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Recovery of valuable metals from electronic scraps by clays and organo-clays: Study on bi-ionic model solutions

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ABSTRACT

The demand of valuable metals, as precious metals and rare earths, is constantly increasing in the global market, as many and different technological applications exploit these materials because of their unique properties. Since natural resources are located just in focused areas, an interesting possibility could be the recovery of metals from Waste Electrical and Electronic Equipment (WEEE). The aim of this work is to evaluate the recovery potentialities of clays and organo-clay based systems towards the metals contained in the solutions of electronic scraps dissolved in strong acid, by preliminary tests on bi-ionic model solutions. Lanthanum has been chosen as representative of the rare earths while copper has been considered since it is by far the most used metal in electric and electronic equipment. The considered sorbents are a montmorillonitic clay and two polyamine based organo-clays. Uptake and release processes have been carried out in order to assess the performances of these solids and to evaluate the uptake and release mechanisms. The results showed that the cationic exchange is the prevailing mechanism in the case of pristine clay, while both coordinating effect due to amino groups and cationic exchange occur in the case of modified clays, respectively accounting for copper and lanthanum uptake. Furthermore the pH was found having a great influence in both the adsorption and desorption phenomena.

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1. Introduction

The approach known as “urban mining” bases on the development of “Best Practices” for collection, transportation, recycling and recovery of precious metals and rare earths from electrical electronic waste (E-waste, or WEEE), so as to transform the waste into an added economic value product and, at the same time, minimizing its environmental impact (Cossu and Williams, 2015).

Currently each EU citizen produces about 17 kg of waste electrical and electronic equipment per year. According to European Union estimates, this value is expected to rise to 24 kg by 2020 (EU, 2012). Unfortunately, nowadays the collection is able to intercept only 25–40% of the generated WEEE (UNU, 2008). These wastes are rich in precious and strategic metals and are characterized by very higher contents than those of natural minerals (Bigum et al., 2012; Cucchiella et al., 2015; Guezennec et al., 2015). For these reasons, the study of an efficient rare earths (REs) and precious metals recovery from WEEE can lead to undeniable

socio-economic and environmental benefits (Akcil et al., 2015; Iannicelli-Zubiani et al., 2012).

The European Union (EU, 2010) pointed out some elements, for which there are no primary sources in the EU territory and which are fundamental to the development of European manufacturing. For what concerns these elements, the suppliers are often located in politically hazardous areas or the reserves are limited, so that these materials can easily create geopolitical and economical tensions and prices instability. All the precious metals (gold, silver and copper), the elements belonging to the platinum group metals (PGMs: platinum, palladium, rhodium, ruthenium, iridium, osmium) and the ones belonging to the group of REs can be found in this group of critical elements (EU, 2010). In all these cases the recovery from worn out or obsolete products is still limited if not completely absent.

Today, different technologies are operating for the recovery of precious metals and REs from WEEE, which, however, or are not resolving, or are energetically expensive, or require too sophisticated or too large equipment to be managed by medium small companies (Jadhav and Hocheng, 2015).

From a technological point of view recycling of end-of-life WEEE and recovering of metals therein contained can be divided into three major steps (Cui and Forsberg, 2003):

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- (1) Disassembly: selectively disassembly, identification of the hazardous or valuable components for special treatment, is an indispensable process in recycling of e-waste.
- (2) Upgrading: using mechanical processing and/or metallurgical processing to up-grade desirable materials content.
- (3) Refining: in the last step, recovered materials are retreated or purified by using chemical processing. This final step can be realized by different metallurgical techniques: pyrometallurgical, biometallurgical and hydrometallurgical.

Among the others, hydrometallurgical method has been reported to be more interesting with respect to pyrometallurgy, having the following advantages:

- (1) Reduced risk of highly toxic and polluting emissions, in particular the ones caused by the presence of halogenated flame retardants (HFR), which can lead to the formation of dioxins and furans (Chen et al., 2015b).
- (2) Efficient separation of metals avoiding purification steps (Cui and Zhang, 2008).
- (3) Low energy requirement by the plants, where it is not necessary a source of organic fuel (Chen et al., 2015b).
- (4) No combustion residue currently sent to final disposal (landfill).
- (5) No dust emission that can be a risk to both the environment and the human health (Tuncuk et al., 2012).

On the other hand, some disadvantages of this process are:

- (1) The large number of process steps.
- (2) The consumption of large amounts of chemicals.
- (3) The generation of large amounts of waste water (Iannicelli-Zubiani et al., 2012).

In particular, some of the reported disadvantages are related to the step of metal ions removal from aqueous solution (Iannicelli-Zubiani et al., 2015b).

Recently, the use of adsorption onto solid sorbents is obtaining more and more attention because of its high recovery efficiency, short extraction time, high enrichment factor, low cost and low consumption of organic solvents over liquid-liquid extraction (Li et al., 2011). In particular, adsorption has become one of the alternative treatments (Rao et al., 2008), being a simple and potentially low cost process. Indeed, technical applicability and cost-effectiveness are the key factors in the selection of the treatment technology (Nurchi and Villaescusa, 2012).

Typical adsorbent materials include: inorganic oxides, activated carbon, porous organic polymers, organosiloxane-bonded silica materials, class-specific sorbents (Huck and Bonn, 2000), immunosorbents, surface-bound macrocyclic ligands (Rahman et al., 2013), restricted access materials (Poole and Poole, 2012) and often the same agents used in liquid-liquid extraction but immobilized on solids. In particular, in this study montmorillonitic clays have been investigated. Indeed, the quoted expandable and exchangeable features of this material, together with low toxicity, good rheology, discrete establishing properties, good acid tolerance and low costs, make these clays to be optimal adsorbing agents which can be even considered for industrial applications (Meunier, 2005). Furthermore clays can be modified with suitable agents in order to improve the adsorption efficiency.

From preliminary analyses on the chemical composition of the leached WEEE scraps, copper resulted to be the most present ion, confirming the literature (Calgaro et al., 2015; Chen et al., 2015a). For this reason in this study three ideal solutions are considered all of them containing a fixed amount of lanthanum ions (19 mM), but copper in different initial concentrations (19, 50,

100 mM). The uptake performances of three different solids were evaluated: the pristine clay STx, and two organo-clays modified with different polyamines. At the end, the question about the release of the adsorbed ions is faced.

The results obtained in adsorption and desorption tests and the discussion of the involved mechanisms can present innovative aspects in metal adsorption since new and cost effective materials, characterized by easy technical applicability, are used. Furthermore, the proposed process valorizes natural resources as clays, using them as solid matrices and modifying them, synthesizing completely new materials, which have never been tested for the proposed application of metals recovery from WEEE.

2. Experimental

2.1. Materials and characterization techniques

On the basis of literature reports (Chegrouche et al., 1997; Spencer et al., 2007), a natural smectite samples was used as sorbents, namely a Ca-montmorillonite (STx-1b which stands for “State of Texas”, STx hereafter) which was provided by the Clay Minerals Society. From the supplier datasheet, the crystal formula is: ${}^{\text{IV}}\text{Si}_{4.0}{}^{\text{VI}}(\text{Al}_{1.21}\text{Fe}_{0.05}{}^{3+}\text{Mg}_{0.36}{}^{2+}\text{Ti}_{0.02}){}^{\text{XII}}(\text{Ca}_{0.14}\text{Na}_{0.02}\text{K}_{0.01})\text{O}_{10}(\text{OH})_2$.

The natural smectite samples were suitably modified by different intercalating agents.

Two different polyamines were used as intercalating compounds (Table 1), characterized by different amino groups number and structure: a N-(methoxy-polyethylene glycol) ethylene diamine (L2) from Bozzetto Group and a linear ethylene-based amines (L6) from Sigma Aldrich, technical grade.

The other reactants used in this study were lanthanum nitrate ($\text{La}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ 99.99%, Sigma Aldrich), copper nitrate ($\text{Cu}(\text{NO}_3)_2 \cdot 2.5\text{H}_2\text{O}$ 98%, Sigma Aldrich), HNO_3 (ACS, Sigma Aldrich), NaOH (ACS, Sigma Aldrich), and deionized water.

All the metal ions concentrations in solutions were measured by Inductively Coupled Plasma - Optical Emission Spectroscopy (ICP-OES) analyses using a Perkin Elmer Optima 2000DV spectrometer. The measurement error is estimated to be within 1%.

X-ray powder diffraction (XRPD) patterns of clays were recorded after each step of the recovery process with a Bruker D8 Advance diffractometer using graphite monochromated $\text{Cu K}\alpha$ radiation; the scan step was $0.02^\circ 2\theta$ and the measurement time was 1 s per step. The XRPD line profile analysis was performed with TOPAS P 2.1 software (Bruker AXS, Karlsruhe, Germany) using a Pearson VII profile function, after background subtraction. The calculated profiles were used for the determination of basal spacing (d_{001}) of the clays.

In the case of organo-clays, Chemical Oxygen Demand (COD) analyses were carried out using a Spectrodirect Lovibond instrument in order to determine the amine content in the modified montmorillonites. In a standard instrumental procedure the unknown sample was oxidized by heating for 120 min at 150°C (ASTM, 2006).

2.2. Clays modification

The preparation of the organo-clays was performed according a procedure developed elsewhere (Iannicelli-Zubiani et al., 2013, 2015c), via an intercalation reaction. In a typical experiment, 2.5 g of clay were mixed, in a jacketed reactor under vigorous stirring, with 50 mL of aqueous polyamine solution (90 mM) for a fixed time of 90 min.

All the experiments were carried out at the controlled temperature of 30°C .

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