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Removal of emulsified oil from polymer-flooding sewage by an integrated apparatus including EC and separation process

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ABSTRACT

Removing emulsified oil from polymer-flooding sewage (PFS) is a tough problem during oil field exploitation as the viscosity and emulsification degree of the oily sewage increased with the addition of polymer in the recovery process. This study aimed to propose an integrated apparatus composed of reaction and enhanced settlement process for small footprint and high settlement efficiency. The effects of current density, flow rate and tilt angles of parallel-plate electrodes (APE) were explored using the integrated apparatus in a continuous system. Furthermore, the comparison between field and laboratory experiments was performed. The results showed that the integrated apparatus had higher removal efficiency. For a given current density of 14 mA cm^{-2}, flow rate of 5 L h^{-1} and APE 80°, the oil and turbidity removal rate of simulated sewage were 96.32% and 97.3%, respectively. Meanwhile, the energy consumption was 2.32 kWh m^{-3}. For on-site sewage, the treatment effects were better and the removal rate of oil and turbidity were 97.2% and 98.2%, respectively. In addition, it was found that the entire EC process was divided into reactive stage and stable stage. The higher the current density, the better purification effect and the shorter time required to reach the stable. As current density increased from 7 to 14 mA cm^{-2}, the stable time decreased from 100 to 40 min while current density increased from 14 to 28 mA cm^{-2}, the stable time changed a little with the removal increased less than 5%. Besides, the results showed that the final removal decreases with the increase of APE, but except APE ɑ which was defined as a special orientation of electrode when the upper end of anode and the lower end of cathode are in a vertical line (Liu et al., 2017) and that a smaller flow rate required longer time to reach the steady stage with a satisfactory purification effect.

1. Introduction

The production of oily wastewater increases with the extensive application of "Enhanced Oil Recovery" (EOR) technologies used for improving oil recovery, particularly in older and declining oil wells/oil reservoirs. "Polymer Flooding" is a dramatic improvement in water-flooding and is quickly becoming one of the preferred EOR. The oily sewage produced by "Polymer Flooding" is not only characterized by high oil concentration, high salinity and micron sized oil droplets with high stability [1], but also the viscosity and emulsification degree of the oily sewage increased with the addition of polymer in the recovery process [2]. The traditional treatment process "natural oil removal (by gravity)–coagulation sedimentation–gravity or pressurized filtration" adopted in oily wastewater treatment cannot achieve the National standards [3], and conventional methods were unfit for demulsification of O/W emulsions due to some limitations [4,5], as shown in Table 1. Nevertheless, EC technology is receiving an increasing acceptance by industry in the light of its advantages compared to other methods. The EC, combined the advantage of chemical coagulation (CC) and electrochemical methods of sewage treatment and characterized by low sludge production, no requirement for chemical use, and easy operation [6], is an efficient and environmentally friendly water purification technology. This process has proven to be very effective in removing varieties of contaminants from water. Chen [7] tested the effectiveness

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Table 1
Comparison of different methods.

Methods	Types	Advantages	Disadvantages
Physical methods	Gravity separation coarse graining flotation, filtration etc.	Easy-control Low-cost	Large footprint long treatment-time unfit for emulsified oil
Chemical methods	Coagulation water quality modification chemical oxidation etc.	High treatment efficiency strong stability	High cost large amounts of sludge
Biological methods	Activated sludge process biological filter process oxidation pond method etc.	No second-pollution low operation cost low sludge production	Large footprint long treatment-time low efficiency poor shock resistance

of gravity separation, centrifugation, flotation, and EC on demulsification of O/W emulsions, and the results showed that EC has the highest removal efficiency.

EC processes carried out in aqueous solutions are quite complex and are a combination of various mechanisms (including direct and indirect redox, coagulation, flotation, etc.). Characterized by strong adsorption capacity of aluminum coagulants and a large number of bubbles existing in aluminum flocs, the aluminum electrodes have been proved to be more efficient than iron electrodes [8]. Besides, the effluent with aluminum electrodes is very clear and stable, while the effluent with iron electrodes appeared greenish first, and then turned yellow and turbid [9]. Therefore, aluminums were adopted as electrodes in our study and the main reactions are as follows [10]:

For anode:

$$Al - 3e \rightarrow Al^{3+} \quad (1)$$

At alkaline conditions:

$$Al^{3+} + 3OH^{-} \rightarrow Al(OH)_{3(s)} \quad (2)$$

At acidic conditions:

$$Al^{3+} + 3H_2O \rightarrow Al(OH)_{3(s)} + 3H^{+} \quad (3)$$

In addition, there is oxygen evolution reaction:

$$2H_2O - 4e \rightarrow O_2\uparrow + 4H^{+} \quad (4)$$

The reaction at the cathode is:

$$2H_2O + 2e \rightarrow H_2\uparrow + 2OH^{-} \quad (5)$$

as Eqs. (1)–(3),a consumable electrode, usually aluminum, used to supply ions into the water stream within the EC reactor, the metal ions combine with water or OH^{-} ions to immediately undergo further spontaneous hydrolysis reactions to form various monomeric species such as $Al(OH)^{2+}$, $Al(OH)_2{}^{+}$, and $Al(OH)_4{}^{-}$, polymeric species such as $Al_2(OH)_2{}^{4+}$ and $Al_2(OH)^{5+}$, which finally transform into amorphous $Al(OH)_{3(s)}$ and less soluble species as Al_2O_3 in terms of many complicated processes [11], as Eqs. (6).

$$Al^{3+} \rightarrow Al(OH)_3{}^{(3-n)} \rightarrow Al_2(OH)_2{}^{4+} \rightarrow Al_{13}complex \rightarrow Al(OH)_{3(s)} \quad (6)$$

Thus, the coagulating ions in EC process are produced 'in situ' for breaking of emulsion, and demulsification mechanism depending on factors such as pH and current density, can be summarized as three points [12]:

(i) Under the action of an external power source, organic substances in the oily wastewater undergo oxidation reactions producing low molecular organic compounds, CO_2, H_2O, and other oxides. At the same time, the cathode produces [H] 'in situ' with a strong reduction capacity, which can degrade the pollutants.
(ii) The metal ions generated at the anode undergo a series complicated hydrolysis and polymerization reactions to form mononuclear or multinuclear polymers. These highly active polymers with strong adsorption properties can destabilize colloidal particles, forming large aggregates that are removed by sedimentation and flotation. The process generally includes charge neutralization, adsorption bridging, double layer compression, and sweep coagulation.
(iii) When direct current is applied to water through a pair of electrodes, the energy barrier is overcome, and the cathode produces

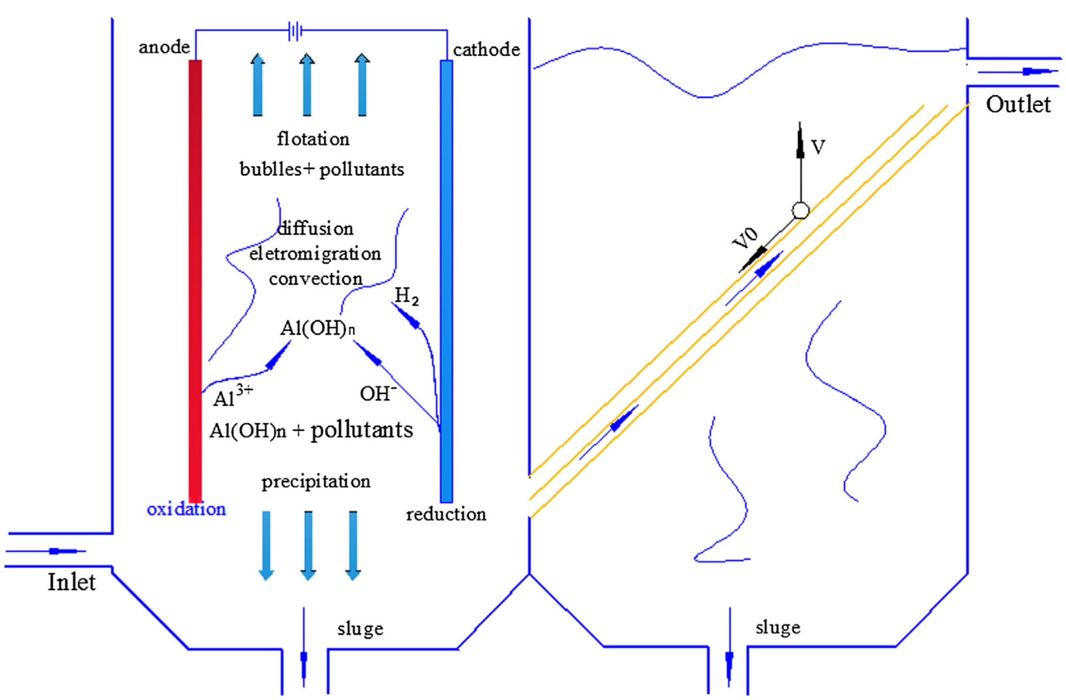

Fig. 1. Working principle of electrocoagulation.