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Concentration-polarization in nanofiltration of low concentration Cr(VI) aqueous solutions. Effect of operative conditions on retention

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Nanofiltration of low concentration Cr(VI) aqueous solutions by using an AFC80 membrane from PCI has been shown to be feasible with high retentions that increase for higher pressures, temperatures and recirculation speeds. This makes nanofiltration a suitable tool to decrease Cr(VI) levels below those recommended by the world health organization. In some cases this would require a few consecutive NF steps.

Nanofiltration of Cr(VI) has been modelled by using the Spiegler-Kedem theory modified to include concentration-polarization. This allowed us the simultaneous evaluation of the three characteristic parameters of the model: reflection coefficient σ, solute permeability P and mass transfer coefficient K_m from rejection experiments. Both σ and P are shown to decrease with concentration and to reach a relatively high concentration plateau. Other methods habitually used to evaluate the mass transfer coefficient have also been compared showing, when applicable, similar results for K_m.

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1. Introduction

Although the importance of trustworthy water sources is universally known, the growth of their contamination is also general. Water resources can be contaminated by industrial or urban waste waters or indirectly through: soil, atmosphere and rain waters (Vijayaraghavan and Yun, 2008). Especially relevant as water contaminants are heavy metals - with atomic weights between 63.5 and 200.6 and specific weights over 5 (Bilal et al., 2013)- that are toxic even at low concentrations. For example, mercury, lead, cadmium and chromium (VI) are quite hazardous, while other not so highly toxic metals as copper, nickel, cobalt and zinc, can be problematic due to their growing concentrations in water (Moore, 1990; Rozada et al., 2008; Volesky, 1990). Heavy metals are highly soluble and accumulate in living organisms eventually reaching quite high concentrations with devastating effects on health. This explains why wastewaters must be treated to eliminate heavy metals prior to discharging (Barakat, 2011; Kaur et al., 2013; Kurniawan et al., 2006).

Chromium is usually present in environment and water, as either Cr (III) or Cr (VI). The International Agency for Research on Cancer, IARC, classifies Cr (III) within the group 3 (that includes chemicals that are considered as not classifiable as to its carcinogenicity to humans) while includes Cr (VI) within group 1 (human carcinogens) (McNeill et al., 2012; Pellerin and Booker, 2000; US-EPA, 1998; WHO-IARC, 1990). The effects of Cr (VI) after ingestion were revised by the US Environmental Protection Agency, US-EPA (US-EPA, 2014a, b).

More and more restrictive normative is being put in place in developed and developing countries concerning industrial discharges that are increasingly monitored and restrained (US-EPA, 1992, 2006). The US Environmental Protection Agency established the maximum contaminant level of total chromium in 0.1 mg/L (100 ppb) (US-EPA, 2009). The World Health Organization, WHO, established as a provisional maximum value for total chromium in drinking water the concentration of 0.050 mg/L (50 ppb) (WHO, 2003).

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Some sources of chromium contaminated effluents include those from: inorganic dyes with chromium and cadmium sulfide; tanning; galvanoplasty; oil refining that generates chromium along with vanadium and nickel; and others where chromium can be used as catalyzer (Jordão et al., 1997; Kotaś and Stasicka, 2000; Romero-González et al., 2005; Sörme and Lagerkvist, 2002). Galvanoplasty is one of the industries that generate more water contaminated with heavy metals including Cr, Cd, Cu, Ni,Zn, As, Pb (Kurniawan, 2002).

Different procedures have been used to eliminate Cr (VI): selective precipitation (Djedidi et al., 2009); bioadsorption on active carbon (Hong et al., 2008; Wu et al., 2008); other adsorbents (Acosta et al., 2005; Vaghetti et al., 2008); conventional electrochemical reduction (Lakshmipathiraj et al., 2008); microbial fuel cells (Li et al., 2008) and photoelectrochemical techniques (Shim et al., 2008). Nevertheless, most of these and other conventional procedures are inefficient or too expensive, especially when concentrations are in the range from 1 to 100 mg/L (Ahluwalia and Goyal, 2007; Nourbakhsh et al., 1994), because Cr (VI) elimination would require too many chemicals or lead to sewage sludge that would result quite difficult to decontaminate.

Electrodialysis (Nataraj et al., 2007) has been tested to minimize the appearance of contaminated slurry ionic exchange (Rengaraj et al., 2001). Several other membrane processes are also being investigated including: liquid membranes (Alonso et al., 1994; Kumbasar, 2008) or activated transport (Gherasim and Bourceanu, 2013; Melita and Popescu, 2008). Micellar and complexing ultrafiltration have also been tested (Bade et al., 2008). All these techniques have some drawbacks such as their energy cost, the addition of chemicals or the procedures are not appropriate for the treatment of large volumes of effluent.

A few years ago, nanofiltration started to be investigated in view of the elimination of chromium (VI) (Liu et al., 2008) due to its low energy consumption and the high fluxes attained by the process. Different nanofiltration membranes have been used (Hafiane et al., 2000; Muthukrishnan and Guha, 2008; Ren et al., 2010; Taleb-Ahmed et al., 2002; Xu et al., 2014).

Here we will test the capacities of a nanofiltration membrane, the AFC80 from PCI to eliminate low concentration Cr (VI). A deep study of the influence of operative conditions including pressure, concentration, speed through the feed loop and temperature on flux and retention is also presented. Observed and true retention are studied and modelled within the frame of thermodynamics (Spiegler-Kedem transport model), for the transport through the membrane, and fluid mechanics (film layer model), for the mass transfer through the feed channel. Of course, the use of Thermodynamics has its advantages and drawbacks. By definition, it doesn't need any knowledge of the detailed mechanism of the process at a molecular level what frees the results from any bias caused by an insufficient knowledge of these details. This also makes impossible to test these detailed molecular level models but make their results robust. This work participates of this strength, although we will make reasonable assumptions on the causes of the behavior of the thermodynamic parameters obtained.

2. Theory

2.1. Spiegler-Kedem equations

The classical Spiegler-Kedem model, which is founded on Irreversible Thermodynamics, offers a simple description of solute transport in both RO and NF processes (Kedem and Katchalsky, 1963; Spiegler and Kedem, 1966). The Irreversible Thermodynamic study of transport through a convective and diffusive membrane leads to the following expressions for the volume, J_V, and solute, J_s, fluxes in terms of the drop in hydraulic pressure Δp and in osmotic pressure $\Delta\pi$:

$$\left.\begin{array}{l} J_V = L_p(\Delta p - \sigma\Delta\pi) \\ J_s = \bar{c}(1-\sigma)J_V + P\Delta\pi \end{array}\right\} \tag{1}$$

$\bar{c}$ is the average concentration along the membrane, $(c_p + c_m)/2$; c_p corresponds to the permeate concentration and c_m to the concentration on the membrane active layer. These equations appear in terms of three parameters: the hydraulic permeability, L_p, the permeability, P, and the reflection coefficient, σ.

According to this, the incomplete semipermeable character of a membrane is characterized by σ. An osmotic pressure difference, $\Delta\pi$, across a non-totally semipermeable membrane can be compensated by an applied pressure, Δp, giving a zero volume flow. This means that Δp would be smaller than $\Delta\pi$ and it must be $\sigma < 1$.

If it is assumed (Ahmed, 2013) that only pure water would pass through a certain portion of the membrane, perfectly rejecting the solute, with a volume flux J_w while the solute passes only along some water through the rest of the membrane giving a volume flux J_m, in such a way that:

$$J_V = J_w + J_m \tag{2}$$

And assuming that the flux, J_w, is proportional to the effective trans-membrane pressure (difference between applied and osmotic pressure across the membrane), whereas in the non-ideally rejecting portion, the flux, J_m, is simply proportional to the applied pressure, according to Darcy's law, we would obtain, from Equation (1):

$$\left.\begin{array}{l} J_w = \sigma L_p(\Delta p - \Delta\pi) \\ J_m = (1-\sigma)L_p\Delta p \end{array}\right\} \tag{3}$$

But:

$$\left.\begin{array}{l} J_s = J_V c_p \\ J_s = J_m c_m \end{array}\right\} \tag{4}$$

Then:

$$\frac{c_p}{c_m} = \frac{J_m}{J_V} = \frac{(1-\sigma)L_p\Delta p}{L_p(\Delta p - \sigma\Delta\pi)} = \frac{(1-\sigma)\Delta p}{\Delta p - \sigma\Delta\pi} \tag{5}$$

And thus, σ is:

$$\sigma = \frac{\Delta p R}{\Delta p - \Delta\pi(1-R)} = \frac{R}{1 - \frac{\Delta\pi}{\Delta p}(1-R)} \tag{6}$$

with $R = 1 - \frac{c_p}{c_m}$ the retention coefficient. Or equivalently:

$$R = \sigma\frac{\Delta p - \Delta\pi}{\Delta p - \sigma\Delta\pi} = \sigma\frac{1 - \Delta\pi/\Delta p}{1 - \sigma\Delta\pi/\Delta p} \tag{7}$$

Eqs. (6) and (7) allow to conclude that when $\Delta\pi/\Delta p \rightarrow 0$ then $R = \sigma$. Of course, $\Delta\pi = 0$ only if the membrane were totally non retentive. This would lead to $R = \sigma = 0$. When concentrations are low, even quite retentive membranes can lead to very small $\Delta\pi$ and $R \approx \sigma \neq 0$.

Eq. (1) can be rearranged, using Eq. (3), to:

$$\mathrm{R} = \frac{\sigma(1-F)}{1-\sigma F} \tag{8}$$

$$F = \exp\left[-\frac{(1-\sigma)}{P}J_V\right] \tag{9}$$

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