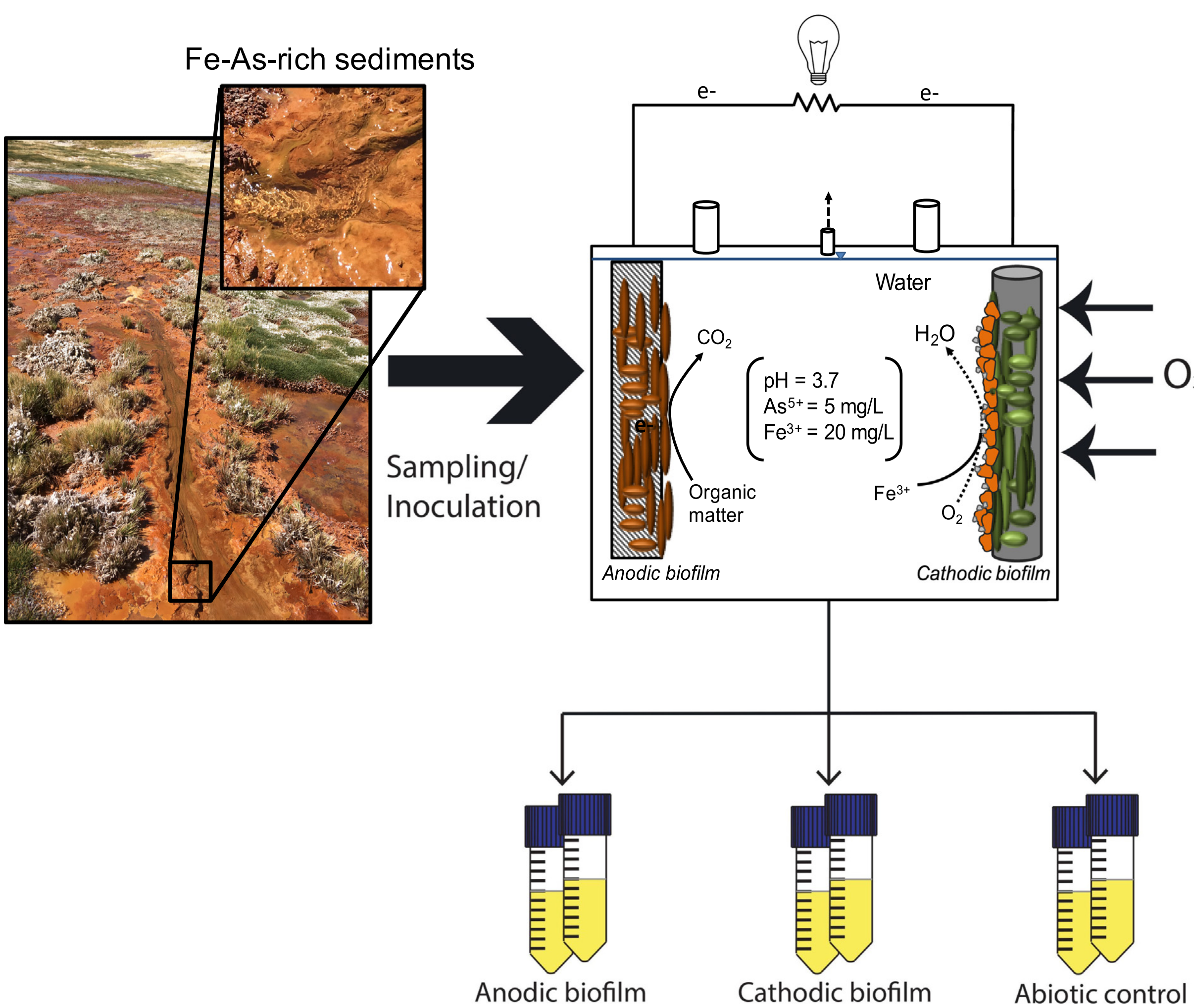

Fig. 1. Conceptual model and experimental design used in the study. Microorganisms extracted from Fe-As-rich surface sediments located in a hydrothermal area in northern Chile were enriched and used to inoculate SC-MFC reactors. As and Fe removal was evaluated in SC-MFC reactors and in batch reactors (non-MFC) inculcated with electrode biofilms (Anode and Cathode).