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Antifouling grafting of ceramic membranes validated in a variety of challenging wastewaters

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ABSTRACT

Compared to traditional separation and purification techniques, membrane filtration is particularly beneficial for the treatment of wastewater streams such as pulp and paper mill effluents (PPME), olive oil wastewater (OOWW) and oil/gas produced water (PW). However, severe membrane fouling can be a major issue.

In this work, the use of ceramic membranes and the potential for the broad applicability of a recently developed antifouling grafting was evaluated to tackle this issue. To this end, the fouling behavior of native and grafted membranes was tested in the selected difficult wastewater streams, both in dead-end and in cross-flow mode. In addition, the quality of the produced permeate water was determined to assess the overall performance of the investigated membranes for reuse or recycling of the treated wastewater.

The obtained results show that grafting significantly enhances the antifouling tendency of the ceramic membranes. Particularly, the membrane grafted with methyl groups using the Grignard technique (MGR), showed in all cases no or negligible fouling as compared to the native membrane. As a consequence, the process flux or filtration capacity of the MGR membrane in cross-flow is always higher and more stable than the native membrane, even though the grafting lowers the pure water flux. Hence, the inert character of the MGR membrane is repeatedly proven and shown to be broadly applicable and generic for anti-fouling, without loss in permeate quality.

Moreover, in case of OOWW, the quality of the MGR permeate is even better than that of the native membrane due to its lower fouling. All results can be explained taking into account the physico-chemical properties of foulants and membranes, as shown in previous work. In conclusion, the use of MGR membranes could provide an optimum economical solution for the treatment of the selected challenging wastewaters.

1. Introduction

The production of wastewater is unavoidable, as water is a vital part of operational processes in different types of industries. The increasing demand of water for industrial use, originating from increasing economic activity (UNESCO, 2012) and the prompt industrial growth, has led to plenty of wastewater production every year (Chakrabarty et al., 2008; Cheryan and Rajagopalan, 1998; Ju et al., 2008; Lin and Lan, 1998; Yusoff and Murray, 2011; Zaidi et al., 1992; Zhu et al., 2014). This wastewater is one of the biggest threats for water supply worldwide. The reliable key factor for diminishing the water scarcity would be the purification or treatment of industrial wastewater (McCloskey et al., 2010; Morales Chabrand et al., 2008; Cambiella et al., 2007).

In this work, we focus on three important industrial wastewater streams: 1) pulp and paper mill effluents, 2) olive oil wastewater and 3) oil/gas produced water (wastewater produced during the pumping out of oil/gas from the earth known as produced water), which contribute extensively to the whole of industrial wastewater in the world. These industrial wastewater streams are not only large in quantity, but they are also very difficult to treat because of their complex chemical composition.

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1.1. Pulp and paper mill effluents (PPME)

Pulp and paper production is a water-intensive process. Paper industry ranks third in the world after metal and chemical industries in terms of fresh water consumption and wastewater creation (Beril Gönder et al., 2011). At present, a number of pulp and paper mills treat their wastewater by biological treatment systems. However, after the biological treatment, the effluent still contains significant amounts of microorganisms, dyes and suspended solids. In addition, inorganic compounds cannot be removed effectively by biological treatment. Moreover, the pulp and paper mills generate a variety of pollutants depending upon the type of the pulping process, making it difficult to treat wastewater by biological treatment.

Hence, advanced treatment is essential to improve the discharge quality of the PPME and/or to recycle it (Mänttäri et al., 2006). Among the advanced treatment processes, membrane technology is an attractive alternative to treat PPME. In literature, a limited number of studies have been found dealing with membrane-based treatment of PPME. Jonsson and coworkers (Jonsson et al., 1996) reported that membrane-based treatment is suitable for removal of color from paper mill effluent; however, the composition of the color had an important influence on the membrane performance. A few other researchers also reported that the membrane separation is an appropriate technique for removal of adsorbable organic halogens, chemical oxygen demand (COD), and color from PPME (Afonso and Pinho, 1991; De Pinho et al., 2000; Mänttäri et al., 2006; Zaidi et al., 1992). Pizzichini (Pizzichini et al., 2005) performed experiments using ceramic microfiltration (MF), polymeric MF, ultrafiltration (UF) and reverse osmosis (RO) modules to treat the PPME wastewater with the aim of reusing the treated water in the manufacturing process. Furthermore, Mänttäri (Mänttäri et al., 2006) evaluated the possibilities of polymeric nanofiltration (NF) for the purification of discharge water from the activated sludge process and they reported that NF is an attractive process to purify the paper mill wastewater.

However, the most important limitation that appeared in PPME treatment by membrane processes is membrane fouling, causing a rapid flux decline. Lipophilic extractives are potential foulants for membrane applications, which are abundantly present in PPME (Dal-Cin et al., 1996; Ragona and Hall, 1998; Ramamurthy et al., 1995). Flux decline caused by the irreversible adsorption of these foulants is a major problem for the economic implementation of NF for the purification and recycling of this type of wastewater.

1.2. Olive oil wastewater (OOWW)

The management of OOWW is a very important issue as this is one of the major sources of pollution of the water environment, especially in Mediterranean countries. Similar to the pulp and paper production process, also the olive milling is a water intensive process, causing the OOWW to be the main by-product of the olive oil extraction (Borja et al., 1992; Erguder et al., 2000; Tsonis et al., 1989). Generally, the OOWW is characterized by high concentrations of several organic compounds, such as sugars, organic acids, polyalcohols, lipids, proteins and polyphenolic substances, which make OOWW difficult to treat (Erguder et al., 2000; Gavala et al., 1996; Jaouani et al., 2003; Rozzi and Malpei, 1996).

There are different ways or methods adopted to process or dispose this wastewater. For example, disposal of the OOWW to agricultural soils (Di Giovacchino et al., 2002; López et al., 1996; Riffaldi, 1993; Tamburino et al., 1999), natural evaporation (Cegarra et al., 1996; Fiestas Ros de Ursinos and Padilla, 1992), thermal concentration (Fiestas Ros de Ursinos and Padilla, 1992), treatment with lime (Aktas et al., 2001; Al-Malah et al., 2000) and oxidation (Marques et al., 1997) have been reported. Composting (Bouranis et al., 1995; Fiestas Ros de Ursinos and Padilla, 1992; Marques, 2001) and biological treatment (Ammary, 2005; Borja et al., 1992; Fountoulakis et al., 2002; Marques, 2001) are among the methods that are suggested most for the management of the OOWW. However, the efficiency of the process and the complexity might vary significantly. Importantly, the cost involved for processing the OOWW by these methods is quite high. As a rule, high costs is quite often the main reason for not adopting efficient treatment methods. Therefore, the OOWW treatment by traditional techniques is limited (Cheryan and Rajagopalan, 1998; Paraskeva et al., 2007).

Membrane technology has already been reported as a better alternative for the treatment of the OOWW (Bódalo-Santoyo et al., 2003). It offers a number of benefits such as low energy consumption, no additives required, no phase changes etc. Compared to the traditional techniques for irrigation or even for recycling of the OOWW (Akdemir and Ozer, 2009; Borsani and Ferrando, 1996; Coskun et al., 2010; Paraskeva et al., 2007; Stoller, 2008; Turano et al., 2002). Recently, combined membrane processes for the selective fractionation, recovery and concentration of polyphenols from the OOWW also have been proposed (Paraskeva et al., 2007; Garcia-Castello et al., 2010; Russo, 2007).

However, fouling, not surprisingly, is again cited as the major factor that limits the application of membrane technology for the OOWW treatment (Stoller, 2013). Several approaches to mitigate this problem have been attempted (Belkacem et al., 1995; Cheryan and Rajagopalan, 1998). Nevertheless, membrane fouling still remains one of the main challenges for the implementation of membrane technology in the OOWW treatment.

1.3. Produced water (PW)

Oil/gas is one of the key energy sources worldwide and its production is still indispensable to fulfil the energy demand. While oil is produced, some unfavorable environmental effects occur, e.g. PW is produced during oil production and considered the largest by-product associated with the oil production (Folarin et al., 2013; Siriverdin and Dallbauman, 2004). The PW is a continuous source of contaminants to the ecosystems (Bakke et al., 2013).

On the other hand, the composition of the PW is very complex, having distinctive characteristics due to the presence of dispersed oil, production chemicals, corrosion products, heavy metals, large amounts of organic material, inorganic salts and natural radioactive minerals (Dyke and Bartels, 1990; Farnand and Krug, 1989; Zaidi et al., 1992). In addition, characteristics of the PW usually vary significantly depending on the location of the field, produced hydrocarbon (Bakke et al., 2013) and life of the well (Kose et al., 2012). Therefore, treatment of PW is a growing challenge in all oil producing regions and its management has become a major issue for the public and regulators (Wandera et al., 2011). The large volume of the PW presents not only environmental challenges but also potential opportunities for beneficial reuse, recycling and disposal alternatives (Horner et al., 2011). Thus, it is absolutely necessary to improve innovative technologies for the treatment of the PW, not only to meet the increasingly stringent environmental regulations, but also to improve the economic viability of the processes (Xu and Drewes, 2006) and possibly leading to a new source of water.

In the past few decades, various conventional methods have been developed for the treatment of the PW, including biological (Li et al., 2005, 2010), physical (Bayati et al., 2012) and chemical (Shokrollahzadeh et al., 2012) or a combination of these (Ge et al., 2014; Li et al., 2011; Younker and Walsh, 2014; Zhang and Seeger, 2011). However, these methods are more or less energy and time consuming, suffer from low efficiency and are not effective for treating tiny oil droplets. A number of reports stated that these