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Comparison of Nanofiltration and Direct Contact Membrane Distillation as an alternative for gold mining effluent reclamation

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ABSTRACT

Effluent reuse is becoming an environmentally-economically viable option for industries that have been struggling with environmental/legislative pressure and high costs of water supply and effluents discharge. Mining effluents are promising for reuse, but for this, (hazardous) metals and ions such as sulfate must be removed efficiently. This study aimed to compare the technical-economic performance of Nanofiltration (NF) and Direct Contact Membrane Distillation (DCMD) on gold mining effluent treatment. The results showed that both technologies are promising for mining effluent reclamation as high pollutants removal rates were observed (rejection > 73% and > 96% for NF and DCMD, respectively). DCMD showed lower flux than NF and was limited to operate up to permeate recovery rate (RR) of 40% due to severe fouling ($CaSO_4$ precipitation), but produced a higher quality permeate with no reuse restriction, low energy requirement and lower associated costs due to use of the effluent residual heat. Capex and Opex (for membrane lifespan of 1–5 years) were estimated at US$ 575,490.30 and 2.00–2.10 US$/m^3 for NF and US$ 305,483.85 and 0.13 to 0.27 US$/m^3 for DCMD systems. The permeate reuse would reduce water consumption in 490,560 m^3 year^{-1}, which represents almost 50% of the industrial water demand.

1. Introduction

Despite the great economic importance of gold-based ores mining, this activity generates several environmental impacts. The production of contaminated effluents is a drawback that has been intensified because of the lower quality of the available ores, requiring more water for its processing, and mining activity intensification.

The composition of the mining wastewaters is very varied, being dependent on the type of ore/metal mined, local geochemical characteristics, the processes that are used, etc. But in general, regardless of their specificities, mine waters present important constituents such as Al^{3+}, Si^{+4}, Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, Cl^{-}, CO_3^{2-}, HCO_3^{-} and SO_4^{2-} ions and may also present heavy metals/metalloids such as Hg, Cd, Cr, Hg and As [1,2]. This is an aggravating factor and makes mine waters require a proper treatment.

The environmental and legislative pressure that the industry has been suffering in addition to the high costs of water supply and effluents discharge has led to the reuse of effluents which is becoming an environmentally and economically viable option for industries. Mining effluents are promising for reuse, but the presence of metals, especially hazardous metals, and ions such as sulfate (corrosive potential and high fouling potential) represent a barrier to such use and must be removed.

In this context, membrane separation processes such as nanofiltration (NF) and membrane distillation (MD) stand out. Both NF and MD are systems able to retain salts and dissolved molecules, presenting a great application in the treatment of effluents and reuse water generation [3–5].

The NF has already been applied to remove arsenic and sulfate from contaminated water and synthetic solutions [6–10] and also for the treatment of mining effluents [11–17], showing good results. Despite its removal efficiency, NF membranes are sensitive to high temperatures [18], which represents a limitation for the treatment of certain wastewaters, like gas scrubber effluents (60 °C). The strategy of reducing the temperature before membrane filtration requires expenditure on cooling [19], making the use of MD a promising alternative.

MD is a process that has an even higher efficiency in the removal of pollutants, being able to concentrate solutions until their saturation without a significant decline of permeate flux [20–22]. Direct Contact Membrane Distillation (DMCD) is the most used and operationally simplified configuration, being much used in desalination plants, but

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also showed good results in acid production/concentration [23] and in the treatment of a mining solution containing sulfuric acid and several metals [24]. Besides this effectiveness and minimum cost of operating capital, this process requires a low heat supply, which can be supplied by industrial residual heat or solar energy, resulting in a low fossil energy requirement [21,22,25].

Therefore, the objective of this work was to compare the use of NF and DCMD in the treatment of a real gold mining effluent. The processes effectiveness was characterized in terms of permeate flux and rejection of arsenic, sulfate, calcium, and magnesium. Moreover, the fouling behavior of NF and DCMD were assessed and compared. This study is timely and appropriate in light of the technical and economical comparison between two well-known effective technologies for the removal of pollutants and wastewater reclamation in the mining industry. It allows for a better understanding and orientation of decision making in industry exploring a case of a real effluent with its specifications.

2. Materials and methods

2.1. Gold mining effluent

The effluent used in this study comes from the sulfuric acid production plant of a gold mining company located in the state of Minas Gerais (Brazil). This effluent is generated in the gas scrubber used to remove impurities from the gas generated after the ore, which is rich in sulfur and arsenic, is boiled. The gas is composed mostly of sulfur dioxide, which is processed and used in the manufacture of sulfuric acid.

The effluent generated in the gas scrubber is acid, contains high concentrations of ions such as calcium, magnesium and mainly sulfate, and also presents arsenic contamination, both arsenite [As(III)] and arsenate [As(V)], in high concentrations (Table 1).

Before being subjected to NF and MD, the mining effluent underwent previous filtration to remove the suspended solids. Filtration was performed in a vacuum pump using a commercial cellulose filter containing 2.5 μm pores (Whatman® quantitative filter paper, ashless, grade 42). No pH adjustment was performed.

2.2. Experimental setup

NF assays were performed on a laboratory scale in the filtration unit shown in Fig. 1. It consisted of a feed tank, followed by a rotary-vane pump, which is connected to a speed controller, a rotameter and a stainless steel membrane module for a flat sheet membrane. After the cell, there is a manometer, thermometer and between those, a needle valve for pressure adjustment. Before being recirculated to the feed tank, the concentrate passed through a chiller in order to maintain the effluent at the temperature of the test (25 °C).

In the stainless steel cell, a new 9 cm diameter flat membrane (0.0064 m^2 filtration area) was placed along with a 28 mil (25.4 μm) feed spacer at the top to ensure flow distribution. The membrane used was a polyamide NF90 membrane (Dow Filmtech) with a molecular weight cut-off between 100–200 Da [26]. The measurement of the permeate flux was done by collecting the volume of permeate in a measuring cylinder over a period of 60 s.

MD assays were also performed on a laboratory scale in the filtration unit shown in Fig. 2. MD module is composed of two compartments between which the membrane was disposed. In one of the compartments, the effluent was circulated and in the other, cooled water. The feed tank is followed by a thermometer, a peristaltic pump, and a rotameter before the membrane module. Before being recirculated to the feed tank, the concentrate passed through a heating system in order to maintain the effluent at the temperature of the test (60 °C). The cold water reservoir (distillate tank) is placed on a digital balance and it is also followed by a thermometer, a peristaltic pump, and a rotameter before the membrane module. The distillate passes through a cooling system (chiller) before being recirculated to the distillate tank.

The temperature differential is needed so that the permeate flux can be established. This flux was measured using the data collected in the digital balance as the increase in the distillate mass is provided by permeate production.

For the membrane cell, a Sterlitech® (CF042D Crossflow Cell) cell was used, in which was positioned a new Sterlitech® PTFE microfiltration membrane. This membrane has an average pore size of 0.2 μm, liquid entry pressure < 14.5 Psi and for the experiment, the filtration area was 0.0042 m^2.

Table 1
Physicochemical characterization of the gold mining effluent.

Parameter	Unit	Mean and standard deviation
pH		1.80 ± 0.23
Conductivity	(μS/cm)	12420 ± 1010
Sulfate	(mg/L)	5501 ± 614
Calcium	(mg/L)	444 ± 263
Magnesium	(mg/L)	288 ± 129
Arsenic	(mg/L)	580 ± 94
As (V)	(mg/L)	*88 ± 51*
As (III)	(mg/L)	*440 ± 126*

2.3. Experimental procedure

Before the tests, the membranes passed through a cleaning procedure in which they were submerged in a hydrochloric acid (HCl) 0.2% w/w solution for 20 min and then washed with distillate water. After chemical cleaning, water permeability was measured.

Before the systems were used to treat the real effluent, distilled water was used as feed in both NF and DCMD systems and permeation occurred until reaching a steady permeate flux.

NF and DCMD tests were performed using real mining effluent and adopting the following operational conditions: feed velocities were maintained at 1.9 m/s (Re = 839) and 0,1 m/s (Re = 581) for NF and DCMD respectively. In NF, the applied transmembrane pressure was 10 bar and the temperature was maintained at 25 ± 2 °C. Tests were performed in batch mode with recirculation of the concentrate. In DCMD, temperatures were maintained at 60 °C for the feed and 20 °C for the distillate. Both NF and DCMD were performed for at least the recovery rate of 45%. The permeate flux and conductivity were continuously measured and permeate samples were taken out at different times for physicochemical analysis.

2.4. Analytical methods

The mining effluent and permeates were characterized in terms of the following physicochemical parameters: pH (pHmeter Qualxtron QX 1500); conductivity (Hanna conductivity meter HI 9835); calcium and magnesium cations (Dionex ICS-1000 ion chromatograph equipped with AS-22 and ICS 12-A columns); total arsenic, As (III) and As (V) [27]; and total dissolved solids, sulfate and chloride (APHA, [48]).

2.5. Calculations

The NF permeate flux [$J_{P(NF)}$] can be calculated by Eq. (1):

$$J_{P(NF)} = \frac{\Delta V_p}{A_m \cdot \Delta t} \quad (1)$$

where ΔV_p is the volume of permeate collected, Δt is the collection time, and A_m is the effective membrane area. The permeate recovery ratio (RR_{NF}) can be defined by Eq. (2):

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