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Treatment of WEEE industrial wastewaters: Removal of yttrium and zinc by means of micellar enhanced ultra filtration

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ABSTRACT

In this paper, the efficiency of micellar enhanced ultrafiltration technique (MEUF) was tested for the removal of yttrium and zinc ions from synthetic industrial liquid wastes. UF membranes (monotubular ceramic membranes of 210 kDa and 1 kDa molecular weight cut-off) were used with adding an anionic surfactant, sodium dodecyl sulfate (SDS). A two - level full factorial design was performed in order to evaluate the effect of molecular weight cut-off, sodium dodecyl sulfate concentration and pressure on the permeate flux and rejection yields. It was found that the single factors presented the largest influence on the permeate flux: the membrane pore size and the pressure had positive effect, instead the SDS had negative effect. Regarding the metal rejection yields the main relevant factors were the membrane pore size with a negative effect, followed by the surfactant concentration with a positive effect. The effect of the pressure seemed to be almost negligible, for zinc removal experiments had a positive effect in the interactions with the surfactant and membrane pore size. The results showed that very good removal percentages up to 99% were achieved for both metals under the following conditions: 1 kDa membrane MWCO, in the presence of the surfactant at a concentration above CMC independently of the investigated pressure.

1. Introduction

Wastewaters from industrial uses contain high concentration of heavy and dangerous metals. These residual solutions cannot be discharged into the sewer without an appropriate treatment. The dangerousness of the liquors depends on the type and concentration of the metal ions, linked to the industrial activity from which wastewaters derive. As for example, residual solutions coming from electronic industry and from recycling plants of waste electrical and electronics equipments (WEEE) contain substances such as Tetramethyl Ammonium Hydroxide (TMAH) and heavy metals (*e.g.* zinc, nickel, cobalt, manganese, lead and others), respectively.

The choice of the best treatment of the residual solutions depends on the type, metal concentration and eventually on further reuses of the treated water (Fu and Wang, 2011; Barakat, 2011, Prisciandaro et al., 2016). Adsorption and chemical precipitation are the most widely used techniques to remove heavy metals from wastewaters (Prisciandaro et al., 2006; Karatza et al., 2013; Sen et al., 2015; Ippolito et al., 2016; Wu et al., 2016). Other processes used for the wastewater treatment are membrane processes (mostly ultrafiltration), (Prisciandaro and Mazziotti di Celso, 2010) that are more selective and may represent a reliable alternative to conventional processes for the removal of pollutants from wastewaters (Mazziotti di Celso and Prisciandaro, 2013; Prisciandaro and Mazziotti di Celso, 2016). However, due to the permeability of the membranes, ultrafiltration does not allow the separation of the metal ions from contaminated wastewaters without the use of an element that tends to increase the size of the solutes. Micellar enhanced ultrafiltration (MEUF) is a technique wherein a surfactant is added to the solution in such concentrations as to form micelles on which the pollutants (*e.g.* metals) are adsorbed. Such structures can be held by an ultra-filtration membrane with a suitable cut-off, while only free ions and the monomers pass through the pores. Removal efficiency of the process depends on the characteristics and concentrations of the metal and of the surfactant, the solution pH and ionic strength of the solution and by ultrafiltration operating parameters. However, during membrane wastewater treatment process, due to membrane fouling and concentration polarization, severe flux decline decreases filtration

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efficiency, increases the operation cost, and restricts its sustainable process and further industrial application (Zhang et al., 2012, 2013, 2016).

MEUF has been proven to be an effective separation technique to remove metal ions from wastewater, following a list of scientific works about this scientific fields were reported. Landaburu-Aguirre et al. (2009) studied the removal of zinc from synthetic wastewater by MEUF using anionic surfactant, sodium dodecyl sulfate (SDS). The removal efficiency was greater 99% when the surfactant to metal molar ratio was above 5.

Samper et al. (2009) described the removal of cadmium, copper, nickel, lead and zinc from synthetic aqueous solution in presence of two anionic surfactant, SDS and linear alkylbenzene sulfonate (LAS). The experimental results showed that around 90% of rejection yields were obtained for cadmium, copper, lead and zinc using an LAS and SDS concentration below critical micellar concentration (CMC).

Jung et al. (2008) studied the feasibility of MEUF for the treatment of the washing effluent after soil washing. In the initial effluent, total metal concentration and complexing agent was 10.14 and 9.03 mM respectively and the pH was 6.34. Under optimal conditions (when the concentration ratio of metal and surfactant was 10) the removal efficiency were over 92%.

Other research activities about the removal of heavy metals by MEUF technique were presented and discussed by Fun and Wang (2011) and Mungray et al. (2012).

The MEUF technique was already studied by this research group for the removal of heavy metals such as chromium, nickel and cobalt from wastewaters (Tortora et al., 2016a; 2016b, 2017a; 2017b; Innocenzi et al., 2017a). The results showed that MEUF with an anionic surfactant was able to remove metal ions, with a strong increase of rejection coefficient with respect to UF, reaching the highest removal efficiencies when the surfactant concentration was above its critical micellar concentration (CMC).

More recently Lin et al. (2017) in their paper discussed the removal of heavy metals from mining wastewater. The effect of surfactant to metal (S/M) and pH solution on metal rejection was studied by Montecarlo-based artificial neural network modelling approach. The results showed that an S/M ratio equal to 8.5 and a pH of 8–10 were the optimal conditions for MEUF operation. In these conditions the metal removal yields exceeded 99% for copper, nickel and cobalt.

Generally, the MEUF experiments include the separate study of each factors as a function of time. This method consists of numerous tests; therefore, it makes use of significant amounts of raw materials (water, metal salts, organic substance and surfactants) and time, with an inefficient use of resources and energy. Moreover, this approach cannot evaluate the magnitude of interactions between the investigated factors, losing some important information about the process especially if the aim is to optimize it from an industrial point of view. To this scope, a statistical analysis should be desirable. One of the main advantages of statistical designs is the possibility of finding the optimal combinations of the investigated factors to increase treatment the efficiency.

In this manuscript, the factorial design was conducted in order to reduce the number of the experiments and define the effect of several factors on the removal of metals, such as zinc and yttrium, from synthetic solutions. Other articles were foundthat reports the description of statistical designs applied to MEUF (Badan Ribeiro et al., 2006; Cojocaru et al., 2007; Xiarchos et al., 2008; Landaburu-Aguirre et al., 2009, 2010, 2012; Namaghi and Mousavi, 2016). Badan Ribeiro et al. (2006) applied a full factorial design to investigate the recovery of solvent from soybean oil/hexane miscellas, Landaburu-Aguirre et al. (2009,2010) studied the recovery of zinc and cadmium from synthetic aqueous solutions. The same authors applied the statistical analysis for the recovery of heavy metals from phosphors real rich wastewater by MEUF.

Namaghi and Mousavi (2016) in their paper studied micellar-enhanced ultrafiltration with LAS in order to treat soft drink processing wastewater using factorial experimental design.

In the present work, MEUF technique is presented when applied to the treatment of residual solutions coming from a hydrometallurgical process for the treatment of cathode ray tubes (CRTs). CRTs are classified as WEEE and are extremely dangerous since they contain a lot of metals as zinc, lead, iron and they need to be appropriately collected and disposed in according to European directive of WEEE (Directive 2012/19, European Parliament). As for almost all the electronic wastes, CRTs contain, in addition to base metals, a number of critical materials mainly rare earths (REEs), (European Commission, 2010). Yttrium is the most concentrated REEs with a high percentage in the initial fluorescent powders, followed by zinc. During the recycling process, the most part of the metals (as yttrium) is recovered while a small percentage remains in the residual solutions.

Fig. 1 reports the hydrometallurgical processes for the recovery of metals from WEEEs, in particular from cathode ray tubes (Innocenzi et al., 2013a, 2013b). The process was developed within the European FP7 framework: HydroWEEE (2009–2012) and HydroWEE-DEMO (2012–2016). A part of this project was focused on the recycling of rare earths from other electronic wastes, such as fluorescent powders of lamps (Innocenzi et al., 2014, 2016a, 2016b; Ippolito et al., 2017) and industrial catalysts (Innocenzi et al., 2015; Ferella et al., 2016).

The WEEEs, after collection, sorting and dismantling, need of a pre-treatment that includes disassembly and CRT recycling for example with a diamond cutting technology. The initial materials coming from pre-treatment are the feed of the hydrometallurgical process. The characterization of the metals shows that the main elements are yttrium and zinc with an average concentration of 15%w and 25%w, respectively. These metals are present as oxides and sulphur compounds. The feed of the leaching is the pre-treated materials (after dissolution with sulfuric acid to dissolve rare earths as sulphates). Yttrium is recovered by precipitation using oxalic acid and after calcined to obtain oxides. The metal recovery is near to 70% and the final oxide is rich in yttrium with a grade purity greater than 95%. After precipitation and filtration, the residual solutions cannot be directly discharged but must to be treated to remove and recover residual metal ions. In the case of CRT process the solutions contain a series of metals, for example yttrium and zinc in the following average concentrations: Y = 30 mg/L and Zn = 2 mg/L.

In this paper, the possibility of applying micellar enhanced ultrafiltration technique (MEUF) to the removal of Zn and Y ions from synthetic liquid wastes is verified; the synthetic solution simulates the residual solutions coming from the hydrometallurgical process for the recovery of metals from CRTs. Two monotubular ceramic ultrafiltration (UF) membranes of 1 kDa and 210 kDa MWCO were used with adding an anionic surfactant (SDS). The experiments were conducted in according to a full factorial plan in which the effect of three factors were investigated: pore size of the membrane, surfactant concentration and pressure. The results of the experiments were expressed in terms of permeate flux and rejection coefficient of the metals. The treatments were performed in according to statistical approach in order to evaluate the main effects and interactions of the factors.

The aim of this research activity is to investigate the efficiency of the MEUF for the wastewater coming from the treatments of the recycling of WEEE in order to (a) reduce the amount and the toxicity of the residual solutions; (b) reduce the wastewater treatment costs; (c) re-use the treated water and finally d) develop an integrated recycling process reducing the liquid waste.