Contents lists available at SciVerse ScienceDirect

ELSEVIER

Chemical Engineering and Processing: Process Intensification

journal homepage: www.elsevier.com/locate/cep



A one-step electrochlorination/electroflotation process for the treatment of heavy metals wastewater in presence of EDTA



A. Khelifa^a, S. Aoudj^{a,b,*}, S. Moulay^c, M. De Petris-Wery^d

^a Laboratoire de Génie Chimique, Département de Chimie Industrielle, Faculté des Sciences de L'Ingénieur, Université Saâd Dahlab de Blida, Route de Soumâa, 09000 Blida, Algeria

^b Centre de Recherche en Technologie des Semi-conducteurs pour l'Energétique (CRTSE), 2, Bd Frantz Fanon BP140, Alger – 7 merveilles, 16038 Alger, Algeria ^c Laboratoire de Chimie-Physique Moléculaire et Macromoléculaire, Département de Chimie Industrielle, Faculté des Sciences de L'Ingénieur, Université

Saâd Dahlab de Blida, Route de Soumâa, 09000 Blida, Algeria

^d IUT Département Mesures Physiques d'Orsay, Plateau du Moulon, 91400 Orsay, France

ARTICLE INFO

Article history: Received 31 December 2012 Received in revised form 24 April 2013 Accepted 25 April 2013 Available online 3 May 2013

Keywords: Metal finishing industry Heavy metals EDTA Ti/RuO₂ anode Electroflotation Electrochlorination

ABSTRACT

This study demonstrated the feasibility of simultaneous removal of heavy metals and EDTA in an electrolytic undivided cell equipped with Ti/RuO₂ as anode and stainless steel as cathode. In absence of EDTA, results show that nickel and copper removal by EF process is pH sensitive. In addition, nickel and copper may be substantially removed by EF. Removal efficiencies were 99.6% and 97%, respectively. In presence of EDTA, the metal removal by the EF process was inhibited. The inhibition rate was found to be dependent on EDTA/metal molar ratio. A one-step process, involving the combination of two techniques electrochlorination (EC) and electroflotation (EF), was set thanks to chloride addition. In situ generated active chlorine allowed the decomplexation of M-EDTA. Then, free metal ions were removed by precipitating and subsequent floating to the surface by rising electrogenerated bubbles. The obtained results revealed that, with 0.6 EDTA/metal molar ratio, removal efficiencies were 77% and 78% for nickel and EDTA, respectively, in the case of nickel–EDTA solutions. Removal efficiencies were 89% and 96% for copper and EDTA, respectively, in the case of copper–EDTA solutions. Furthermore, heavy metal removal efficiency by the combined process showed to be affected by chloride content and current intensity.

© 2013 Elsevier B.V. All rights reserved.

1. Introduction

Wastewaters from metal finishing industries are hazardous to the environment and deleterious to the public health because they contain unsafe substances in startling levels, such as heavy metals, organic complexing agents, and suspended matters [1–3]. The most common technique for the removal of heavy metals from industrial effluents is the precipitation [3]. The latter technique is, however, unsuited when the examined waste contained chelating organic components such as ethylenediaminetetraacetic acid (EDTA), citrates, and gluconates. As a matter of fact, these substances react as ligands with metallic ions to form stable complexes or chelates

E-mail addresses: khelifaab@hotmail.com (A. Khelifa), aoudjsalah@yahoo.fr (S. Aoudj).

that defy removal, unless the ligands are degraded by oxidation or other phenomena [4]. A conventional process for such effluents consists in applying several techniques in a sequential fashion, as follows: oxidation via a chlorination or an ozonation, neutralization to precipitate the metal, and, finally, removal of the precipitate by solid/liquid separation [2-4]. Bober et al. [5] patented a technique for the treatment of waste which contains EDTA and heavy metals like iron and silver. EDTA solutions are chlorinated to destroy EDTA and thereby increase the biodegradability of the solution. Chlorination can be effected by bubbling chlorine gas or by the use of hypochlorite solution. The heavy metal precipitates as hydroxide and is easily filtered or otherwise removed as a precipitate. However, some pitfalls as to this process should be borne in mind: (1) the oxidation step implies the use of oxidizing agents, which are not always safe, (2) the precipitation generally affords a copious sludge, and (3) the kinetics of the aggregation and the decantation of the precipitates are usually slow, which entail a low removal efficiency of metal [4,6].

On the other hand, the electrochemical processes are potentially viable and widely spread, and are increasingly gaining a foothold in industry due to their multifaceted peculiarities, among which there are: the reducing and oxidizing capacity at the surfaces of

Abbreviations: M, metal; DC, direct current; EF, electroflotation; EC, electrochlorination; EDTA, ethylenediaminetetraacetic acid; DSA, dimensionally stable anode; η , removal efficiency; K_{MY} , formation constant; Y, EDTA anion; Å, Angstrom (10^{-10} m) ; K_h , hydrolysis constant.

^{*} Corresponding author at: Centre de Recherche en Technologie des Semiconducteurs pour l'Energétique (CRTSE), 2, Bd Frantz Fanon BP140, Alger – 7 merveilles, 16038 Alger, Algérie. Tel.: +213 21 279880x172; fax: +213 21 433511.

^{0255-2701/\$ -} see front matter © 2013 Elsevier B.V. All rights reserved. http://dx.doi.org/10.1016/j.cep.2013.04.013