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Integrated UF–NF–RO route for gold mining effluent treatment: From bench-scale to pilot-scale

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ABSTRACT

This study focused on the performance of an UF–NF–RO integrated system treating a gold mining effluent from the pressure-oxidation stage in order to concentrate metals, and recover acid and water. Aspects such as the most suitable cross-flow velocity, cleaning procedure, and water recovery fraction were evaluated. For cross-flow velocities, a threshold value of 1.5×10^{-2} m/s was found for NF and RO stages. Besides, the system had its best performance while operating at recovery fractions of 90%, 40–50%, and 50% for UF, NF, and RO, respectively. Concentration factors of 2.0 and 2.7 were found in metals from the NF retentate and sulfuric acid from the RO retentate respectively. For cleaning purposes, hydrochloric and oxalic acid demonstrated the best cost-benefit. A long-term operation inside the mining company was assessed and the results confirmed that NF and RO association allowed for the recovery of a purified acid stream, which may be reused in the ore processing; the production of a metal enriched stream, that can be transferred to a subsequent metal recovery stage; and the generation of high quality reuse water. The total cost of the proposed route was US\$ 1.137/m^3 of effluent, including UF and NF concentrate neutralisation.

1. Introduction

Gold is a valuable metal that possesses a wide range of uses. However, the environmental impact associated with gold exploitation is one of the main challenges that the mining sector must overcome [1], since the pressure for cleaner production principles is increasing worldwide, along with more restrictive environmental standards, including those associated with water resources [2].

Normally, effluents resulting from ore processing contain hazardous substances such as heavy metals, metalloids, acids and process chemicals [3]. To address their treatment, different methods can be employed, such as neutralisation, ion-exchange, electrochemical and membrane separation processes [4,5]. Up until now, the commonly used solution in industry is the neutralisation process, which uses large amounts of alkaline reagents such as calcium carbonate, calcium oxide, calcium hydroxide, magnesium hydroxide, and sodium hydroxide [6]. Although this process is simple, its main disadvantage is the generation of large volumes of sludge containing heavy metal compounds that must be disposed of. Moreover, valuable acids and metals are lost during the neutralisation processes that could instead, for example, be recovered and reused in the hydrometallurgical processes.

As an alternative to neutralisation, the membrane processes present some attractive advantages, such as the generation of a permeate with higher quality that can be directly reused for industrial purposes, the concentration of metals, and the recovery of acids. This is a viable approach that not only benefits the environment and the economics of the operation, but it can also provide a secondary source of valuable metals [7].

In this context, the conjugation of ultrafiltration–nanofiltration–reverse osmosis (UF–NF–RO) processes may be applied to concentrate the metals, and recover acid and water. This integration is becoming especially interesting due to the development of membranes more resistant to extreme pH conditions [8] and the ability to treat water to an excellent quality for reuse, let alone the possibility of concentrating wastewater byproducts for recovery. This relies on the fact that NF membranes combine the characteristics of both ultrafiltration (UF) and RO membranes, thus presenting a higher flux than that observed in RO. NF membranes also have good retention for multivalent ions (e.g. metals) [9,10]. On the other hand, RO membranes are mainly dense and therefore the retention of compounds with lower molar mass [11], such as acids, is very high. The UF membrane can be used as pre-treatment for the removal of suspended solids and to

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mitigate fouling of membranes in the subsequent NF and RO processes. Despite this potential, membrane fouling still remains a challenge to membrane systems. Fouling raises costs due to an increased energy demand, additional labour for maintenance, and chemical costs for cleaning, as well as reducing the usable life of the membranes [8].

Taking into consideration the intrinsic characteristics of NF and RO membranes, studies that were conducted at a bench-scale focused on the treatment of gold mining effluent treatment through membrane technology [12–16]. In a previous study, the authors evaluated the integration of MF–NF–RO processes in the treatment of gold mining effluent from the pressure-oxidation (POX) stage. The results showed that the NF attained high sulfuric acid permeation (82%) and substantial acid purification (77%), whereas RO increased the acid content by 99% in relation to the feed solution. The final RO permeate had low conductivity and reduced the contents of total solids and contaminants, thus revealing a promising potential in industrial reuse. Furthermore, the acid recovery did not lead to the loss of metals of high commercial value, which were present in the raw effluent since their retention in the concentrates of the UF and NF was above 95%.

However, although these studies are indispensable in understanding the fundamentals of membrane fouling, a few aspects, such as hydraulic conditions and operating time, still remained as issues since these conditions were very different in a bench-scale compared to what occurred after scaling-up [17]. Hence, in order to evaluate these issues, piloting is an alternative that allows a more representative assessment of full-scale membrane installations [18].

Consequently, this study evaluated the performance of a pilot-scale UF–NF–RO integrated system treating a gold mining effluent from the pressure-oxidation (POX) stage. Aspects such as the most suitable cross-flow velocity, cleaning procedure, and water recovery fraction (RR) were evaluated. In addition, a long-term operation inside the gold mining company was assessed in order to observe the system's behaviour and potentials.

2. Materials and methods

2.1. POX effluent

When the exploitation of gold is made from refractory ore minerals, a pre oxidation step is required before the cyanidation leaching, since gold is mainly locked up in sulfidic host minerals and therefore cannot be directed leached. In this case, the gold ore slurry goes through a hydrometallurgical oxidation under elevated pressure and temperature, in the presence of acid [19]. Consequently, an effluent, rich in sulfuric acid and metals such as nickel, copper, and cobalt, is generated. Table 1 presents the main characteristics of POX effluent collected from a gold mining company located in Minas Gerais, Brazil.

Table 1
Main physicochemical characteristics of POX effluent and its deviations.

Parameters	POX effluent
pH	1.3 ± 0.2
Conductivity (mS/cm)	18.7 ± 15.6
Suspended solids (mg/L)	73 ± 23.8
Sulfate (mg/L)	18.2 ± 3.8
Calcium (mg/L)	347 ± 35
Nickel (mg/L)	102 ± 19
Cobalt (mg/L)	20 ± 4
Copper (mg/L)	95.7 ± 24.6
Aluminium (mg/L)	445 ± 183
Arsenic (mg/L)	14.9 ± 3.8
Iron (mg/L)	467 ± 206
Manganese (mg/L)	2575 ± 178
Zinc (mg/L)	82 ± 8.7
Potassium (mg/L)	6.4 ± 0.7
Sodium (mg/L)	18.6 ± 7.2
Chloride (mg/L)	36 ± 3

2.2. Description of the treatment process

Fig. 1 shows the schematic of the proposed route for POX effluent treatment. In this process, the UF process works as a pretreatment to avoid major particles in the NF and RO systems. Hereinafter, the NF stage retains and concentrates metals on the NF retentate as the acid permeates the membrane, hence separating them. Afterwards, the NF permeate (a metals-free solution with a low concentration of acid) is then subjected to the RO stage. At this point, the water (RO permeate) is separated from the acid, concentrating the acid in the RO retentate. Thus, by the end of the process, three streams can be seized: NF retentate with elevated metal concentration, RO retentate rich in acid, and RO permeate having water quality for industrial reuse.

2.3. Membranes

Ultrafiltration was performed with a submerged commercial hollow fiber membrane module (ZeeWeed) as a PVDF-based polymer with an average pore diameter of 0.04 μm. On the other hand, nanofiltration was performed with a thin-film polymer composite DK membrane from GE, with a molecular weight cut-off of 150–300 DA, operational pH range of 3–9, and a $MgSO_4$ (2000 mg/L) rejection of over 98% [20]. Reverse Osmosis was performed with a polyamide composite SG membrane, also from GE, with an average NaCl (2000 mg/L) retention of 98.5% and operational pH range of 2–10 [21]. The membranes were selected according to preliminary lab-scale tests, which suggested the suitability of these membranes for large-scale evaluations based on their high permeate flux and retention values, apart from elevated membrane chemical stability.

2.4. Experimental setup

Tests were conducted in bench and pilot-scale units. Tests conditions are described below.

2.4.1. UF pretreatment

The UF system consisted of a submerged UF membrane with a filtration area of 0.9 m^2 with a maximum operational pressure of 0.7 bar, and provided with a diaphragm pump equipped with a speed controller. The ultrafiltration was conducted at a pressure of 0.7 bar and a recovery fraction of 90% for all tests performed, including long-term operation.

2.4.2. Bench-scale NF/RO

The unit used for NF and RO bench tests consisted of a supply tank, a rotary vane pump (maximum operational pressure of 15 bar), a rotameter, a membrane cell, a manometer, and a thermometer as described in a previous publication [15]. Transmembrane pressure and feed flow rate were adjusted by a needle-type valve. The stainless-steel filtration cell had a filtration area of 75 cm^2 (a diameter of 9.8 cm), and a feed spacer with 711 μm (28 mil) was placed over the membrane to promote flow distribution. For the bench-scale tests, the UF permeate was stored and placed manually in the NF feed tank in each of the performed tests. The same was done for NF permeate used as feed in the RO stage. All bench scale tests were operated in a flat sheet membrane, under tangential flow.

2.4.3. Pilot-scale UF–NF–RO

For pilot-scale tests, the units described in Section 2.4.2 were adapted, and the stainless-steel bench cell was substituted with a vessel containing a spirally wound NF and an RO membrane with a filtration area of 2.6 m^2. In these vessels, the feed entered in parallel to the membrane, characterizing a cross-flow operation. Fig. 2 shows a schematic of the process.

Membranes operated in both batch and continuous modes. During