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Water impact

Oily bilge water is chemically complex and difficult to treat due to emulsion formation. This study provides insights on how electrocoagulation (EC) can be operated more efficiently by looking at the effects of power source (DC vs. AC), current densities, frequencies, and salinity. The findings from this study provide new understanding on saline oily wastewater treatment not only for bilge water but for oil/gas produced water and other similar industries as well.

Oily Bilge Water Treatment Using DC/AC Powered Electrocoagulation

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Abstract

Bilge water is a complex oily wastewater that may contain a variety of contaminants and salts. Electrocoagulation (EC) recently emerged as an efficient process for bilge water treatment, but its energy cost and lifespan are primary challenges. This study compared the oil removal and energy consumption by both DC and AC powered EC units and investigated the effects of salinity for the first time. Different current densities, voltages, and operation modes (constant current and constant voltage) were investigated to understand the electrochemical kinetics for high salinity bilge water. Results showed that both DC and AC power sources satisfied the oil reduction requirement with over 99% oil removal and high current density was found more efficient in oil removal. High salinity (e.g. 35 g/L) could slightly improve demulsification during electrocoagulation by reducing the zeta potential. Different frequencies (0.25, 0.5 and 1 Hz) of AC power were applied to EC units and it was found that low frequency was more efficient for oil removal. The results also showed that constant current AC was more efficient to maintain the same performance with less energy cost increment than DC mode.

Keyword: Bilge water, electrocoagulation, alternating current, oil removal

1. Introduction

Bilge water is a mixture of wastewater, sea water, oil, sludge, metals, and other chemicals, specifically surfactants, that accumulate at the lowest compartment on a ship[1-3]. Because bilge water has high oil content and thus toxicity, the international maritime organization (IMO) published the international convention for the prevention of pollution from Ships (MARPOL) to regulate bilge water discharge and protect the marine environment. The Annex I of MARPOL 73/83 requires that the oil concentration of discharged wastewater cannot exceed 15 ppm [4]. Therefore, appropriate treatment is required to remove both free and emulsified oil before discharge. The technologies commonly used for oil removal include oil-water separation by gravity, biological degradation, coagulation, membrane filtration, and electrochemical methods [5-11]. In bilge water, oil can exist either in form of free oil or as an oil-in-water emulsion. The free oil can be separated easily by gravity, but it is difficult to separate small, stable emulsified oil droplets from water without proper physical and chemical treatment [12, 13]. For bilge water, due to the complex composition and the presence of surfactant, oil can be dispersed well and kept stable for a long time. Although membrane separation such as ultrafiltration (UF) or nanofiltration (NF) is used, membrane fouling due to oil contamination has been a challenge. Previous studies found cake layer formation, pore blocking, oil coalescence, and membrane wetting greatly reduced the performance of membrane filtration [14-17].

Electrocoagulation (EC) recently emerged as an alternative method for bilge water treatment [1, 8, 9, 18]. EC is a versatile process to break oil emulsion and separate oil from liquid via multiple pathways. By applying appropriate power sources, coagulants such as aluminum hydroxide complex can be generated to adsorb fine oil droplets. In oily wastewater treatment, EC has been successfully tested to treat oilfield wastewater, petroleum refinery wastewater, metal processing wastewater, and bilge water[19]. Studies found different factors such as pH, temperature, electrode materials, and reactor configurations could affect EC performance[20, 21]. However, some major issues remain including electrode passivation and the high energy consumption. Recently, alternating current (AC) and pulsed-voltage power source received attention for its high treatment performance, effective electrode protection, and low energy demand. Vasudevan et al. used high frequency AC-powered EC for cadmium removal and found AC-EC consumed 55% less energy than DC-AC for achieving similar removal performance[22]. Lobo et al. designed granular carbon filled EC for produced water treatment, and they found more than 60% energy was saved comparing with DC. Similarly, 48% energy was saved for oily wastewater treatment by using pulsed voltage powered EC equipment [23]. The EC operation can also be integrated with microbial fuel cells (MFCs) with the later serving as the energy source for EC[24]. These previous studies indicate that AC can be effective in reducing bilge water treatment energy consumption, which is believed primarily attributed to reduced electrode passivation. However, few studies looked at how salinity and long-term operation affect such treatment, which is important for practical applications in rough shipboard environment. High conductivity improves current flow, and it is therefore hypothesized to accelerate coagulant formation. The addition of salts can also reduce oil-in-water emulsion stability by reducing zeta potential of oil droplets[25-27]. The increased ion strength can reduce electrostatic repulsion between oil droplets and reduce emulsion stability. However, high conductivity may also lead to accelerated passivation and affect floc formation.

In this study, both DC and AC power sources were investigated for oil removal from bilge water. Both constant current and constant voltage modes were applied in DC and AC operations to understand the impacts on oil removal and energy consumption. The effects of salinity on demulsification was characterized in different conditions. In addition, the effects of long-term operation on oil removal and energy consumption were investigated for both DC and AC operation modes.

2. Materials and methods

2.1. Bilge water preparation

To simulate bilge water, potable water well mixed with 1,000 mg/L of mixed oil, 100 mg/L of mixed detergent and 500 mg/L of particulates (Arizona test dust (ISO 12103-A4/A2), 50% coarse, 50% fine) (Table S1). The mixed oil was composed (by volume) of 50% diesel fuel marine (MIL-PRF-16884N), 25% 2190 TEP steam lube oil (MIL-PRF-17331H) and 25% 9250 diesel lube oil (MIL-PRF-9000H). The mixed detergent was composed (by volume) of 50% Type 1 general purpose detergent (MIL-D-16791G), 25% commercial detergent Tide Ultra (liquid) and 25% degreasing solvent (MIL-PRF-680, Type III)[24]. Based on the characteristics of actual bilge water, 15 g/L sea salt (ASTM D1141-52) was added to make the conductivity at about 25 mS/cm. The sea salt concentration was kept the same unless specified in section 3.2.

2.2 Experimental

Cylindrical reactors were used in batch EC experiments, and each reactor had a diameter of 3 cm and height of 12 cm. For each EC cell, two 2-mm thick aluminum plates (470002-028, VWR, USA) were used as electrodes (both anode and cathode) and placed in parallel to each other with a distance of 3 mm. The effective surface area of each electrode was 14 cm^2, and the effective liquid volume was 40 mL. A potentiostat (PC4/300, Gamry Instruments, NJ) was used to provide DC or AC power source and monitor the current and voltage. In constant current mode, three current densities were used, including 3.6, 7.1 and 10.7 mA/cm^2. The EC cells were also operated in constant voltage mode by applying 1-2 V DC power. To investigate the effects of polarity changing frequency, 1, 0.5, and 0.25 Hz were applied for alternating pulsed current (3.6, 7.1 and 10.7 mA/cm^2) and voltage sources. For alternating pulsed voltage mode, a periodically alternated voltage of 2 V was applied with different frequencies (1, 0.5, and 0.25 Hz), and the oil removal and energy consumption were compared. Each batch test lasted for 5 to 50 min depending on conditions, and for each test the final oil concentration needed to be 15 mg/L or less.

To investigate the effects of long-term operation on oil removal and energy consumption, two sets of EC cells were operated for 24 h-batch in 1-L bilge water solution with the same current density of 3.6 mA/cm^2 by using DC or AC (0.25 Hz) power source. The oil removal and energy consumption of long-term used electrodes were compared with brand new electrodes.

2.3 Analyses and calculations

Solution conductivity was measured using a conductivity meter (HQ440d, Hach, USA). Voltage and current were monitored by a potentiostat. Power and energy were calculated according to equations (1) and (2).

$$\mathrm{P} = \mathrm{U} \times \mathrm{I} \qquad (1)$$

$$\mathrm{E} = \frac{P \times t}{V \times 3.6 \times 10^{6}} \qquad (2)$$

where P is power (W), U is voltage (V), I is current (A), t is time (s), E is energy consumption (kWh/m^3), V is the volume of treated bilge water. The oil concentration was measured by bench-top oil analyzer based on UV fluorescence method (TD-3100, TURNER, USA). All extraction and measurements followed manufacture instruction. Zeta potential of bilge water solution was determined by using Nano Zetasizer (Malvern, ZEN 3690). The surface structure and the elemental composition change of electrodes were analyzed through scanning electron microscopy (SEM) and energy-dispersive spectroscopy (EDS) by using a JEOL JSM-6480 electron microscope[28].

3. Results and discussion

3.1 Oil removal by DC powered electrocoagulation

Fig. 1 shows oil removal profile under either constant voltage or constant current mode using a DC power source. The results indicate that both modes were effective in oil removal. For constant current operation (Fig. 1A), under 10.7 mA/cm^2 power input, 98.8 ± 0.2% oil was removed, and the final oil concentration was 10 ± 2 mg/L after 10 min. In contrast, to achieve a similar performance, the low current density required much longer time. For example, it took 15 and 30 min for 7.1 and 3.6 mA/cm^2 to remove 99.1 ± 0.3% and 98.3 ± 0.4% oil, respectively. The oil concentration by the end of each cycle were 8 ± 2 and 14 ± 2 mg/L, respectively. Apparently, the removal rate was improved as the current density increased. As Table S2 shows, during the first 5-min treatment, 3.6 mA/cm^2 only removed 58% oil with a removal rate of 98.3 mg/L·min. When the current density was increased to 7.1 mA/cm^2, over 90% oil was removed while both the removal efficiency and rate were ~150% higher. However, further increment of current from 7.1 to 10.7 mA/cm^2 only improved about 7% on removal rate. Theoretically, current flow determines the amount coagulants and hydrogen gas generated[19], therefore it is understandable that higher current led to more coagulants generation, which co-precipitated more oil droplets. In the meantime, higher gas generation brought more demulsified droplets up to the surface. However, such relationship is not always linear, as the optimal amount of coagulant and gas generation depends on the amount of oil to be removed, so a matching current density to the oil content will be more energy efficient than high current.

Under constant voltage condition, higher voltage led to faster oil removal (Fig. 1B), but the rates correlated with the applied voltage. This operation is more popular than constant current due to easier control[1, 8, 9, 29]. As observed previously, high current generated from high voltage input benefited oil removal. The voltage applied in this study was much lower than previous studies mainly because the distance between electrodes and size of the electrodes were smaller. This enables higher sensitivity response to voltage change, where 0.1 V increment could make a difference in current density and oil removal. Although the voltage control is more straightforward, constant current could be a safer and more delicate method to control electrochemical reactions.

3.2 Effects of salinity on oil removal

Salinity can fluctuate dramatically in real bilge water, and salt concentration can be as high as sea water (35 g/L) in bilge. In general higher salt concentration leads to high conductivity and therefore lower internal resistance, but the mixed salt content may also affect the stability of oil-in-water emulsion and accelerate passivation[27]. Table S3 shows how different concentrations of sea salt change the zeta potentials of synthetic bilge water. The zeta potential was reduced with salt addition, indicating salinity destabilizes the emulsion. Fig. 2 shows how salt concentration influences oil removal under constant DC current. It demonstrates that under low current density salinity does improve oil removal. For example, under 3.6 mA/cm^2 DC current, oil concentration was reduced from 890± 10 to 10 ± 1 mg/L (98.9% removal) in 20 min when the salt concentration was 35 g/L. In the meantime, under the same current but lower salinity (5 g/L), oil concentration decreased from 890± 10 to 27 ± 3 mg/L, a 96.9% removal. As current density increased to 7.1 mA/cm^2, high salt concentration still showed benefits to oil reduction. Within 15-min operation, oil content reduced by 99.2% in bilge water containing 35 g/L salt, while the removal was only 97.2% in the water with 5 g/L salt. However, when the current density increased further to 10.7 mA/cm^2, the effect of salinity on oil removal became insignificant (Fig. 2C). These results suggest that salinity could help increase oil removal by reducing emulsion stability, but this benefit can also be achieved by increasing current density, though it will increase energy consumption. Such findings can help guide the operation strategies when dealing with actual bilge water with fluctuating salt content.

Although salinity can be beneficial to electrochemical reactions and demulsification, one adverse impact was the increased salt deposition on electrodes due to the formation of metal hydroxides and carbonates, such as $CaCO_3$ and $Mg(OH)_2$ (Fig.3). Table S3 lists the percentage of major elements on the electrodes after 12-hour operation measured by SEM EDS. Aluminum and carbon were major elements for new electrode, but oxygen content increased significantly from 4.46% (new electrode) to 46.9% and 49.2% on used anode and cathode electrodes, respectively. Since little calcium or magnesium was found on used anode, most oxygen containing compounds were believed to be aluminum hydroxide and aluminum oxide. In contrast, the cathode in EC could be passivated due to the deposition of metal hydroxides and carbonates[30-32]. It was found that the concentration of Ca and Mg on the surface of cathode increased to 1.33% and 4.61%, respectively. Even though the cathode electrode was producing hydrogen gas that could break or slow down deposition, the metal deposits was still observed on the electrode surface. On the other side, the dissolution of electrode might facilitate the detachment of deposits. Therefore, the periodical change of polarity could be an effective way to inhibit the formation of both oxide layer on the anode and metal deposits on the cathode.

3.3 Oil removal by AC powered electrocoagulation

DC power has been a common source for EC operation, but electrode passivation caused by impermeable oxide layer during DC operation has been reported to significantly decrease treatment efficiency [19]. To resolve this issue, AC or pulsed voltage powered EC with frequent change of polarity could inhibit the formation of impermeable layer [23, 33, 34]. Fig. 4 shows oil removal in EC by using alternating pulsed current and voltage sources with different frequencies. When constant current was applied, no significant difference was found among three frequencies. At 3.6 mA/cm^2, 99.2 ± 0.1% oil was removed under 0.25 Hz (Fig. 4A), which is slightly higher than 1 Hz (98.8 ± 0.1%, Fig. 4B). As current increased, increased performance was observed in all frequencies. The clear relationship between frequency and current density couldn't be identified in the range of frequencies and current densities tested.

The blue curves (Fig. 4) show oil removal in EC by using periodically alternated voltage mode (+2 V/-2 V with equivalent current density of 3.6 mA/cm^2). The final oil concentration at 1 Hz was 217 ± 55 mg/L whereas it was 83 ± 6 mg/L for 0.25 Hz after 30-min operation. The low frequency showed an oil removal advantage. To provide an electrochemical basis for this trend, the current vs. time profiles of three frequencies were compared as shown in Fig. 5. Since the current generation is directly related to electrochemical reactions, it seemed that the process required a "start-up" period. At high frequency (1 Hz), the polarity changed fast and periodically, and the current generation gradually became stable after a long period (~180s). However, at low frequency (0.25 Hz), the current gradually increased and current was higher than that at high frequency. To investigate an extreme condition, a function generator was used to supply an alternating-pulsed-voltage of 2 V and 60 Hz where little current was observed and no oil removal was detected (data not shown). The phenomena indicated that an excessively high frequency for polarity change might limit electrochemical reactions required for contaminants removal. In addition, such high frequency could cause energy loss because high voltage would be needed to trigger the reactions as well but some of the electrons might move back and forth between electrodes to generate heat. Another issue for alternating voltage was biased current generation caused by unequal electrode reactions. As shown in Fig. 5, the net current value generated from one direction was not always equal to the inversed one. For example, at 0.5 Hz, the positive current was higher than the negative direction, but the negative current was higher than the positive one at 0.25 Hz. The purpose of using alternating current or voltage power is to reduce the impermeable layer formed on the electrode surface by hydrogen gas production, but such shift mentioned above could be caused by unequal passivation/deposition [18]. When the electrode reaction rate from one side is slower due to shifted current production, the oxide compounds and deposited materials could accumulate and cause the irreversible passivation. Therefore, using appropriate alternating pulsed current instead of using voltage is preferred to process the treatment with accurate electrochemical reaction and minimum passivation effects by enforcing the electrode reaction at early time.

3.4 Effect of long-term operation on oil removal and energy consumption

The oil removal and energy consumption were compared in Fig. 6 between new and long-term used electrodes. Both DC- and AC (0.25 Hz)- powered EC cells with new and long-term (24 hours) used electrodes could remove oil effectively (> 98%), but their energy consumptions were different. The new EC cell powered by DC consumed relative low energy of 0.378 kWh/m^3, but the energy consumption increased by 258% (0.977 kWh/m^3) after 24 hours' operation. In contrast, the energy consumption of AC powered EC from new to used were similar, only increasing from 0.787 kWh/m^3 to 0.936 kWh/m^3 or 19%. This demonstrates the benefits of AC as it may save significant amounts of energy during the lifetime of the EC equipment. It should be noted that the energy consumption of AC condition was higher than that of DC power at the beginning of operation in this study, which may be caused by higher voltage at the beginning of every shift of electric field direction. The advantage of AC powered EC became obvious after 24 hours' operation, which showed slight lower energy consumption compared with that of DC powered EC. The low energy use is believed to be due to reduced passivation under AC conditions, as the electrochemical reactions from alternating electrodes limited passivation under this periodical current change. However, previous studies reported that AC powered EC could save ~60% energy compared with DC operation [18, 22]. The main discrepancy is due to the difference of AC power source applied in this study. Alternated constant current AC mode was applied in this study to control the current flow of EC system, while alternating-pulsed-voltage (APV) or high-voltage AC power source for the EC process were used in previous studies. Alternating-pulsed-voltage (APV) or high-voltage AC power source may negate the passivation benefits observed in low frequency AC because of biased current generation. The quick diffusion of hydroxide and metal ions in the bulk solution would be the primary pathway to inhibit the formation of impermeable layer if the APV shifted to pulsed voltage with one direction. Therefore, the alternating-pulsed-current operation may be an alternative to protect electrode and control the reaction at the same time.

4. **Conclusions**

This study demonstrated the treatment process for high-salinity bilge water by using both DC and AC powered electrocoagulation. The low frequency and high current AC power demonstrated high efficiency removal, and alternating pulsed current could control the electrochemical process relatively precisely without current shifting. This balances electrode reactions during polarity change. Salinity was found beneficial to the EC electrochemical reaction by increasing conductivity and decreasing emulsion stability, but ions like calcium and magnesium could form carbonates and hydroxides and scale the electrode surface. The alternating-pulsed current operation showed good oil removal and higher energy efficiency in long term operation so should be considered as a viable approach for EC operation. Further investigations could include operation optimization by reducing electrode spacing, conditioning solution pH, and including heavy metal removal and other functions.

Conflicts of interest

There are no conflicts to declare.

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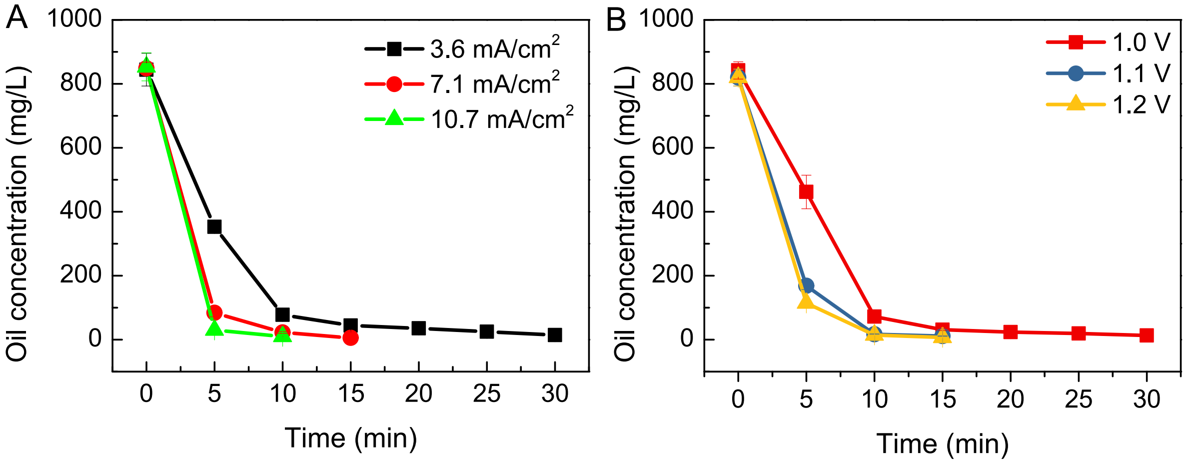

Fig. 1 Bilge water oil removal profile by electrocoagulation powered by either constant DC current (A) or constant DC voltage (B).

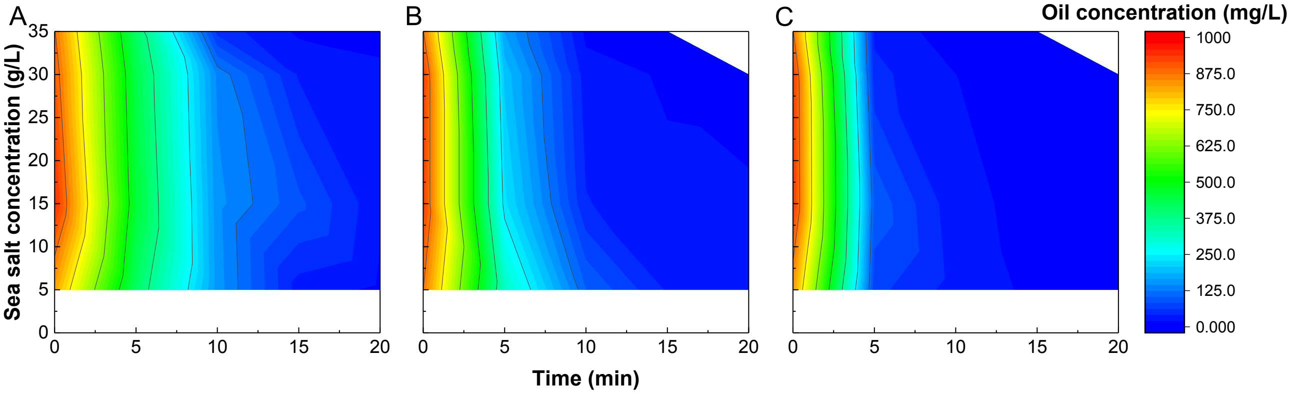

Fig. 2 Oil removal from bilge water under the influence of salt concentration (5-35 g/L) at constant DC current 3.6 mA/cm^2 (A), 7.1 mA/cm^2 (B), or 10.7 mA/cm^2 (C).

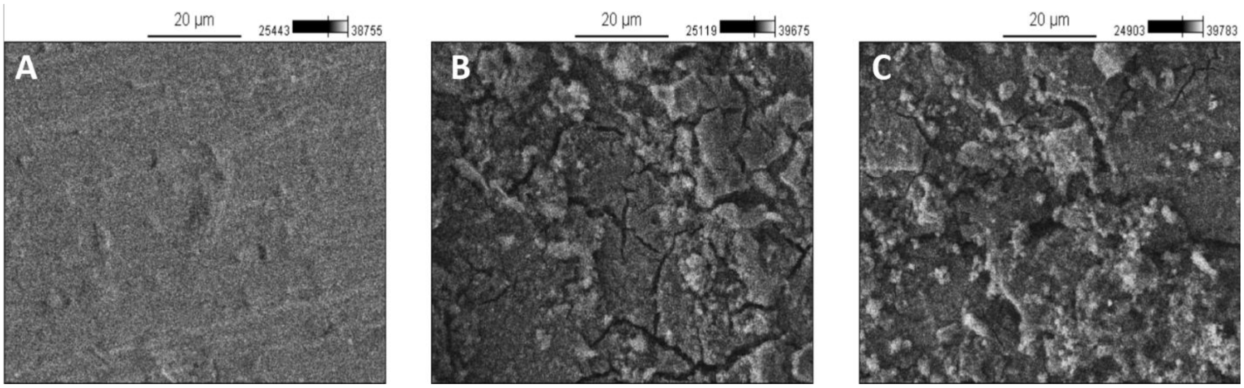

Fig. 3 Morphology changes of aluminum electrodes used in bilge water electrocoagulation: brand new (A), used anode (B), and used cathode (C).

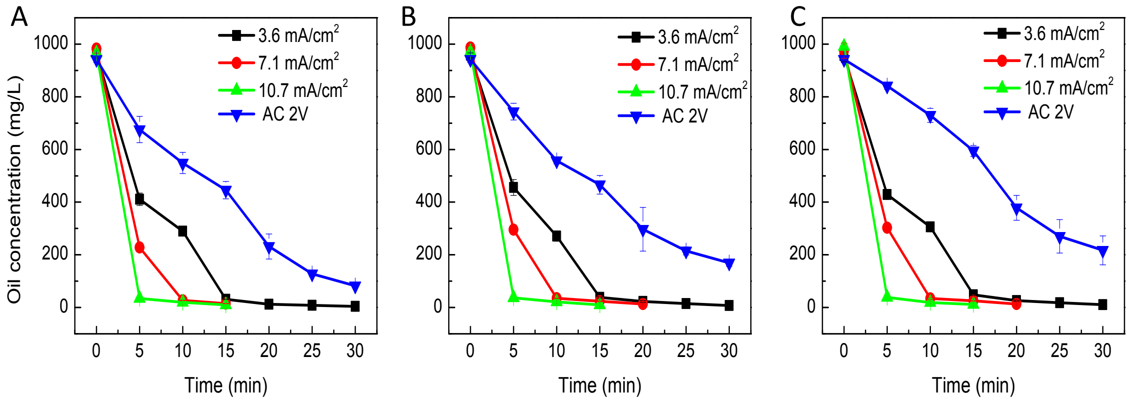

Fig. 4 Bilge water oil removal profile by electrocoagulation powered by AC current (alternated constant current with different current densities 3.6, 7.1 and 10.7 mA/cm^2) and AC voltage (AC 2V: alternated +2 V/-2 V with equivalent current density of 3.6 mA/cm^2) with different frequencies: 0.25 Hz (A), 0.5 Hz (B), and 1 Hz (C).

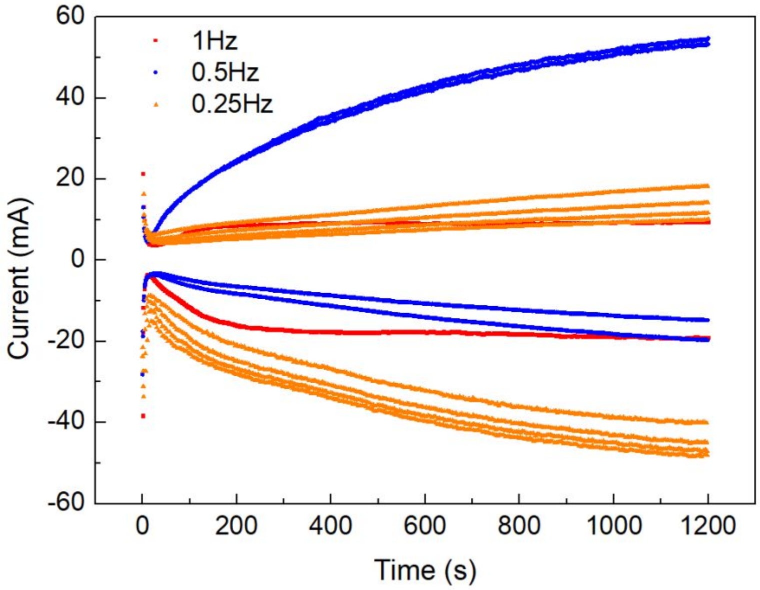

Fig. 5 Current generation from alternating-pulsed-voltage (±2V) electrocoagulation under different frequencies.

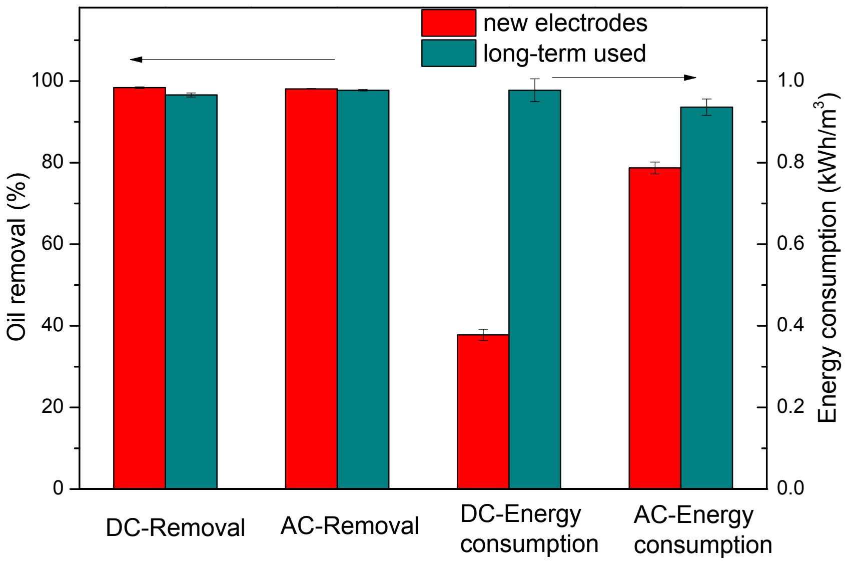

Fig. 6 Comparisons of oil removal and energy consumption under different electrocoagulation modes with new or long-term used electrodes.

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DC and AC powered electrocoagulation were compared for oil removal from bilge water.