



# Comparative study of electrochemical wastewater treatment processes for bilge water as oily wastewater: A kinetic approach



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## ARTICLE INFO

### Article history:

Received 23 September 2014  
Received in revised form 30 March 2015  
Accepted 4 April 2015  
Available online 6 April 2015

### Keywords:

Bilge water  
Oily water  
Electro-fenton  
Electrocoagulation  
Electroflotation  
Pseudo-first-order  
Pseudo-second-order

## ABSTRACT

In this study treatability of bilge water was investigated by electrochemical processes: electrocoagulation/electroflotation process and also the electro-fenton process as an indirect electrooxidation process. As the seawater is mixed into bilge water, the chloride content and the conductivity of bilge water are quite high, which is a great advantage in terms of electrochemical wastewater treatment. Treatability of bilge water by electrocoagulation/electroflotation process is investigated by using aluminum and iron electrodes in accordance with pH, time, temperature and current density. When the aluminum and iron electrodes are compared in electrocoagulation–electroflotation process, the results clearly showed that the aluminum electrodes were more efficient and as well as in a short period of time as 10 min. Chemical oxygen demand and oil–grease removal values obtained as 64.8% and 57% from Al and 36.2% and 12.5% from Fe, respectively. On the other hand by electro-fenton process, it was seen that higher removal efficiencies were obtained in comparison to the electrocoagulation method, with the COD and oil–grease removal being equal to 71% and 69%, respectively. The discharge standards specified by MARPOL were obtained by the electro-fenton process. When processes are analyzed in economically way, the electricity cost of electrocoagulation process by aluminum, iron and electro-fenton process was 0.54 \$/m<sup>3</sup>, 0.53 \$/m<sup>3</sup>, 0.51 \$/m<sup>3</sup>, respectively. In addition, pseudo-second-order kinetic model was fitted for both COD and Oil&Grease removal in electrocoagulation process.

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## 1. Introduction

Maritime transportation has recently gained great importance due to the increasing demand for energy and raw material worldwide. The international conventions regarding the prevention of marine pollution led by the ships first appeared in the mid-20th century. The first international convention is the International Convention on Preventing Sea Pollution caused by Oil and Petroleum-derived products (OILPOL, 1954). However, due to maritime accidents, it was soon understood that these measures were not sufficient, and the International Convention on Preventing Pollution from Ships (MARPOL, 73/78) was introduced in 1973 by the International Maritime Organization (IMO). Today, the sea pollution problem has become an increasingly important issue not only for coastal countries, but also for all societies. Ships discharge their wastes (bilge waters, ballast waters, domestic waters, sludge, slop, etc...) randomly into the seas, hence causing pollution.

Bilge waters are of great importance among all liquid wastes including petroleum and petroleum-derived products. The bilge waters that are stored at the bottom of the ships are greasy wastewaters, including a number of pollutants with various characteristics. The oil and grease leaking from the engine room are also stored in the bilge, which in turn increases the oil concentrations of bilge waters. Generally, bilge waters can be briefly defined as saline and greasy wastewaters. The oil and grease are mixed with the sea water, which affects many marine species. The vulnerability of plankton, which are a basic food source in the seas, to oil and grease based pollutions have been experimentally proven [1]. When plankton are harmed by oil and grease, the adverse effects will take hold of all the other links in the food chain, and eventually will affect all marine species.

Classical methods widely used for the aim of the treatment of bilge waters are chemical or physical processes such as flotation, separation by centrifuge, filtration, and coagulation. Because the major part of the oil in bilge waters is emulsified, the physical methods may fail to satisfy the targeted treatment levels. Caplan et al. [2] and Woytowich et al. [3] both stated that they failed to remove emulsified oil droplets smaller than 20 μm by means of conventional oil/water separation systems. For bilge water

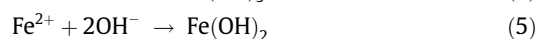
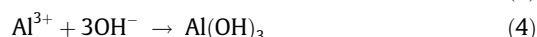
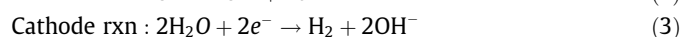
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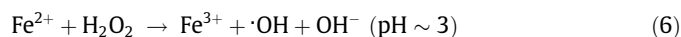
treatment there are also a limited number of studies in which ultrafiltration [4], UF/membrane distillation [5,6] air-stripping and photocatalysis [7], air flotation [8], electrocoagulation [3] methods are applied.

The most rapidly developing and most commonly used processes that are considered to be used in the future among advanced wastewater treatment processes is the electrochemical wastewater treatment process. Besides electrochemical treatment of domestic wastewater is applied successfully, it is proved to be effective in treatment of phenol, aniline, olive oil, cyanide and industrial wastewaters from leather tanning, textile, paint industry [9–17].

Electrochemical processes include electrocoagulation, electroflotation, and direct and indirect electro-oxidation processes. In electrocoagulation process anode electrodes (aluminum and iron electrodes) dissolves as  $\text{Al}^{3+}$  and  $\text{Fe}^{2+}$  ions as coagulant, which form metal hydroxides with high adsorption capacity in water. Electrical charged metal ions can form high-gravity flocks by binding emulsified materials, suspended solids and colloidal materials:



In addition,  $\text{H}_2$  gas produced in cathode generates large surface areas for the adsorption of flocks and precipitates, and removes them by floating (electroflotation process). Generally in electrochemical oxidation process, the desired oxidation is provided by  $\text{H}_2\text{O}_2$  and  $\cdot\text{OH}$  radicals formed via produced gases ( $\text{O}_2$  and  $\text{H}_2$ ) by insoluble electrodes (Ti, Ru, Pt, BDD, etc.). Electrooxidation processes divided to direct electrooxidation and indirect electrooxidation. In direct electrooxidation,  $\cdot\text{OH}$  radicals are formed on electrode surface (Ti, Pt, BDD, etc.), but in indirect electrooxidation process (electrofenton),  $\cdot\text{OH}$  radicals are formed when oxidative chemical ( $\text{H}_2\text{O}_2$  etc.) is added to system:



There are a limited number of studies regarding the treatment of bilge waters through electrochemical processes. The studies focusing on the wastewaters with oil–grease content showed that oil and grease in these wastewaters are removed by absorption into metal-hydroxides formed as a result of the electrocoagulation, or with electroflotation by means of gases produced as a result of electrocoagulation. For the electrocoagulation-based treatment of such wastewaters, aluminum and iron electrodes were typically used, and it was shown that aluminum electrodes led to better results in terms of the efficiency of the treatment of wastewaters with a certain oil concentration [18–20]. This study, on the other hand, aims to investigate the efficiency of electrochemical processes for the treatment of bilge waters. It also examined how electrocoagulation, electroflotation, and electro-fenton processes affect the COD and oil–grease removal.

## 2. Materials and method

### 2.1. Wastewater

The bilge wastewater used for this study was supplied by Haydarpaşa Waste Receiving Facilities. The bilge waters taken by tankers are first brought into the balance tank and stored so that oil and water are separated from one another under gravitational forces and with the aid of a heating procedure. Wastewaters taken from the bottom of this tank are brought into the balance reservoir

**Table 1**  
Characteristics of bilge water.

Parameters	Concentration
pH	6.86
Conductivity, mS/cm	31.1
Turbidity, NTU	35.5
TSS, mg/L	111
Chloride, mg/L	11860
COD, mg/L	2247
BOD, mg/L	490
Oil&Grease, mg/L	93

of a chemical treatment unit and are sent to the treatment facility by means of pumps. The bilge waters used in this study for testing purposes were taken from the balance reservoir, just prior to being sent in for chemical treatment. Characteristics of wastewater can be seen in Table 1.

### 2.2. Reactor and operation

The reactor used in electrocoagulation and electrofenton processes was made of Plexiglas and was 60 mm × 60 mm × 170 mm in size (width × breadth × length), while the aluminum and iron electrodes were 50 mm × 200 mm (breadth × length). The distance between the electrodes was 60 mm. The batch system reactor volume for each study was 500 ml. Mixing within the reactor was accomplished by the gasses produced in electroflotation system. The power supply used at the reactor was a DC supply, GW Instek model GPS-3030 DD.

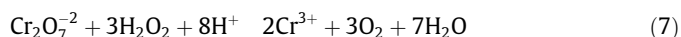
### 2.3. Experimental procedure

Treatability of bilge water by electrocoagulation/electroflotation process was investigated by using aluminum and iron electrodes in accordance with pH, time, temperature and current density. The experiments were carried out with 500 ml bilge water. Initial conditions to reach optimum conditions were fixed between pH: 4.5–10, time: 5–120 min., temp.: 4–60 °C and current density: 2.5–25 mA/cm<sup>2</sup> (1 A current equals to 7.5 mA/cm<sup>2</sup> in 6.5 V).

For electro-Fenton processes as different from the foregoing conditions,  $\text{H}_2\text{O}_2$  dosages between 1250 and 6250 mg/L and pH: 2, 3, 4 and original pH values were arranged. Current density was about 6.8 mA/cm<sup>2</sup> (1 A) and voltage was about 6.2.

After the electrolysis, final pH values were documented, anionic polyelectrolyte with a concentration of 0.2 mg/L was added to the samples in order to facilitate the coagulation process and to accelerate clarity.

In electrofenton process study, hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) should be disintegrated to determine the COD concentrations of samples. It has to be certain that the sample is not containing  $\text{H}_2\text{O}_2$  before COD analysis. Otherwise,  $\text{H}_2\text{O}_2$  will give positive attempt to results as in Eq. (7):



In order to remove  $\text{H}_2\text{O}_2$ , which can react with dichromate in COD analysis, permanganometric method is used [11,21]. Permanganometric method can be used to determine oxidant forms such as  $\text{H}_2\text{O}_2$ ,  $\text{O}_2$ , etc. This method is a well-known method to analysis  $\text{H}_2\text{O}_2$  and it is sometimes called as Ghormley Method [11,21]. For this aim, the pH values of the samples were increased to a value higher than 10 by NaOH solution (0.2 N). After pH adjustment, 1 ml of effluent sample was taken homogeneously in order to analysis  $\text{H}_2\text{O}_2$  by permanganometric method. Addition of NaOH solution is continued until the effluent sample is not

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