



Cathodic abatement of Cr(VI) in water by microbial reverse-electrodialysis cells



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ABSTRACT

For the first time a microbial reverse electrodialysis cell (MRC) was used for the treatment of water contaminated by Cr(VI). It has been recently shown that both inorganic and organic pollutants can be removed by reverse electrodialysis processes (RED) using water with different salinity without the supply of electric energy. However, a high number of membrane pairs is usually necessary for the treatment of wastewater by RED. Here, it was showed that a lower number of membranes can be used by the utilization of a MRC (i.e., a RED cell with a biotic anode) for such purposes. Indeed, the abatement of Cr(VI), chosen as model pollutant, was successfully achieved by cathodic reduction in short times using a drastically lower number of membrane pairs with respect to that required by a conventional RED. The effect of the number of membrane pairs and of the salinity gradient was studied in detail and the performances of MRC was systematically compared to that of RED.

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1. Introduction

Reverse electrodialysis (RED) is an innovative method to convert salinity gradient into useful power, based on the use of many pairs of anion and cation exchange membranes situated between two electrodes [1–10]. Salinity gradient energy can be obtained from seawater and freshwater sources, from salt ponds and seawater/river water or using thermolytic solutions [5] that can be concentrated with waste heat (>40 °C) energy [6]. The supply of solutions with different salt concentrations gives rise to an electromotive force E for a stack assembly of N membrane pairs [3,7] (see Eq. (1)), that can be used to produce electric energy by adopting suitable redox processes:

$$E = 2N\alpha RT \ln(a_c/a_d)/F \quad (1)$$

where R is the gas constant, T the absolute temperature, α is the average permselectivity of the membrane pair, F the Faraday constant and a_c and a_d the solute activities in concentrated and diluted solutions, respectively.

A proper selection of redox species and of electrode materials is necessary in order to develop the RED process on an applicative scale [4,8,9]. Redox processes based on the utilization of iron specie were proposed in order to reduce the potential penalty given by electrode processes [4,8,9]. Other authors recently studied the

possible utilization of capacitative electrodes for RED processes [10]. It has been recently shown by some of the authors that a proper selection of the redox processes can allow to use salinity gradients in a RED stack to achieve both energy generation and the effective treatment of wastewater contaminated by inorganic or organic compounds resistant to conventional biological processes, such as Cr(VI) or acid orange 7 [11–13]. However, many membrane pairs are needed for using salinity gradients for the production of electricity and/or the treatment of wastewater, resulting in significant costs for RED systems. In order to reduce the number of membrane pairs necessary for the cathodic abatement of Cr(VI), we have here evaluated the substitution of previously adopted anodic processes (water or chlorides oxidation), which require quite high anodic potentials (the oxidation of water to oxygen requires about 1.5 V at Ru anodes [14]), with the oxidation of waste organics by exoelectrogenic microorganisms at a biotic anode, that usually involves a working potential close to -0.2 V [15–18]. The RED stack equipped with biotic anode is called Microbial reverse electrodialysis cell (MRC) and it was proposed in literature to effectively capture energy from salinity gradients using the bacterial oxidation of organic matter and oxygen reduction as favorable electrode reactions [19]. MRC were also used for hydrogen production [20] and the production of chemicals for the carbon dioxide capture [21].

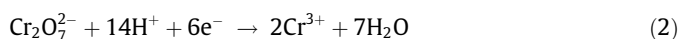
In this study, we examined the utilization of a MRC for the cathodic reduction of Cr(VI) to Cr(III) with the aim to achieve a fast abatement of the pollutant coupled with the utilization of a small

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number of membrane pairs (3–7, Fig. 1). Cr(VI) was chosen as a model pollutant for several reasons:

- Chromium compounds are widely used in metal plating, paints and pigments, leather tanning, textile dyeing, printing inks and wood preservation [22,23] and, as a consequence, relevant amounts of waste waters containing chromium, as Cr(VI) and Cr(III), are discharged by industries and have to be properly treated [24].
- Trivalent chromium is relatively harmless, whereas hexavalent chromium is about 100–1000 times more toxic [24]. Indeed, the concentration of Cr(VI) is limited in groundwater by a World Health Organization provisional guideline value of 0.05 mg dm^{-3} [25].
- The cathodic reduction of Cr(VI) into Cr(III) (Eq. (2)) on carbon electrodes (such as carbon felt or reticulated vitreous carbon) is an effective approach widely studied in literature [26–31] but requires the supply of electric energy, thus hindering up to now a wide application of such methodology.



- Both microbial fuel cell (MFC) [15,32–36] and RED were applied for the treatment of wastewater contaminated by Cr(VI) but MFC gives rise to very long treatment times [33] while RED, as above mentioned, can require a significant number of cell pairs [11,12]. As an example, the reaction time required for the complete removal of 100 mg dm^{-3} of Cr(VI) by MFC using graphite electrodes (11.25 cm^2) was 150 h at pH 2 [33]. Some of the authors have recently shown that a quite fast electrochemical abatement of Cr(VI) is achieved by using a RED stack equipped with 40 or 50 cell pairs [11,12] while Z. An and co-authors have very recently proposed a microbial desalination cell to desalinate saline water using a synthetic Cr(VI)-containing wastewater as the catholyte [37].

In particular, in this study we show that a water solution contaminated by Cr(VI) can be effectively and rapidly treated using a MRC equipped with a very low number of membrane pairs.

2. Materials and methods

2.1. Experimental procedures and apparatuses

RED and MRC experiments were performed in a custom made stack [11] equipped with two electroodic chambers ($10 \text{ cm} \times 10 \text{ cm} \times 2 \text{ mm}$), carbon felt cathode and anode (Carbone Lorraine, geometric area 100 cm^2), n (3, 5 and 7) cation- and $n+1$ anion-exchange membranes (Fuji), gasket integrated with spacers (Deukum, 0.28 mm thickness) and two external

cationic membranes (Nafion) to separate electrode compartments and side ones, creating n pairs of alternating high concentrated (HC) and low concentrated (LC) cells (Fig. 1). Anode and cathode are connected by an external circuit equipped with a resistance, an amperometer and a voltmeter (overall electrical resistance about 4.6 Ohm). Solutions used in HC and LC compartments were prepared by dissolving NaCl (analytical grade, Sigma–Aldrich) into deionized water. The LC solution entered from the cell next to the anode chamber and flowed in parallel through the LC cells in the stack, exiting from the cell next to the cathode chamber. The HC stream entered in the RED stack near the cathode and flowed in parallel through the HC cells in the stack, exiting from the cell next to the anode chamber. Two peristaltic pumps (General Control SpA) continuously fed the HC and LC solutions. Two closed-loop hydraulic circuits were used for electrode solutions that were continuously recirculated by two peristaltic pumps (General Control SpA) to the electrode compartments and to two different reservoirs maintained at 30°C . The cathode solution contained an aqueous solution of Na_2SO_4 (0.1 mol/dm^3 , Sigma–Aldrich) in the absence and in the presence of Cr(VI) ($\text{K}_2\text{Cr}_2\text{O}_7$, 25 mg dm^{-3} , grade Sigma Aldrich) at a pH of 2 obtained by addition of sulfuric acid (Sigma Aldrich). The anode solution contained for RED an aqueous solution of Na_2SO_4 (0.1 mol/dm^3) and for MRC experiments a LB broth (sodium chloride 5 g/L, tryptone 10 g/L, yeast extract 5 g/L) [38]. Flow rate of electrolyte solutions: 75 mL/min; flow rate of HC and LC solutions: 190 mL/min. Volume of electrolyte solutions: 250 mL each. For MRC experiments, the carbon felt was pre-acclimated in an undivided electrochemical cell equipped with compact graphite cathode and SCE reference with a working potential of -0.2 V vs. saturated calomel electrode (SCE) (by a Amel 2055 potentiostat) for at least 12 h with an organic solution consisting of 1/4 of *Shewanella putrefaciens* (SP) grown and 3/4 of Luria–Bertani (LB) broth. Once the current density has reached a plateau value, the acclimated anode was transferred to the MRC. SP was grown in LB medium. The SP strain was grown aerobically in a 50 mL flask in LB broth (Difco Laboratories, Detroit, MI). This culture was incubated at 30°C for 24 h with shaking at 100 rpm [38]. Cell counts about 1×10^8 cells/mL as determined by plating after serial dilution. Experiments were repeated at least twice with an error lower than 10%. Facultative SP, that is currently the object of a detailed investigation in our lab, was chosen as model exoelectrogenic microorganism for its tolerance to air, which makes easier the operations during the preparation of the MRC stack with the pre-acclimated anode.

Focused cyclic voltammetry was performed at 10 and 50 mV/s in a single-compartment, three-electrode cell, under nitrogen or air atmosphere, in order to roughly evaluate the anode and cathode potentials of some redox processes at carbon felt electrodes: reduction and oxidation of water and reduction of oxygen and Cr(VI). Working electrode was carbon felt (wet geometric surface

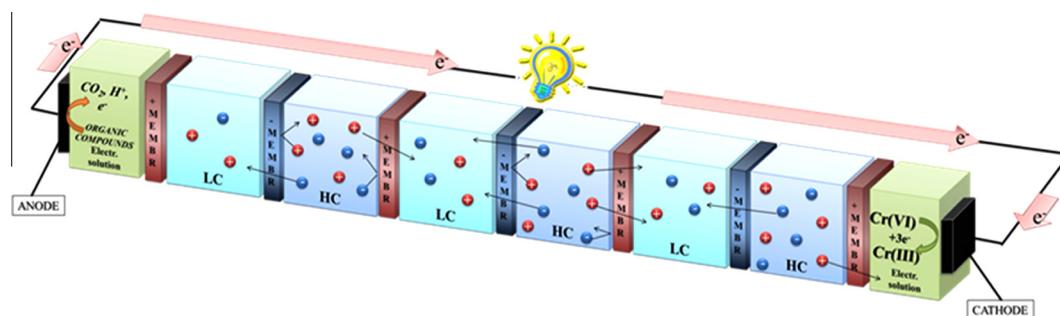


Fig. 1. Scheme of adopted MRC stack equipped with n cell pairs. “– membrane” indicates an anion-exchange membrane, “+ membrane” a cation-exchange membrane and HC and LC the high concentrated and low concentrated saline solutions, respectively.

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