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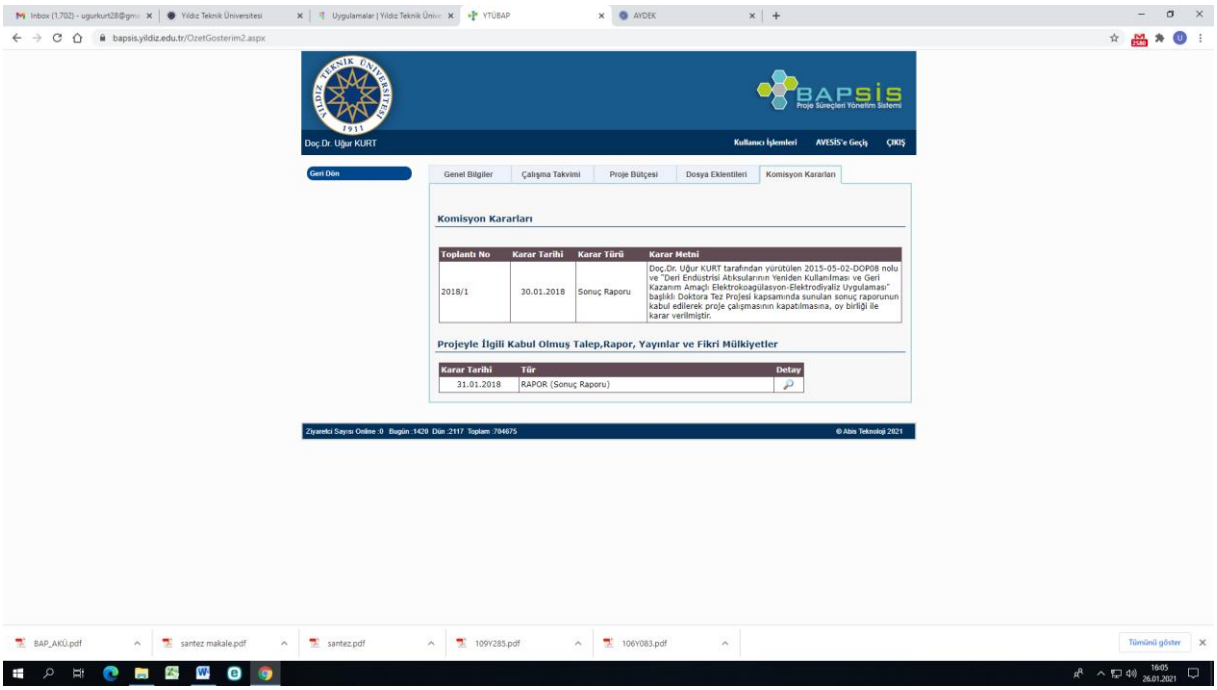
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GRADUATE SCHOOL OF NATURAL AND APPLIED SCIENCES

APPLICATION OF
ELECTROCOAGULATION/ ELECTRODIALYSIS TREATMENT FOR THE RECOVERY AND REUSE TANNERY WASTEWATER

ABDALHADI DEGHLES

PhD. THESIS
DEPARTMENT OF ENVIRONMENTAL ENGINEERING
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İSTANBUL, 2016

REPUBLIC OF TURKEY

YILDIZ TECHNICAL UNIVERSITY

GRADUATE SCHOOL OF NATURAL AND APPLIED SCIENCES

APPLICATION OF ELECTROCOAGULATION/ ELECTRODIALYSIS TREATMENT FOR THE RECOVERY AND REUSE TANNERY WASTEWATER

A thesis submitted by Abdalhadi DEGHLES in partial fulfillment of the requirements for the degree of **DOCTOR OF PHILOSOPHY** is approved by the committee on 04.02.2016 in Department of Environmental Engineering, PhD of Environmental Engineering Program.

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APPLICATION OF ELECTROCOAGULATION/ELECTRODIALYSIS TREATMENT FOR THE RECOVERY AND REUSE TANNERY WASTEWATER

Abdalhadi DEGHLES

Department of Environmental Engineering
PhD. Thesis

Adviser: Assist. Prof. Dr. Ugur KURT

Tannery industry is known widely as one of the highest consumers of water as well as playing a key role in the economic sector of many countries. A few decades ago, water and energy sources were classified as the top two challenges for the twenty-first century. Wastewater treatment has been considered as an efficient tool for managing water and energy sources. Until relatively recently, little has been done of effective technique "zero effluent" to conserve them.

In this context, electrochemical treatment technology has achieved much attention due to its attractive advantages as: simple, fast, reliable, and cost-effective operation for the treatment of wastewater.

In this work, a batch electrocoagulation (EC) technique for tannery wastewater treatment was examined using iron and aluminum electrodes. It involves dissolution of metal from the anode with simultaneous formation of hydroxyl ions, and generation of hydrogen gas at the cathode The effects of operating parameters that include current density, initial pH, and electrolysis time on EC performance were accomplished by measuring of COD, Color, Cr, and NH_3-N as control parameters.

ÖZET

DERİ ENDÜSTRİSİ ATIKSULARINDA MADDE VE ENERJİ GERİ KAZANIMI, SUYUN YENİDEN KULLANILMASINDA ELEKTROKOAGÜLASYON-ELEKTRODİYALİZ ARITMA YÖNTEMİ

Abdalhadi DEGHLES

Çevre Mühendisliği Anabilim Dalı

Doktora Tezi

Tez Danışmanı: Yrd. Doç. Dr. Uğur KURT

Deri Endüstrisi bir çok ülkenin ekonomisinde oynadığı önemli rol kadar, yüksek su kullanımı olan bir sektördür de. Son yıllarda, 21. Yüzyıl için olabilecek en büyük iki sorun su ve enerji kaynaklarındaki sıkıntılar olarak sıralanmaktadır. Atık su arıtımı su ve enerji kaynakları yönetiminde etkin bir araç olarak dikkate alınmaktadır. Bu kaynakların korunması için elzem olan "Sıfır Atık Teknolojis" son zamanlarda biraz biraz kullanılmaya başlanmıştır.

Bu bağlamda, Elektrokimyasal Arıtma Teknolojileri atık su arıtımında kolaylığı, hızlı olması, güvenilirliği ve ekonomik işletme maliyetleri gibi cazip avantajları nedeniyle oldukça başarılı olmaktadır. Hem değerlendirilebilir arıtılmış atık su eldesi, hem de enerji geri kazanımı açısından önem arz etmektedir.

Bu çalışmada, Tabakhane Atıksuyu arıtımı için elektrokoagülasyon (EC) tekniği demir(Fe) ve aluminyum(Al) elektrotlar kullanılarak çalışıldı. EC işlemi eş zamanlı olarak anottan koagülan metalin çözünmesi ve katotta hidroksil iyonları ve hidrojen gazı

CHAPTER 1

INTRODUCTION

1.1 Literature Review

A few decades ago, water and energy sources were classified as the top two challenges for the twenty-first century [1]. Increasing pollution from point and non-point sources such as industry and agriculture, respectively, the ever-increasing population, urbanization and climatic changes, all of these are being leaded to decline water quality and quantity [2, 3]. Wastewater treatment has been considered as an efficient tool for managing water and energy sources. Until relatively recently, little has been done of effective technique "zero effluent" to conserve them.

Billions of liters of industrial wastewater are produced every day with large quantity of aqueous waste [4]. Leather and environment can be described as two sides of the same coin due to the fact that potential wastes from meat industry (i.e. skins and/or hides) are used in the leather industry via tanning process [5]. Overall, tannery industry is known widely as one of the highest consumers of water as well as playing a key role in the economic sector of many countries. The global leather industry produces about 18 billion square feet of leather yearly with an estimated value of about $40 billion [6].

Besides, leather industry is described as a hard process which involves in serious operations can be given as: beam house operation, tanyard operation, post-tanning operation and finishing operation [7]. At each operation several of chemical materials are used such as acids, alkalis, chromium salts, tannins, solvents, sulphides, dyes and auxiliaries. Ultimately, they produce large amounts of potentially toxic wastewaters containing both organic and inorganic compounds [8, 9].

Tanneries, ultimately, generate wastewater in the range of 30–35 L Kg^{-1} skin/hides process with variable pH, high concentration of suspended solid, BOD, chromium,

CHAPTER 3

MATERIALS AND METHODS

Based on literature reviews, so far no other studies have taken into account the effective factors on EC treatment of tannery wastewater using the Taguchi method. And also so far no other studies have taken into account the aspect of hydrogen recovery from the electrocoagulation process. Moreover Until relatively recently, little has been done of effective technique "zero effluent" to conserve water and energy. The idea of ''zero effluent'' may be linked to a hybrid process i.e EC- ED process. This chapter has been outlined the wastewater sources and characteristics and the experimental set up and procedure as well as the analytical procedure.

3.1 Wastewater sources and characteristics

The samples analyzed in this work were collected from different outflow wastewaters of the Organized Tannery Industrial Region (OTIR) which is located in the Tuzla quarter of Istanbul, Turkey. With regard to OTIR, the treatment plant receives wastewater from 100 small tannery plants based on chrome and vegetables tanning. Generally, it has four treatment steps that can be given as: equalization, settling, aerobic activated sludge, and physiochemical treatment by chemical coagulation. The samples were put into PE containers, transported to laboratory in one hour, mixed very well, and conserved at 4°C during the experiments. The composition of the wastewater sample is presented in Table 3.1. The sample was collected in December 2014.

CHAPTER 4

RESULTS AND DISCUSSION

In this work, the electrochemical techniques for tannery wastewater treatment have been conducted. The elctrocoagulation process was applied as a batch mode and continuous mode reactors using iron and aluminum electrodes. The effects of operating parameters that include current density, initial pH, and electrolysis time on EC performance were accomplished as a batch mode reactor. Taguchi method was carried out in order to design the experiments and to optimize the experimental results. L_{25} orthogonal array (OA, three factors in five levels), signal-to-noise (S/N) ratio (the larger-the-better), and analysis of variance were applied to find the optimum levels and relative magnitude of the effects of parameters.

And also the performance of a continuous electrocoagulation (EC) process was investigated. The effects of the operating parameters, such as current density, pH of solution and inlet flow rate, on pollutants removal efficiency, sludge production and recovery of hydrogen gas were investigated in order to optimize process performance. At optimum operating conditions that were determined in continuous mode reactor, the treated tannery effluents were imposed to electrodialysis process to achieve enhanced treatment results. The results and discussion for these processes can be emphasized as below:

4.1 Batch mode EC reactor

4.1.1 Determination of optimum parametric levels

The collected data were analyzed using Minitab "16 version" statistical software for the estimation of the effect of each parameter on the optimization criteria. Experimental

Conclusion and recommendations

In this work, Taguchi design experiment (L$_{16}$) was carried out to optimize the effective parameters on the removal efficiency of tannery wastewater pollutants by EC process. The larger-the-better S/N ratio was used to analyze the result experiments. A confirmation experiment within the optimal process parameters was conducted to assess the usefulness of the Taguchi optimization design technique. In view of our results, the conclusion can be given as below:

- For iron electrode: current density was the most significant parameter on COD, total chrome, color, and its percentage value was 61.6, 42.4, and 32.5%, respectively. Together with, the second important factor for COD, total chrome and color was electrolysis time and its percentage contribution value was 29, 22.6, and 22%, respectively. Finally, the slightest factor was the initial pH. According to the result statistical analysis, for COD removal, the optimum condition was achieved as: current density 50 mA/cm^2, initial pH 7, and electrolysis time 25 min, for total chrome removal: current density 30 mA/cm^2, initial pH 8, and electrolysis time 25 min, and for color removal: current density 5 mA/cm^2, initial pH 7, and electrolysis time 25 min. In addition, the operating costs for both of COD and color removals were computed as 0.88 $/m^3, besides, for chrome removal was 1.70 $/m^3.
- For aluminum electrode: the initial pH was significant parameter on COD and its percentage contribution value was 45.1%, hence both current density and initial pH have the same effect on total chrome and their percentage values were 31.4% and 31.4% respectively. With regard to color, current density was the most significant and its percentage contribution value was 38%. The second important parameter on COD was the current density with percentage value 31.8%. On the contrary, for color the second important factor was electrolysis time and its percentage value was 27.6%, and also it can be concluded that the color is independent on the change of initial pH and its percentage value was 19.6%. Electrolysis time for COD and total chrome was the slightest parameter with the low percentage contribution value 10% As said by the results statistical analysis, the best of optimum condition for COD, total chrome and color removal was current density 50 mA/cm^2, initial pH 7, and electrolysis time 15 min.

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