

Organics removal in oily bilgewater by electrocoagulation process

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Abstract

This study investigated the treatment of oily bilgewater using an electrocoagulation technique. Electrocoagulation process was evaluated at laboratory scale (1.71 electrolytic cell) and involved utilization of two kinds of electrodes (iron and aluminium) arranged either in bipolar (BP) or monopolar (MP) configuration. Results showed that the best performance was obtained using mild steel MP electrode system operated at a current intensity of 1.5 A, through 60 or 90 min of treatment. Under these conditions, removal yields of $93.0 \pm 3.3\%$ and $95.6 \pm 0.2\%$ were measured for BOD and O&G, respectively, whereas COD_s and COD_l were removed by $61.3 \pm 3.6\%$ and $78.1 \pm 0.1\%$, respectively. Likewise, $99.4 \pm 0.1\%$ of *n*-C₁₀ to *n*-C₅₀ hydrocarbons was removed from oily bilgewater. Electrocoagulation was also efficient for clarification of OBW. Removal yields of $99.8 \pm 0.4\%$ and $98.4 \pm 0.5\%$ have been measured for TSS and turbidity, respectively. Electrocoagulation process operated under the optimal conditions involves a total cost of 0.46 US\$ per cubic meter of treated OBW. This cost only includes energy and electrode consumptions, chemicals, and sludge disposal.

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1. Introduction

Wastewater generated from different industries and household uses have become a serious concern to society. Wastewater treatment and disposal is not only very expensive and time consuming, but also extremely harmful to the environment. Among many contaminants, a particular contaminant of concern is petroleum hydrocarbon present in shipyard wastewater, including the oily wastewater resulting from cleaning of ship bilges and fuel tanks.

Normally, engine oil in boats and ships tends to accumulate in bilges. If no precautions are taken, the oil is pumped overboard along with the bilgewater. This action not only degrades water quality but is also toxic to human and marine life and environment, in general. Moreover, the Clean Water Act (33 CFR 153.305) also prohibits the use of soaps or other dispersing agents to dissipate oil on the water or in the bilge without

the permission of the Coast Guard. Information concerning oily bilgewater (OBW) is difficult to find and it is not possible to know the exact annual load of this water for Canada or Quebec. However, Tomaszewska et al. [1] have reported that oily bilgewater is accumulated at intervals from approximately $0.5\text{--}50\text{ m}^3\text{ day}^{-1}$ per boat and accounts for about 20% of the million tonnes of oily water poured each year into the oceans worldwide.

Oily bilgewater has two distinct components (generally, petroleum hydrocarbons) – a gravity separable phase and an emulsified phase – and both must be addressed to produce clean bilge water [2]. Petroleum hydrocarbons are present as “free oil” (petroleum hydrocarbons that separate from wastewater and float to the liquid surface) and as “emulsified oil” (petroleum hydrocarbons that remain in stable suspension and do not separate from the wastewater). Currently available treatment technologies for OBW effluent include, adsorbents, flocculents, ultrafilters, chemical addition with temperature-enhanced centrifugal devices and polymeric surface modified filtration [3,4]. Each treatment technology encompasses its pros and cons depending on complexity of operations, and capital and

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Nomenclature

Al-BP	aluminium bipolar electrode system
Al-MP	aluminium monopolar electrode system
BOD ₅	biological oxygen demand (mg l ⁻¹)
BP	bipolar electrode system
C ₁₀ –C ₅₀	petroleum hydrocarbon (mg l ⁻¹)
COD _s	soluble chemical oxygen demand (mg l ⁻¹)
COD _t	total chemical oxygen demand (mg l ⁻¹)
Fe-BP	mild steel bipolar electrode system
Fe-MP	mild steel monopolar electrode system
MP	monopolar electrode system
OBW	oily bilgewater
O&G	oil and grease (mg l ⁻¹)
P _t	total phosphorus concentration (mg l ⁻¹)
TOC	total organic carbon (mg l ⁻¹)
TS	total solids (mg l ⁻¹)
TSS	total suspended solids (mg l ⁻¹)
VS	volatile solids (mg l ⁻¹)
VSS	volatile suspended solids (mg l ⁻¹)

operating costs. As said earlier, there is no single off-the-shelf technology that can meet all requirements. Nevertheless, electrocoagulation is a technology which is widely growing over the last few decades as an economical and robust technical solution to numerous environmental pollution problems [5–8]. It has great potential as an excellent treatment technology for oily bilgewaters.

Electrocoagulation directly addresses three main factors that lead to a stable suspension of suspended solids and emulsified oils: ionic charge, droplet or particle size, and droplet or particle density. The process uses an electrical current to dissolve a sacrificial anode and thereby introduces aluminum/iron ions with a positive charge into the wastewater stream [8]. In fact, electrocoagulation requires less quantity of reagents (*in situ* coagulant generation) resulting in volume reduction of sludges when compared to conventional chemical coagulation [9]. These ions are attracted to the very fine negatively charged droplets and particles of the contaminants. The resulting agglomerations increase in size until they are no longer stable in suspension. The gases simultaneously formed by hydrolysis result in very fine bubbles that associate with the coagulated contaminants and buoy them up for removal by flotation.

In this context, the present study investigates electrocoagulation process to remove soluble and insoluble organic matter from OBW studying the effect of the type of electrolytic cell (monopolar and bipolar configuration), type of electrodes (iron or aluminium), current intensity, pH and electrical conductivity. The feasibility of the electrocoagulation process was tested for treating oily bilgewaters and rendering them suitable for disposal into urban sewers. Efficiency of the technology was evaluated in terms of volume of the sludge residues produced, electrical energy consumed, electrodes consumed, and organics removal (BOD₅, COD_s, COD_t, O&G and C₁₀–C₅₀ hydrocarbons). Studies were also carried out on the compaction of metallic sludge

by conditioning electrocoagulated effluent by means of organic polymers.

2. Materials and methods

2.1. Oily bilgewater

Wastewater resulting from the washing of the bilges of boats was procured from an industry having stock reservoirs at Lévis (Que., Canada). Sampling was directly carried out from the tanks which primarily contained water from washings. Samples were collected in polypropylene bottles, shipped cold, and kept at 4 °C, until further use. The effluent was sampled twice and the initial characteristics varied with time (Table 1).

2.2. Electrocoagulation experimentation set-up

Electrocoagulation treatments were carried out in a batch electrolytic cell made of acrylic material {12 cm (wide) × 12 cm (long) × 19 cm (deep)}. The electrode sets (anode and cathode) comprised eight parallel pieces of metal plates (10 cm width × 11 cm high) each, having a surface area of 110 cm², situated 1.5 cm apart and were submerged in the effluent. The electrodes were installed on a perforated acrylic plate placed at 2 cm from the bottom of the cell. Mixing in the cell was achieved by a Teflon-covered stirring bar installed between the perforated plate and the bottom of the cell. For all tests, a working volume of 1.7 l of effluent was used. Between two tests, electrolytic cell (including the electrodes) was cleaned with 5% (v/v) hydrochloric acid solution for at least 15 min and then rubbed with a sponge and rinsed with tap water. The anode and cathode sets were, respectively connected to the positive and negative outlets of a dc power source, Xantrex XFR40-70 (Aca Tmetrix, Mississauga, Canada) with a maximum current rating of 70 A at

Table 1
Characteristics of oily bilgewater

Parameters	Values	Permissive levels ^a
Number of samples	3	–
pH	7.09 ± 0.10	6.0–9.5
Conductivity (μS cm ⁻¹)	668 ± 13	–
Turbidity (NTU)	2210 ± 0	–
BOD ₅ (mg l ⁻¹)	167 ± 49	500
DOC (mg l ⁻¹)	396 ± 0	–
TOC (mg l ⁻¹)	468 ± 0	–
COD _t (mg l ⁻¹)	3400 ± 50	–
COD _s (mg l ⁻¹)	1810 ± 200	–
O&G (mg l ⁻¹)	800 ± 0	150
C ₁₀ –C ₅₀ (mg l ⁻¹)	441 ± 19	20
N-TKN (mg l ⁻¹)	109 ± 10	150
N-NH ₄ (mg l ⁻¹)	85.3 ± 5.2	–
P _t (mg l ⁻¹)	243 ± 5	–
Fe total (mg l ⁻¹)	29.3 ± 1.2	–
Al total (mg l ⁻¹)	5.6 ± 5	–
TSS (mg l ⁻¹)	543 ± 42	600
VSS (mg l ⁻¹)	287 ± 9	–
TS (mg l ⁻¹)	2280 ± 120	–
VS (mg l ⁻¹)	735 ± 213	–

^a Québec City recommendations for effluent discharge in sewer [25].

an open circuit potential of 40 V. Current was held constant for each run with a retention time of 90 min. Different cell arrangements were studied to determine the optimal configuration for organic matter removal from oily bilgewater.

The overall treatment comprised two electrode configurations: monopolar (MP) and bipolar (BP). The dimensional and mounting details of each of the electrodes have been already furnished in an earlier publication [10].

Prior to each test (i.e. before imposing the current intensity), a control sample of oily bilgewater was withdrawn and filtered to determine the initial soluble concentration of organic matter. During an electrolytic run, 10 ml samples were withdrawn at 10 min intervals to follow the changes in COD_s against time. At the end of each test, the pH and the conductivity were measured and the treated effluent was carefully collected, settled for 18 h followed by vacuum filtration (using a Whatman No. 4 membrane filter). A sub-sample (20 ml) was also withdrawn to measure the residual organic concentration. The solid fraction (wet residue) was measured and dried for 24 h to determine total residual solids.

Several assays were performed using two electrolytic units (BP and MP systems) by imposing different current intensities (0.3–3.0 A) and different electrode materials (Al and Fe) through 90 min in view of determining the optimal conditions (reduce cost and increase effectiveness) for treating OBW effluent. During these assays, only the soluble chemical oxygen demand (COD_s) was measured to evaluate the performance of the experimental units in removing organic compounds.

2.3. Sludge compaction study

The OBW sludge produced after electrocoagulation treatment is normally subjected to 18 h sedimentation prior to sampling and filtration. Thus, it is necessary to incorporate a step of chemical flocculation of the sludge to increase compaction as well as decrease the sedimentation time. Various tests were carried out to select an appropriate flocculating polymer/agent and determination of optimal concentrations thereof. Three polymers, namely, Percol 351 (non-ionic), Percol E10 (anionic) and LPM 3135 (cationic) were chosen for the same. In each case, two concentrations (0.010 and 0.025 g l⁻¹) were tested using 100 ml of volume of electrocoagulated effluent. The sedimentation–flotation time varied from 3 to 4 h and the volume occupied by the solid (in ml) was noted at regular intervals.

Once the appropriate dose and type of organic polymer was determined, the optimal conditions of electrocoagulation treatment of OBW (including configuration of electrolytic cell, electrode material, current intensity and treatment time) were repeated in triplicates, followed by flocculation of electrocoagulated effluent and the mixture was then settled for 1 h rather than 18 h. These triplicates assays allowed verifying the efficiency and reproductibility of electrocoagulation process. Soluble and total organics concentrations (COD_s, COD_t, BOD, and O&G), turbidity, total suspended solids (TSS) and total solids (TS) concentrations were measured in each of the replicates.

2.4. Analytical details

A pH-meter (Fisher Acumet model 915) equipped with a Cole-Micrometer electrode was utilized for pH measurements. The conductivity and temperature of the effluent was measured using conductivity meter (Oakton model 510).

COD_s was determined by taking vacuum filtered samples on Whatman 934AH membrane (pore diameter = 1.5 μm) filter, were immediately acidified with 0.2% concentrated sulphuric acid and were preserved at 4 °C, until analyzed. The selected filters prevented passage of TSS as proposed by APHA et al. [11]. The COD_t was measured without filtration according to the method prescribed by Hach (digester HACH, 1995).

The quality of the effluent was also evaluated by measuring turbidity, TS, TSS, VS and VSS. Turbidity (in nephelometric units, NTU) was measured using a turbidimeter Hach 2100AN. Solids were measured utilizing standard methods [11].

The metal analysis was carried out using ICP-AES (Varian, Vista AX model). The dissolved metals were filtered using Whatman 934AH membrane, followed by acidification with nitric acid (5%, w/v) and preserved at 4 °C, until further analysis [11].

Analyses of BOD₅, total oil and grease (O&G) and C₁₀–C₅₀ hydrocarbons was carried out in triplicates by Bodycote Laboratory (Quebec, Que., Canada), accredited by the Department of the Environment of Quebec (standard methods: QC004-92, QC061-97 and QC083-97). The samples were extracted in hexane and analyzed by GC-FID. The concentration of hydrocarbons present in the samples was determined by comparing the total area of group of peaks of *n*-C₁₀ to *n*-C₅₀ with area of the standard curves obtained under similar reaction conditions.

2.5. Process economics

The process economics was evaluated by incorporating the cost of chemicals (industrial grade), electrodes, energy consumption and sludge disposal. The polymer cost was evaluated at 4.31 US\$ kg⁻¹, and the electrolyte (Na₂SO₄) consumption was estimated at 1.53 US\$ kg⁻¹. The sulphuric acid (H₂SO₄) was evaluated at a cost of 0.080 US\$ kg⁻¹. The mild steel and aluminium electrode consumption was estimated at costs of 197 and 1378 US\$ t⁻¹, respectively. The energy consumed was estimated at a cost of 0.05 US\$ kWh⁻¹. The disposal costs (excluding drying costs) for the residual sludge, including transportation and charges for waste disposal, were evaluated at 52 US\$ t⁻¹ of dry residue by assuming the residues to be non-hazardous. Total cost was evaluated as United States dollars spent per cubic meter of treated effluent (US\$ m⁻³).

3. Results and discussion

The oily bilgewater characteristics in the raw effluent and the permitted guidelines for effluent discharge for Quebec City in the urban sewers [25] are presented in Table 1. It was noted that the O&G concentration was five times higher than the permitted concentrations. Likewise, C₁₀–C₅₀ hydrocarbons were twenty times higher than the allowable limits.

Table 2

Experimental conditions, COD_s removal and cost analysis of oily bilgewater using bipolar electrode system (BP)

Parameters	Tests							
	BP-1	BP-2	BP-3	BP-4	BP-5	BP-6	BP-7	BP-8
Electrode type	Fe	Fe	Fe	Fe	Al	Al	Al	Al
Current intensity imposed (A)	0.3	0.5	1.0	1.5	0.3	0.5	1.0	1.5
Mean voltage (V)	15.0	30.0	34.7	39.3	22.1	39.5	39.0	31.7
Treatment time (min)	90	90	90	90	90	90	90	90
Initial pH	7.05	7.18	7.08	7.10	7.25	7.21	6.95	7.01
Final pH	11.8	11.7	12.0	11.8	10.2	9.94	8.74	9.65
Conductivity ($\mu\text{S cm}^{-1}$)	655	653	861	1180	684	671	862	1070
Electrical energy (kWh m^{-3})	3.96	13.2	30.6	52.0	5.84	17.4	34.4	42.0
Electrode consumption (kg m^{-3}) ^a	1.93	3.22	6.43	9.65	0.62	1.04	2.07	3.11
Sludge production (kg m^{-3})	2.24	3.18	6.00	7.12	1.06	1.88	10.7	14.3
Electrolyte Na ₂ SO ₄ (kg m^{-3})	0.00	0.00	1.18	2.94	0.00	0.00	1.18	6.47
Initial COD _s (mg l^{-1})	2800	2620	2730	2640	3460	3580	2690	2890
Residual COD _s (mg l^{-1})	941	659	647	616	1040	1200	711	835
COD _s removal (%)	66.4	74.8	76.3	76.6	69.9	66.5	73.6	71.1
Energy cost ($\text{US\$ m}^{-3}$)	0.20	0.69	1.58	2.69	0.30	0.90	1.78	2.17
Electrolyte cost ($\text{US\$ m}^{-3}$)	0.00	0.00	0.31	0.76	0.00	0.00	0.31	1.68
Cost of electrode consumption ($\text{US\$ m}^{-3}$)	0.38	0.63	1.27	1.90	0.86	1.43	2.85	4.28
Sludge disposal cost ($\text{US\$ m}^{-3}$)	0.12	0.16	0.31	0.37	0.05	0.10	0.55	0.74
Total operating cost ($\text{US\$ m}^{-3}$)	0.70	1.48	3.47	5.72	1.21	2.43	5.49	8.87

^a The electrode consumption was calculated using Faraday's law.

Thus, OBW was required to be treated prior to discharge, and electrocoagulation was one of the techniques which can be exploited to accomplish the objectives. COD_s was chosen as the principal parameter to define effluent treatment as analysis of other components, namely, O&G, and C₁₀–C₅₀ hydrocarbons was laborious.

3.1. Treatment using bipolar electrode configuration

The percentage removal of COD_s from OBW along with the experimental conditions utilized at various current intensities (0.3, 0.5, 1.0 and 1.5 A) for a period of 90 min using either mild steel (Fe) or aluminium (Al) electrodes, is presented in Table 2. The yields of COD_s removal varied between 66% and 77% for Fe electrodes, whereas 67–74% of removal was reported for Al electrodes. From Table 2, it was observed that, for a given current intensity, the depurative efficiency of electrolytic cell (mild steel electrode) was slightly higher than the one recorded using aluminum electrode.

Fig. 1 shows the change in COD_s removal during treatment of OBW using mild steel and aluminium electrodes operated at a current intensity of 1.0 A (assays BP-3 and BP-7), the two assays for which the treatment seemed to be optimal in terms of cost-effectiveness. The COD_s removal decreased rapidly over the first 10 min of the treatment and then increased slightly between 10 and 40 min followed by a steady state until the end of experiment, using either aluminium or mild steel electrodes. Over the first 10 min, more than 50% of COD_s was removed, irrespective of the type of electrodes used. Indeed, during first few minutes of treatment, organic compounds in the form of emulsified oils or suspended colloids were destabilized by neutralization

of the potential energy of repulsion between charged particles owing to Al³⁺ or Fe²⁺ produced by anodic dissolution. Destabilized drops of oil approached one another, flocculated and were separated from the aqueous phase by formation of H₂ gas bubbles generated at the cathode [8]. At the same time, COD_s was removed by cathodic reduction and organic matter was visually seen to be deposited on cathode electrodes. The slight increase in COD_s removal occurring between 10 and 40 min was mainly attributed to co-precipitation of organic matter with metallic hydroxides. It well known that, metallic hydroxides, in the electrocoagulation process are good adsorbents for emulsified and colloidal dispersed oils [12,13]. Moreover, hydrogen

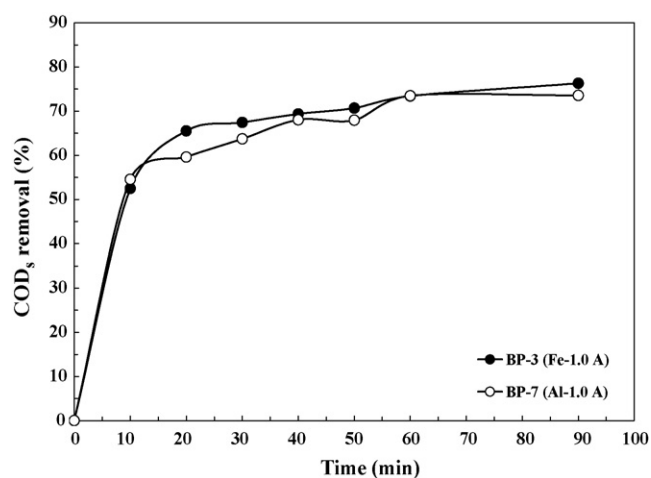


Fig. 1. Variation of COD_s in OBW during electrocoagulation assays using bipolar electrode systems (BP) by means of iron electrodes (BP-3) and aluminium electrodes (BP-7), both operated at the optimal current intensity of 1.0 A.

evolved at the cathode floated the hydroxide flocs with the sorbed organic compounds which caused further clarification of the effluent. Interestingly, only 10 min were required by the cell to produce enough $\text{Al}(\text{OH})_3$ or $\text{Fe}(\text{OH})_2$ and initiate polymerization reaction, inducing the formation of a green precipitate and white gelatinous precipitate using mild steel and aluminium electrodes, respectively. The subsequent formation of polymeric complexes contributed to removal of only a small amount of organic compounds present in OBW by complexation or electrostatic attraction [14]. Similar trend has been also reported in the electrocoagulation treatment of poultry slaughterhouse effluent [10].

Although, BP-3 and BP-7 gave excellent COD_s removals, but the choice of the optimal configuration also depends on the treatment cost and minimal number of treatment steps and other parameters (current intensity imposed, pH variations, electrical conductivity). In fact, cost is a critical parameter with electrocoagulation treatment of such effluents as reported by many authors [15–17]. Thus, BP-5 (BP-Al-0.3A) configuration should be rather selected as an effective treatment for COD_s removal (COD_s removal of 70% as it required no electrolyte addition).

In order to confirm if electrocoagulation contributed to removal of soluble organic impurities, a control test was carried out by subjecting the raw OBW to sedimentation of 18 h. The COD_t concentration of the raw effluent (before sedimentation) was 3420 mg l^{-1} which reduced to 2320 mg l^{-1} after sedimentation, while no change in COD_s was observed. The relatively lower COD_t reduction (approximately 32%) after sedimentation could be due to the settling of a fraction of the organic matter present in colloidal or dispersed form. However, COD_s removal while using a combination of electrocoagulation and sedimentation was much higher (66–77% removal).

The energy consumption with Fe electrodes varied between 3.96 and 52.0 kWh m^{-3} , whereas 5.84 – 42.0 kWh m^{-3} was consumed with Al electrodes. The quantity of metallic residues produced while using the Fe electrodes varied from 2.24 to 7.12 kg m^{-3} , and between 1.06 and 14.3 kg m^{-3} with Al electrodes. The higher metallic residues associated with Al electrodes was due to the higher conducting power of Al electrodes so that anodic dissolution of Al was enhanced. For example, at a given current intensity of 1.0 A , for the same quantity of electrolyte added (1.18 kg m^{-3} of Na_2SO_4), for a relatively similar conductivity (861 and $862 \mu\text{S/cm}$, respectively), 10.7 kg m^{-3} of metallic residues of aluminium hydroxides were reported, whereas 6.00 kg m^{-3} of residues of iron hydroxides were measured (BP-3 and BP-7 tests). It is to be noted that, during the treatment using bipolar configuration, an addition of sodium sulphate (used as supporting electrolyte) was required to be allowed to impose the desired current as the effluent was not sufficiently conductor. For instance, for the same current intensity imposed of 1.5 A , an amount of 2.94 kg m^{-3} of Na_2SO_4 was required for BP-Fe system, compared to 6.47 kg m^{-3} added to the electrolytic cell while using BP-Al system. The relatively high electrolyte required in Al-BP system was probably attributable to the fact that during the assays using BP-Al system, the raw effluent was fewer conductive, compared to the initial effluent while using BP-Fe system. Before adding the electrolyte, the electrical con-

ductivity of the raw effluent varied from 653 to $685 \mu\text{S cm}^{-1}$. After addition of the electrolyte, the electrical conductivity at the end of treatment ranged from 861 to $1180 \mu\text{S cm}^{-1}$ while using BP-Fe system, whereas it varied from 862 to $1070 \mu\text{S cm}^{-1}$ while using BP-Al system.

Meanwhile, in terms of consumption of anodic electrodes calculated using Faraday's law, the quantity of iron consumed (1.93 – 9.65 kg m^{-3}) was three folds higher than with that of aluminium (0.62 – 3.11 kg m^{-3}). However, Faraday's law does not take into account the electrical conductivity of the electrode, and the type of surface (smooth or rough) of the electrodes, important parameters for anodic dissolution. The effect of the electrode surface on anodic dissolution has been well studied for a concentrated oil suspension from machining and drilling operation using electrocoagulation [18].

In addition, the electrolytic reactor equipped with iron (or aluminium) electrodes was able to produce significant quantities of OH^- ions (at cathode) and in fact increased the pH of the effluent. This eventually raised the pH of the effluent from 7.1 to 12.0 by using Fe electrodes and 7.1 – 10.2 in the case of Al electrodes.

It was interesting to compare the total cost (including the consumption of chemicals, energy consumption and the disposal of metallic residues) necessary for the removal of COD_s from OBW. A variable total cost between 0.70 and $5.72 \text{ \$ m}^{-3}$ was observed for the Fe electrodes, compared to an estimate of 1.21 – $8.87 \text{ \$ m}^{-3}$ while using aluminum electrodes. Except for BP-1, BP-2, BP-5 and BP-6 tests (operated without electrolyte addition), other configurations resulted in relatively higher costs, attributed to the consumption of electrolyte and electrodes.

3.2. Treatment using monopolar electrode configuration

As reported earlier, the monopolar electrode configuration for the treatment of oily bilgewater was considered to reduce the consumption of electrical energy, production of metallic residues and to avoid the addition of higher quantity of electrolyte. The COD_s removal and experimental conditions are enumerated in Table 3.

Interestingly, the energy consumption in monopolar configuration was much lower than the bipolar configuration (1.72 – 16.3 kWh m^{-3} instead of 3.96 – 52.0 kWh m^{-3}). For example, at the same current intensity of 1.0 A with Fe electrodes, the energy consumption in bipolar configuration was 30.6 kWh m^{-3} (despite electrolyte addition), in comparison to only 1.72 kWh m^{-3} for monopolar configuration. Likewise, for the Al electrodes, at the same current intensity of 1.0 A , the BP configuration consumed 34.4 kWh m^{-3} (with electrolyte addition), whereas monopolar system consumed only 1.99 kWh m^{-3} . Thus, at the same imposed current intensity of 1.0 A , the energy consumption in bipolar configuration was approximately 17–18 times higher than in the monopolar configuration. The differences could be ascribed to the potential difference between the two electrodes. In fact, for a system functioning in parallel mode (monopolar configuration), the potential difference is equivalent to a simple cell. On the contrary, a system functioning in series (bipolar configuration), the potential dif-

Table 3
Experimental conditions, CODs removal and cost analysis of OBW using monopolar electrode system (MP)

Parameters	Tests							
	MP-1	MP-2	MP-3	MP-4	MP-5	MP-6	MP-7	MP-8
Electrode type	Fe	Fe	Fe	Fe	Al	Al	Al	Al
Current intensity imposed (A)	1.0	1.5	2.0	3.0	1.0	1.5	2.0	3.0
Mean voltage (V)	2.0	3.1	4.6	6.2	2.3	3.1	4.3	5.8
Treatment time (min)	90	90	90	90	90	90	90	90
Initial pH	7.15	7.27	7.00	6.96	7.07	7.20	7.00	7.03
Final pH	11.2	11.7	11.4	11.7	9.96	9.85	9.80	9.58
Conductivity ($\mu\text{S cm}^{-1}$)	655	653	861	1180	684	671	862	1070
Electrical energy (kWh m^{-3})	1.72	4.04	8.12	16.3	1.99	4.04	7.50	15.2
Electrode consumption (kg m^{-3}) ^a	1.61	2.41	3.22	4.83	0.52	0.78	1.04	1.55
Sludge production (kg m^{-3})	2.00	2.78	2.00	4.12	1.63	1.70	4.94	7.18
Electrolyte Na_2SO_4 (kg m^{-3})	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Initial COD_s (mg l^{-1})	3480	2700	3910	2610	2960	2810	2770	2740
Residual COD_s (mg l^{-1})	1270	707	963	591	1200	851	702	709
COD_s removal (%)	63.5	73.8	75.4	77.4	59.5	69.7	74.7	74.1
Energy cost ($\text{US\$ m}^{-3}$)	0.09	0.21	0.42	0.84	0.10	0.21	0.39	0.79
Electrolyte cost ($\text{US\$ m}^{-3}$)	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cost of electrode consumption ($\text{US\$ m}^{-3}$)	0.32	0.47	0.63	0.95	0.71	1.07	1.43	2.14
Sludge disposal cost ($\text{US\$ m}^{-3}$)	0.10	0.14	0.10	0.21	0.08	0.09	0.26	0.37
Total operating cost ($\text{US\$ m}^{-3}$)	0.51	0.83	1.16	2.01	0.90	1.37	2.07	3.30

^a The electrode consumption was calculated using Faraday's law.

ference is higher due to the higher resistance of the electrolytic cells. Thus, at a given current intensity of 1.0 A, the average tension varied between 2.0 and 2.3 V for the MP configuration, and between 34.7 and 39.0 V for the BP configuration.

Despite higher electrical conductivity ($861\text{--}862 \mu\text{S cm}^{-1}$) of the effluent obtained by addition of electrolyte (Na_2SO_4) in BP configuration (at current intensity of 1.0 A), the potential difference was three times higher than that reported for MP configuration without addition of electrolyte (670 and $685 \mu\text{S cm}^{-1}$). The MP configuration produced lower quantities of sludges (2.0 and 1.63 kg m^{-3}) in comparison to BP configuration (6.00 and 10.7 kg m^{-3}) at same current intensity of 1.0 A. The yields of COD_s removal varied between 64% and 77% for Fe electrodes, whereas 60–75% were recorded for Al electrodes. As seen from Tables 2 and 3, using mild steel (Fe) electrodes arranged either in monopolar or bipolar configuration, the yields of COD_s removal increased with the current intensity imposed. As the current intensity increased the anodic dissolution was favoured so that the metallic sludge residues increased and organic pollutants were effectively removed from solution (by co-precipitation or adsorption on metallic sludge). However, using aluminium (Al) electrodes, the yields of COD_s removal did not followed the same trend while increasing the current intensity. This was mainly attributed to the structure of aluminium sludge, which was in a white gelatine-type precipitate and was less stable. By comparison, ferrous sludge formed a stable green colloidal precipitate in aqueous solution, which was more reactive and effective in removing organic pollutants (by precipitation adsorption).

Fig. 2 shows the change in COD_s using mild steel and aluminium electrodes operated at current intensity of 1.5 A (assays

MP-2 and MP-6), the two assays for which the electrocoagulation treatment seemed to be optimal (in term of cost/effective) in mono-polar configuration system. Two main regions could be distinguished in the (COD_s removal, time) plane. In fact, from 0 to 40 min of period of treatment, COD_s removal increased rapidly, then, tended toward a plateau beyond 40 min using either mild steel or aluminium electrodes. The shape of the curves in Fig. 2 was quite similar to the ones reported in Fig. 1, confirming that the mechanism of COD_s removal (neutralization of charged particles, electroflotation, cathodic reduction, and co-precipitation) was similar using either bipolar or mono-polar configuration.

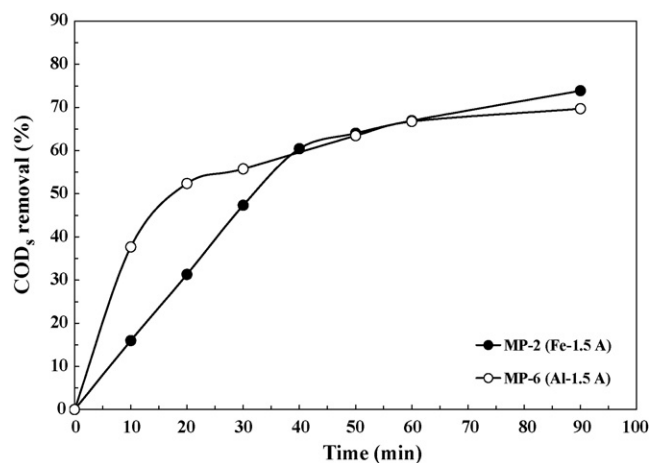


Fig. 2. Variation of COD_s in OBW during electrocoagulation assays using monopolar electrode systems (MP) by means of iron electrodes (MP-2) and aluminium electrodes (MP-6), both operated at the optimal current intensity of 1.5 A.

The optimal conditions (MP-2 and MP-6 tests) in the monopolar configuration resulted in a total operating cost of 0.83 and 1.37 \$ m⁻³ using mild steel and aluminum electrode, respectively. The operating costs were much lower than those selected (3.47 and 5.50 \$ m⁻³, respectively) in BP configuration. In fact, the reduction in cost for MP configuration was obvious due to absence of electrolyte and lower demand for energy as compared to BP configuration. Despite the maximal COD_s removal observed in MP-4 and MP-7 configurations, the MP-2 test was selected as an optimal configuration owing to lower treatment costs with concomitant removal of COD_s (more than 73%).

3.3. Selection of electrolytic cell configuration

When the BP and MP configurations were compared, it was apparent that the MP configuration (MP-2, Table 3) represented a better option (compared to the BP configuration) due to lower operating costs and competitive COD_s removal (more than 73%). However, in large scale application, normally, a BP configuration has an edge over MP configuration due to simple set-up and ease of maintenance [19,20]. Meanwhile, use of MP electrodes with a cheaper electrode material will be a better option as it would tremendously reduce the operating costs as well as provide large range of electrode potential. It was observed from Fig. 1 that the COD_s removal increased with treatment time and also with increase in current intensity imposed (Tables 2 and 3). However, imposition of higher current intensity and increased treatment time incurred treatment costs as the amounts of metallic sludge and the energy consumed increased. In order to further reduce the operating costs, a retention time of 60 min was considered (rather than 90 min) and the MP-2 test (MP-Fe (1.5 A)) was used for sludge compaction study before repeating in triplicates to verify the efficiency and reproducibility of electrocoagulation performance in treating OBW.

3.4. Compaction of electrocoagulation treated OBW sludge—flocculation and sedimentation/flotation

The sludge produced during electrocoagulation treatment of OBW was very fine and difficult to recover. Normally, a sedimentation time of 18 h facilitates the compaction of sludge to a certain degree, but longer sedimentation times at pilot plant level are normally not recommended. Hence, compaction of the treated OBW was carried out and the optimal results using three different flocculating agents for the most preferred electrode configuration, MP-Fe (1.5 A), are presented in Fig. 3. The sedimentation–flotation time varied from 3 to 4 h which accounted for the significant changes in compaction of sludge volume and later (until 18 h), there were no conspicuous variations. When Percol 351 (non-ionic flocculating agent) was used, the OBW showed a distinct solid–liquid separation from the beginning of sedimentation (Figs. 3 and 4). In fact, the clarification was apparent after 30 min and later the sludge compaction did not show much variation, despite increase in concentration of flocculating agent. Thus, the use of 0.010 or 0.025 g l⁻¹ of flocculating agent produced excellent compaction of sludge facilitating recovery.

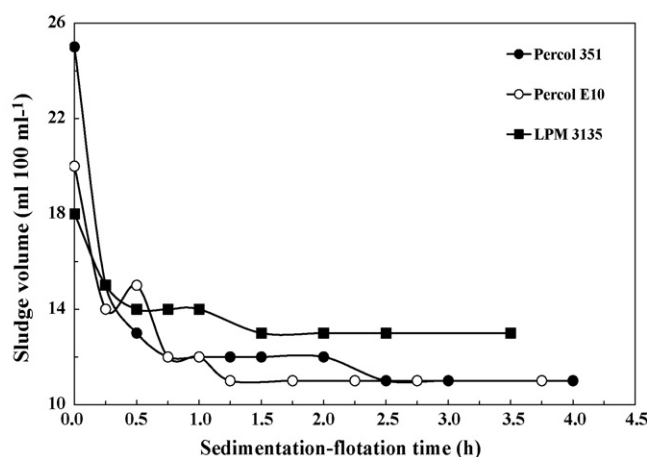


Fig. 3. Compaction of electrocoagulated OBW sludges flocculated with Percol 351, Percol E10 and LPM 3135 with sedimentation time for mild-steel-monopolar (1.5 A) configuration. Polymer concentration = 0.025 g l⁻¹.

Likewise, Percol E10 (anionic flocculating agent) at 0.025 g l⁻¹ resulted in excellent sedimentation of the sludge from the onset of sedimentation. However, in comparison to Percol 351, maximum compaction remained more stable after 1 h of sedimentation. Meanwhile, it was difficult to choose the best concentration as all gave similar results and did not affect the floc constitution.

On the other hand, LPM 3135 (cationic flocculating agent) gave excellent results of all the flocculating agents used. The sludge compaction followed the same course as with Percol E10 (solid–liquid separation, rapid stabilization in less than 30 min, and complete sedimentation of sludges), but the quality of flocs was largely improved. In fact, the flocs were much larger than the other cases, were firmer and very stable, irrespective of the concentrations tested. Thus, LPM 3135 was the most efficient flocculating agent. In fact, LPM 3135 at 0.010 g l⁻¹ allowed an easy recovery of the flocs while reducing the sedimentation time from 18 to 1 h in the optimal condition {MP-Fe (1.5 A)}.

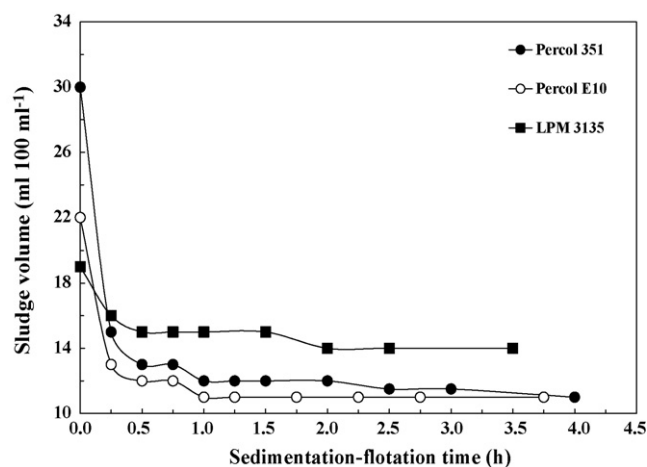


Fig. 4. Compaction of electrocoagulated OBW sludges flocculated with Percol 351, Percol E10 and LPM 3135 with sedimentation time for mild-steel-monopolar (1.5 A) configuration. Polymer concentration = 0.01 g l⁻¹.

In order to determine the efficiency of the system, complementary tests need to be carried out in laboratory in continuous mode which would eventually aid in pilot plant testing of the electrocoagulation process for treatment of OBW. Finally, electrocoagulation process should be tested at the pre-industrial pilot scale for designing treatment facilities so that it may eventually serve as a state-of-the-art technology for treatment of similar wastes.

3.5. Efficiency and reproducibility of the OBW treatment by combining electrocoagulation and flocculation

Electrocoagulation and flocculation combined was repeated in triplicate to verify the efficiency and reproducibility of the process in treating OBW. In particular, before electrocoagulation assay, the initial pH of OBW was adjusted around 4.0 in order to reach a pH below 10 and directly produce an effluent suitable for discharge into an urban sewer. The electrocoagulated effluent was then conditioned by adding the cationic polymer LPM3135 (0.01 g l⁻¹ of polymer) and the mixture was subjected to settling for 1 h rather than 18 h. The results are summarized in Table 4 and it compares different characteristics (COD_s, COD_t, TS, TSS, turbidity, BOD₅, O&G, C₁₀–C₅₀, and others) in raw effluent as well as treated OBW at a treatment time of 60 min.

When BOD₅, O&G and C₁₀–C₅₀ hydrocarbons (three parameters for which the limiting values are recommended by Québec City [25]) were taken into account, electrocoagulation process was effective in reducing the organic compound concentrations

Table 4
Efficiency and reproducibility of the performance of electrocoagulation in treating oily bilgewater using mild steel monopolar electrode system (Fe-MP) (reaction time = 60 min)

Parameters	Characteristics	Removal (%)
Current intensity imposed (A)	1.5	–
Electrical conductivity (μS cm ⁻¹)	652 ± 6	–
Initial pH (pH adjusted)	3.96 ± 0.06	–
Final pH	9.79 ± 0.09	–
Energy consumption (kWh m ⁻³)	2.16 ± 0.09	–
Electrode consumption (kg m ⁻³) ^a	0.92 ± 0.00	–
Sludge production (kg m ⁻³)	1.92 ± 0.16	–
Sulphuric acid consumption (kg m ⁻³)	0.34 ± 0.03	–
Flocculating agent (kg m ⁻³)	0.01 ± 0.00	–
C ₁₀ –C ₅₀ (mg l ⁻¹)	2.80 ± 0.39	99.4 ± 0.1
O&G (mg l ⁻¹)	35.3 ± 1.2	95.6 ± 0.2
BOD ₅ (mg l ⁻¹)	11.7 ± 2.1	93.0 ± 3.3
COD _s (mg l ⁻¹)	697 ± 14	61.3 ± 3.6
COD _t (mg l ⁻¹)	744 ± 9	78.1 ± 0.1
TSS (mg l ⁻¹)	1.33 ± 2.31	99.8 ± 0.4
TS (mg l ⁻¹)	1430 ± 609	37.3 ± 5.8
Turbidity (NTU)	35.4 ± 12.0	98.4 ± 0.5
Electrical energy cost (\$US m ⁻³)	0.11 ± 0.01	–
Electrode consumption cost (\$US m ⁻³)	0.18 ± 0.01	–
Flocculating agent cost (\$US m ⁻³)	0.04 ± 0.00	–
Sulphuric acid cost (\$US m ⁻³)	0.03 ± 0.00	–
Sludge disposal cost (\$US m ⁻³)	0.10 ± 0.01	–
Total operating cost (\$US m ⁻³)	0.46 ± 0.01	–

^a The electrode consumption was calculated using Faraday's law.

below the allowable limits (500 mg BOD₅ l⁻¹, 150 mg O&G l⁻¹ and 20 mg C₁₀–C₅₀ l⁻¹). However, if the electrocoagulation treated OBW has to be rejected directly into the sea (with on-site treatment), it must undergo a secondary treatment to eliminate the residual oil concentration. Moreover, according to 1973 regulation, concentration of oil in the treated effluents to be disposed into the marine environment should not exceed 15 mg l⁻¹ and if the oil concentration is higher, the effluent must be retained on board or else transported to a treatment plant [21].

The residual BOD₅ and O&G concentrations reported at the end of the treatment were 35.3 ± 1.2 and 11.7 ± 2.1 mg l⁻¹, respectively, compared to 167 ± 49 mg BOD₅ l⁻¹ and 800 ± 0 mg O&G l⁻¹ present in the raw effluent. The O&G removal was related to the hydrophobic capacity of O&G, as they have an excellent affinity with the H₂ bubbles released on the cathode. Further, the gas (H₂) bubbles stick to O&G resulting in a (O&G)–H₂ complex which accumulates on the liquid surface as foam and can be easily skimmed. Additionally, there is the phenomenon of adsorption on the surface of metal hydroxides [15]. On the other hand, COD_s and COD_t removal was 61.3% and 78%. Likewise, TSS removal was very high at 98% with a comparatively lower TS removal of 37.3% (attributed to lower removal of dissolved solids). After the treatment, turbidity and TSS reduction averaged 98% and 99%, respectively; the initial green colour disappeared and the effluent became more clear and transparent. Turbidity was effectively removed from OBW owing to aggregating colloidal particles by means of Fe²⁺ and Fe³⁺ produced during electrolysis, which reactants react with colloids to form settleable flocs. Ferrous ions were produced by anodic dissolution whereas ferric ions result from chemical oxidation of ferrous ions in the presence of dissolved oxygen. It well-known that, as the valence of the coagulant increase, the depurative efficiency increase, Fe³⁺ being much more effective in coagulating pollutant than Fe²⁺. Another interesting characteristic of electrocoagulation results from its capability of setting smallest colloidal particles in motion owing to the electric field induced by the potential difference, resulting in an increase in probability of aggregating dispersed or colloids particles [26]. By comparison, suspended solids were mainly removed by adsorption on metallic hydroxides and more importantly by flotation. In fact, during electrolysis the hydrogen gas bubbles attached to the solid particles, and the solids were found to be moving up to the surface with bubbles [8].

It is worth noting that during electrocoagulation treatment, some fine particles of hydroxides re-enter the suspension, which can slightly affect the TS and TSS concentrations (for example, increase in turbidity).

Meanwhile, C₁₀–C₅₀ hydrocarbons removal was very high (99%) which is an indicator of petroleum hydrocarbons which form an important component of OBW. In this experiment, colloidal particles in the OBW trapped hydrocarbons and water. Indeed, the OBW contained large amount of emulsified oils (or emulsified hydrocarbons) that initially formed very stable emulsions with water. During the electrolysis, ferrous ions produced by anodic dissolution reacted vigorously with the emulsified hydrocarbons, leading to phase inversion and breaking of the

emulsion. Stabilized drops of hydrocarbon were then readily sorbed and adhered to the surface of the metallic hydroxides particles [22]. Likewise, hydrogen gas bubbles (H_2) evolved at the cathode electrodes floated the hydroxide flocs together with the hydrocarbons adsorbed on the surface of hydroxides. The two processes (neutralization of charges particles and electroflotation) give a synergistic effect and contribute to efficient removal of hydrocarbons [12,13].

On the contrary, various processes, namely, ultrafiltration/reverse osmosis (UF/RO), photocatalytic processes, wet air oxidation and biological treatment investigated for the possibility of removal of petroleum hydrocarbons from oily bilge water treatment yielded comparatively lower removals [1,4,23,24]. However, severe conditions associated with wet air oxidation mean high installation and operating costs, as well as serious corrosion problems associated, and make it inappropriate as an on board facility. Likewise, many problems related to the long-term efficacy of the UF/RO membranes and issues related to the proper sizing of channels (lumens) within membrane modules remain due to the presence of used oil, detergent and salts in oily bilgewater. Furthermore, bioremediation techniques are slower [24]. In this case, the electrocoagulation shows more promise for treatment of OBW as it is regarded as a flexible, compact and fairly less expensive technology.

Finally, the treatment costs comprising electrical energy, acid requirements of initial adjustment of pH of raw OBW, electrode consumption, flocculating agent and sludge disposal cost were much lower for the Fe-MP (1.5 A) configuration at costs of $0.46 \text{ US\$ m}^{-3}$.

However, an in-depth economical study needs to be carried out to demonstrate the economical advantage of electrocoagulation application (including energy costs, metallic residues disposal costs and the costs required to build and operate the electrochemical reactor) over conventional chemical precipitation.

4. Conclusion

The study demonstrated the efficiency of electrocoagulation process in the treatment of oily bilgewater charged with organic matter. The application of electrocoagulation treatment using iron/mild steel electrodes in monopolar configuration (1.5 A) followed by flocculation of treated effluent was the most efficient mode. Under these conditions, the performances of the system was analysed (by comparison with the raw effluent) and the mean BOD_5 removal was $93.3 \pm 3.3\%$, the mean of O&G removal was $95.6 \pm 0.2\%$, whereas C_{10} – C_{50} has a mean removal of $99.4 \pm 0.1\%$. It is to be noted that BOD_5 , O&G and C_{10} – C_{50} were the three organic parameters for which the limit values were recommended by Québec City for effluent discharge. The relatively high yields of organic removal allowed producing an effluent suitable for stream discharge in the sewer, with residual concentrations of $11.7 \text{ mg } BOD_5 \text{ l}^{-1}$, $35.3 \text{ mg O\&G l}^{-1}$ and $2.80 \text{ mg } C_{10}\text{--}C_{50} \text{ l}^{-1}$ below the limit value recommended by Québec City (500, 150 and 20 mg l^{-1} , respectively). Likewise, the yields of COD_s and COD_t removal had mean values of $61.3 \pm 3.6\%$ and $78.1 \pm 0.1\%$, respectively. Another interest-

ing characteristic of electrocoagulation process results from its capability of clarifying the OBW effluent, the initial green-color disappeared and the treated effluent became more transparent. Removal yields of $99.8 \pm 0.4\%$ and $98.4 \pm 0.5\%$ have been measured for TSS and turbidity, respectively. Electrochemical coagulation operated under the optimal conditions involves a total cost of only $0.46 \pm 0.01 \text{ US\$}$ per cubic meter of treated oily bilgewater. This cost includes energy and electrode consumptions, chemicals, and metallic sludge disposal.

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