



Treatment of acid mine drainage by forward osmosis: Heavy metal rejection and reverse flux of draw solution constituents



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ABSTRACT

Acid mine drainage (AMD) is one of the most relevant potential environmental liabilities of mining activities. They are formed when the excavated ore is exposed to rain and other natural erosion factors. This research evaluated the application of forward osmosis (FO) for the treatment of AMD. Rejection of several metals was determined, using a thin-film composite FO membrane, and two types of draw solutions: NaCl and NH_4HCO_3 . When using NaCl as draw solution, results showed high levels of rejection, exceeding 97%. Individual rejections were Mg: 98.9%, Al: 99.8%, Si: 97.1%, Ca: 99.3%, Mn: 99.6%, Fe: 100%, Co: 99.4%, Cu: 98.9%, Zn: 99.5%. When using NH_4HCO_3 as draw solute, precipitates were observed over membrane surface, preventing normal operation. Results show that transport of ions from draw solution (Ammonia and bicarbonate) is the cause of such phenomenon. FO has the potential to become an alternative for the treatment of AMD, based on the high levels of rejection for metals provided by FO membranes. However, attention must be paid to the reverse flux of ions from the draw solution to the feed, especially when they can react with feed solutes.

1. Introduction

Mining is one of the main industrial activities in Chile. It represents a relevant fraction of gross domestic product. Indeed, Chile is the mayor global copper producer. During 2015, Chile produced over 5,700,000 metric tons, close to 30% of global production [1]. Its reserves are estimated at 210,000,000 metric tons, which would also represent 30% of worldwide reserves. Mining industry has significantly contributed to the economic development of Chile and many other countries. However, this economic activity may generate relevant environmental risks, if measures are not taken to control the release of heavy metals to the environment. One of the most relevant potential environmental liabilities of mining activities are associated with the generation of acid mine drainages (AMD), which are formed when the excavated ore is exposed to rain and other natural erosion factors [2]. AMD is characterized by low pH, high sulphate concentrations and high levels of different metals such as iron (Fe), aluminium (Al), manganese (Mg), molybdenum (Mo), copper (Cu), nickel (Ni), arsenic (As), cadmium (Cd) and mercury (Hg) [3,4]. There is no precise estimation of the amount of AMD being generated in the world. However, it is believed that one mine can lead to the formation of hundreds to thousands of

cubic meters of AMD [5]. In developing countries, still only a fraction of this amount is treated or managed. AMD can contaminate ground and surface water bodies. It is toxic to aquatic organisms, can destroy ecosystems and diminish the available water for human activities [6]. Contamination of the environment with heavy metals can lead to serious health problems for human individuals, as a result of direct contact or food/water intake, if the food chain is contaminated [6].

AMD can be treated with different technologies. A common alternative is the addition of a chemical neutralizing agent, often limestone, to raise the pH and precipitate metals in the form of carbonate and hydroxides [7]. This technology can be efficient, but involves high operational costs and requires the safe disposal of bulky sludge [8]. Other methods such as flotation, adsorption, ion exchange and coagulation can be applied for the removal of metals from wastewaters [9]. The use of membranes seems a promising alternative, based on the ability of some of them to separate small ions. Membrane technologies that have been studied for treatment of AMD are nanofiltration (NF), electro-dialysis (ED), membrane distillation (MD) and reverse osmosis (RO) [6,10–12].

Forward osmosis (FO) is a fairly novel membrane process, that has received increasing attention during the last few years [9]. FO process

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promotes water flux away from a solution to be concentrated, by contacting it with a solution presenting a higher osmotic potential, normally referred to as draw solution (DS). Contact of both solutions is mediated by a membrane that enables the permeation of water, but rejects solutes [13]. Even though first developments in FO were based in the use of RO membranes, specially developed membranes for FO processes are now readily available in the market [14,15]. When the recovery of high quality water is a primary objective, the process requires regeneration of the draw solution. A secondary process is then required. Energy requirement of latter step is decisive to determine overall feasibility of FO [16,17]. The use of draw solutions based on ammonium bicarbonate (NH_4HCO_3) has been proposed to reduce energy requirements of recovery step. This compound can be separated from water by low temperature distillation [18]. Solar radiation may be used to provide the energy required for distillation, in areas presenting high irradiation levels. This is the case of the north of Chile, where most mining operations take place. Then, a process of forward osmosis with ammonium bicarbonate as draw solution and a low energy distillation process for its recovery may represent an interesting alternative for the treatment of acid mine drainage. Water recovery could be provided at the time AMD is concentrated. An increase in concentration would reduce the volume to be treated, facilitating for example operations dedicated to metals recovery or inertization.

This research evaluated the application of FO for water recovery from acid drainage coming from copper mining operations. Rejection of several metals was determined, using a thin-film composite FO membrane, and two types of draw solutions: NaCl and NH_4HCO_3 . Potential limitations of NH_4HCO_3 as draw solution are evaluated and discussed.

2. Materials and methods

2.1. Feed and draw solutions

A sample of real acid mine drainage was obtained from a copper mine from north of Chile. AMD was characterized measuring the concentration of the following elements or compounds: aluminium, calcium, cobalt, chromium, copper, iron, magnesium, manganese, silicon, zinc and sulphate. Draw solutions were prepared using analytical grade reagents and deionized water. NH_4HCO_3 solution was prepared based on the procedure reported by Lu et al. [19]: mixing a 0.9 M solution of ammonium bicarbonate (NH_4HCO_3) and 0.1 M solution of ammonium hydroxide (NH_4OH).

2.2. FO membrane

Thin-film composite (TFC) membrane was used during this study. Membrane was provided by Porifera Inc. (FOMEM-0415 – Hayward, CA, USA). Properties of membrane are shown on Table 1. Water (A_0) and salt (B_0) permeabilities are provided. The structural parameter (S_0) is related to the internal structure of the membrane, and determines internal concentration polarization (ICP) [20]. The zeta potential (ζ) is a measurement of the surface charge of the membrane and impacts the rejection mechanism by electrostatic repulsion. Contact angle indicates the hydrophilicity of the membrane.

2.3. FO filtration setup

Fig. 1 presents a schematic representation of the filtration setups

Table 1
Membrane and active layer properties. Adapted from Blandin et al. (2016a).

A_0 (L/ $\text{m}^2 \cdot \text{h} \cdot \text{bar}$)	B_0 ($\times 10^{-7}$ m/s)	S_0 (μm)	Zeta Potential (pH = 7) (mV)	Contact Angle (degrees)
2.1	1.2	344	-13.7 ± 1.9	36 ± 6

used in this research. Each one consisted of 2 acrylic cylindrical chambers, separated by a FO membrane. Each chamber had an internal diameter of 4.6 cm, and could accommodate 180 mL of liquid. Available membrane area was 13.5 cm^2 . Mixing was provided in both chambers by means of magnetic stirrers. Feed and draw solutions were contained in graduated cylinders. Then, water permeation could be observed by changes in the liquid level of cylinders, enabling evaluation of flux. A peristaltic pump was used to circulate draw solution from the graduated cylinder to the corresponding chamber. A second peristaltic pump was programmed to feed a brine at regular intervals to the graduated cylinder containing draw solution, to keep the draw solution concentration constant at 1.0 M. This was necessary to counteract the dilution effect of water flux. Concentration of the draw solution was estimated by measuring conductivity (OAKTON, Salt 6+) on regular intervals.

2.4. Filtration experiments with model solutions and AMD

A series of filtration experiments were conducted using the setup shown in Fig. 1. Experiments are summarized in Table 2. Assays with clean water were performed to determine maximum water fluxes. Three different concentrations of the tested draw solutions were applied. Filtration experiments were then conducted using CuSO_4 as a model feed, at 3 different concentrations: 1.6, 16 and 160 mM. This compound was tested, considering that sulphate and copper were the most relevant ions present in AMD. Both NaCl and NH_4HCO_3 were tested as draw solutions, at a concentration of 1.0 M. Finally, assays with real AMD were performed, using the two draw solutions.

All experiments were made with the active layer facing the feed solution. No external pressure was applied. All experiments were done in triplicates, at room temperature. Filtration assays with CuSO_4 and AMD were conducted until the volume of the feed was reduced to 50% of its original value (concentration factor of 2).

2.5. Calculation of rejection factors and reversed fluxes rates

Rejection factors were evaluated for different ions present in the tested feeds. Ions concentrations on both draw and feed solution were determined by the end of experiment. Then, rejection (R) was calculated according to Eq. (1):

$$R = 1 - \left(\frac{C_p}{C_f} \right) * 100 \quad (1)$$

where C_f represents the concentration of the ion on the feed side and C_p is the concentration of the ion on the permeate. Concentration on the permeate is evaluated according to Eq. (2):

$$C_p = \frac{C_d * V_d}{V_p} \quad (2)$$

where C_d is the concentration on the draw side, V_d is the volume of draw solution by the end of experiment and V_p is the volume of water that permeates from the feed to the draw side.

Reverse flux is the term usually used to refer to the unwanted transport of solutes from the draw to the feed side, in opposite direction of the water flux. Reverse solutes flux J_S was calculated as described by Eq. (3):

$$J_S = \frac{C_f * V_f - V_p}{A_m * \Delta t} \quad (3)$$

where V_f is the initial feed volume, V_p is the volume of liquid that permeated to draw side, Δt is time and A_m is the effective area of membrane.

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