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Removal of organic matter from water using ultrasonic-assisted electrocoagulation method

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Abstract. Organic matter (OM) is commonly occur in freshwaters, and it poses a threat for health and environment. For instance, high OM concentrations increase the sludge volume and cost of the treatment process in turn. Remediation of water or wastewater from OM has thus become an important issue for the treatment industry. Electrocoagulation (EC) is one of the preferred techniques of treatment for remediation of water from OM and other polluting chemicals, as it utilises simple and easy to run machines and tools, it requires less working area in comparison with conventional methods, and it is more rapidly pollutant separation than other methods. EC technique could be described by passing an electric current through sacrificial electrodes to produce metal hydroxides that separate dissolved contaminants from aqueous phase. On the other hand, the EC method has a limited efficiency in the treatment of high OM concentrations. The present study is thus intended to develop a new technique that combines an EC reactor with an ultrasound (US) field to remediate water from OM. The EC reactor, in this study, was made from four aluminium plats (500 cm^2 in gross area) and plastic container (1 L in volume). The operation of this combined method was optimised for the effects of key factors, such as the pH of solution, electrodes gapping, and current density. The obtained results proved that the US-EC technique removed 97.50 % of OM after 5.0 minutes of US irradiation, 20 minutes of electrolysing at current density 4.0 mA.cm^{-2}, initial pH 7 and gap between electrodes 5.0 mm.

1. Introduction

Organic matter (OM) is described is a wide group of compounds that contain carbon in their chemical structures, and they are abundantly presence in the aquatic environments [1]. For example, all forms of microorganisms, such as bacteria, algae and remains of plants are classified as OM [1-3]. Occurrence of OM in freshwaters poses a threat for health and environment, such as the high consumption of coagulants, production of toxic contaminates by reacting with other chemicals, chelating of non-organic contaminants, and clogging of water supply facilities [4]. In addition, the disposal of agricultural and industrial wastewaters and climate changes has intensified the problem of organic pollution of water [5-11].

Many treatment procedures/techniques therefore were employed to remove OM from water. For example, membrane filtrations, adsorptions, oxidations and biological techniques were broadly used in the remediation of water from OM [1, 12-14]. Additionally, the published researches demonstrated that the EC technique, which is passing an electric current through sacrificial electrodes to produce metal

hydroxides that separate dissolved contaminants from aqueous phase, was extensively applied in the remediation of water from OM and other [15-18]. Also, the EC technique could be described as an *in situ* formation of metal hydroxides by subjecting metal electrodes to electric current [19]. The general configuration of the EC reactors is the immersion of metal plates (electrodes) in solution; electric current is applied to erode the electrodes that results in the formation of metal hydroxides in turn. The formed metal hydroxides act as coagulants to separate dissolved pollutants [18, 20]. Thus, the EC treatment is run without adding external chemicals, which makes this technique an eco-friendly alternative for traditional techniques [18, 20]. Additionally, the EC poses other attractive strengths, such as the cheapness, simplicity, ease of operating, and low sludge production that minimises the additional cost for handling and disposal or cycling of solid wastes [21-30]. Furthermore, this method was used to remove a wide range of pollutants, such as nutrients [31, 32], dyes [33], heavy metals [34] and fluoride [35]. As formerly mentioned, the main weakness of this the EC technique is its limited ability to remove high concentration of OM from solutions [1].

To overcome this weakness point in the performance of the EC technique; this study investigated the combination of two techniques, EC and US, to remove OM from water. The purpose of application of the US irradiation is to breakdown the OM into simple formations that can be, then, handled by the EC reactor. The effects of the US on OM is belong to the ability of the US to elevate both temperature and pressure within the solution being treated, which leads to degrade the OM into simple formations [36]. It is noteworthy to mention that as these two techniques, EC and US, do not need chemicals to complete the treatment process, this new combined method could be an eco-friendly alternative for traditional chemical methods [16]. For future researchers, the EC- US will be made a smart unit by supplying it with electromagnetic sensors [37-41].

2. Methodology

2.1. EC reactor
Electrolysing process was performed using a rectangular EC reactor. The latter consists of a four-sided (rectangle) container (1.0 L in capacity) made from plastic, which in turn contains 4 aluminium perforated plates (holes diameter is 0.5 cm), 500 cm^2 in gross area, of 99.50 % purity, Figure 1.
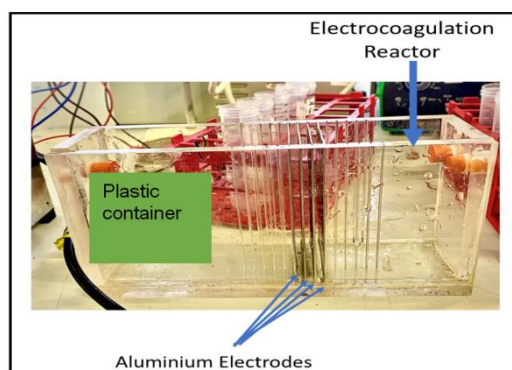
Figure 1. Configuration of the EC reactor.

Aluminium plates were applied, in this study, as anode and cathode because its oxidation potential is low and it can be obtained at relatively low prices [42].

A Fisher Scientific US bath (model: FB-15051) was used to perform the US irradiation process. This US bath was 2.7 L in capacity with variable with variable temperature (upto 60 ºC) and power (upto 280 Watts), and constant frequency (37.0 kHz). Water samples were irradiated for 10.0 minutes before the electrolysing process, this period was chosen according to the relevant literature [36].

2.2. Water sample
The required stock solution to test the efficiency of the US-EC method in the removal of the OM was prepared using deionised water and EDTA (Ethane-1,2-diyldinitrilo), where the required amount of

EDTA (as OM) was added to 1.0 litre of deionised water and properly stirred (to dissolve them). The added amount of EDTA was enough to produce 1000 mg/l of OM. The concentration of the OM in the prepared samples was measured in term of chemical oxygen demand (COD). This stock solution was used to prepare secondary water samples that have less concentration of COD by dilute it with deionised water. The latter were used to run the experiments. The pH value of the secondary samples was adjusted to 4, 7 or 9 by adding the a proper amount of 1.0M HCl or 1.0M NaOH, and sodium chloride was applied to adjust the conductivity to 0.320 mS.cm^{-1}. The needed chemicals, in this study, have been secured from Sigma Aldrich-UK and used as supplied.

2.3. US-EC experiments
The EC treatment was performed by decanting 0.5 litre of the prepared solution into the EC reactor and switching on the DC rectifier to flow the direct electric current through the immersed electrodes in the solution. During the EC experiments, the values of the density of the electric current (CD), pH and gaps between electrodes (ID) were varied from 1.0 to 6.0 $mA.cm^{-2}$, 4 to 9, and 5.0 to 15.0 mm, respectively, to attain the best removal of the OM.

Concentration of OM in water being treated was measured using standard Hach Lange COD cuvettes and Hach Lange spectrophotometer, where 5 mL was taken from the solution at each 5 minutes; filtered at paper filters (0.22 µm) to separate the sludge and foam, and then tested for COD.

The COD removal (RE%) was calculated as follows:

$$RE\% = \frac{C_o - C_t}{C_o} \times 100 \%$$ (1)

Where, C_o is the initial COD concentration, and C_t is measured COD concentration.

Then, in the second phase of this study, freshly prepared samples were irradiated in the US bath for 10 minutes before they were electrolysed in the EC reactor under the same conditions obtained from the previous experiments.

To avoid any unexpected altering of the results due to the accumulation of sludge on the surfaces of the electrodes, the latter were cleaned at the end of each experiments using sandpaper, washed with HCl solution and finally washed with water. The experiments were run for 20 minutes at laboratory that had average temperature of 20±2 ºC.

3. Results and discussion

3.1. OM removal using EC technique

As it was formerly mentioned, this study was initiated by measuring the ability of the EC technique to remove OM from water. In this phase, the effects of pH, CD and ID on the removal of OM were measured in order to reach the best efficiency, which will be compared with best efficiency of the US-EC method.
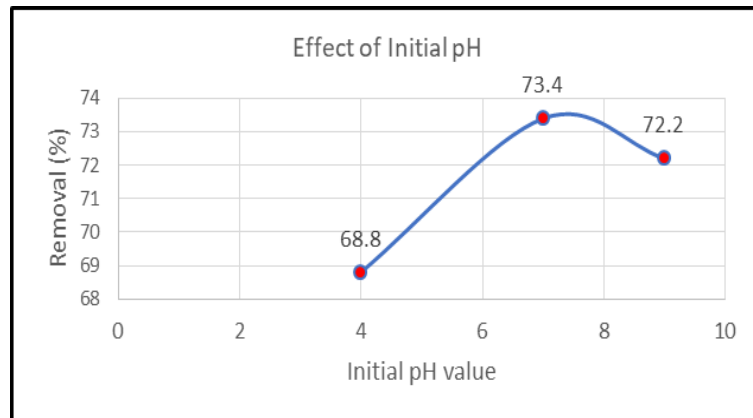
3.1.1. Effects of initial pH
The effects of water pH value was considered in this study as the pH governs the types of the formed metal hydroxides, which directly effects the removal efficiency in turn [34].

The required experiments were run at various values of initial pH (4-9), fixing the CD and ID at 4.0 mA.cm^{-2} and 5mm, respectively.

The findings of these runs are displayed in figure 2, which shows that the best OM removal was 73.4% (in terms of COD) that was reached when the pH was 7.0. At the same time, increasing the pH of water to 9.0 or decreasing it to 4.0 resulted in lower removal of OM, which were about 72% and 69%, respectively.The explanation for this increase/decrease in the removal of OM with the change in initial pH, according to the similar literature, is the change in the chemical form of the produced metal oxides, which influences the removal of COD in turn. In general, formed metal hydroxides in low or high pHs poses a relatively low adsorption capacity for OM.

 Hence, the rest of EC runs were performed at pH 7.

Figure 2. Changes in the removal of OM with the initial pH.

3.1.2. Effects of CD

The second factor that was investigated in this study is CD as it determines the removal of any contaminant by the EC technique. The CD determines the removal of contaminants through determining the released amounts of metal hydroxides and hydrogen gas [34]. Thus, the relationship between the removal of OM and the applied CD has been investigated, in this study, by performing sets of experiments at different CDs (ranging from 1.0 to 6.0 mA.cm^{-2}) at constant pH of 7.0 and ID of mm. The collected results from these experiments are presented in Figure 3, which proves a positive relationship between the OM removal and the applied CD. Where it was found that removal of OM raised from about 60% to 94.5% when the CD changed from 1 to $6\,\mathrm{mA.\,cm^{-2}}$, respectively. The explanation for this increase in the removal of OM with the change in the CD, according to the similar literature, is the CD determines the released amounts of metal hydroxides and hydrogen gas, which determines the removal of OM in turn [34]. At the same time, a negative relationship between the CD and power consumption, hence, the rest of EC runs were performed at CD of $4.0\,\mathrm{mA.\,cm^{-2}}$ and pH 7.
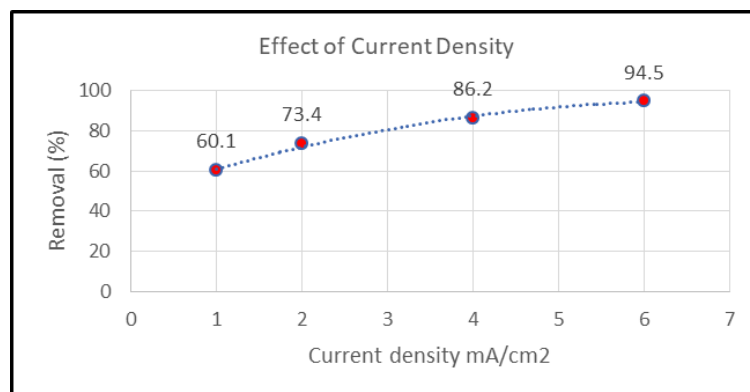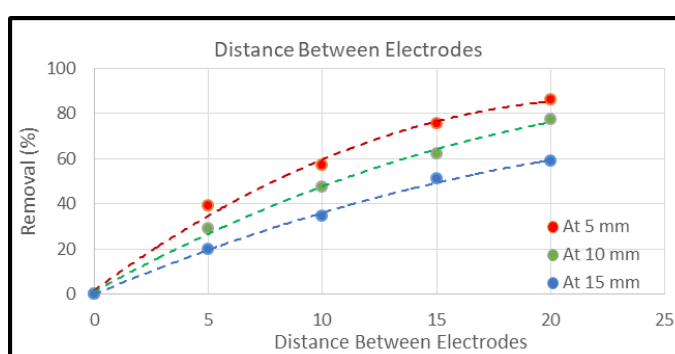

Figure 3. Changes in the removal of OM with the CD.

3.1.3. Effects of ID

The last investigated factor was the electrodes gaping (ID), which was investigated due to its importance in controlling the resistance for current flow through the EC reactor [18]. The experiments were run at ID of 5.0, 10.0 and 15.0 mm, pH of 7 and CD of 4.0 mA. cm^{-2}.

The impacts of this facto on OM removal are displayed in Figure 4, which proves a negative relationship between the OM removal and the ID. Where, it was found that removal of OM dropped from about 86% to 59% when the ID changed from 5.0 to 15.0 mm, respectively.

The previous studies explained the negative relationship between the contaminants removal and the ID by the increase in the resistance (at high ID) for electric current flow that minimises the release of metal hydroxides, and consequently the removal efficiency [32]. Hence, the rest of EC runs were performed at ID of 5.0 mm, CD of 4.0 mA. cm^{-2} and pH 7.


Figure 4. Changes in the removal of OM with the ID.

4. Removal of OM using the US-EC technique

In this phase of the investigation, the new US-EC technique was employed to remove OM from solution. The experiments were started by irradiating water samples in the US bath for 10 minutes at power of 0.28 kW and frequency of 37 kHz as a pre-treatment stage before the EC treatment, then these samples were electrolysed for 20 minutes at pH, CD and ID of 7.0, 4 mA.cm^{-2} and 5.0 mm, respectively (the best operating conditions from the EC experiments).

The outcomes of these experiments, depicted in Figure 5, proved that the US irradiation raised the OM removal from about 86% (without US irradiation) to about 98%. This enhancement in the OM removal could be attributed to the produced temperature and pressure in the bulk solution that breakdown the OM into smaller formations, which in turn improve the removal of the OM [36].

Hence, it could be concluded from the obtained results that the US-EC technique is a promising alternative for water treatment.
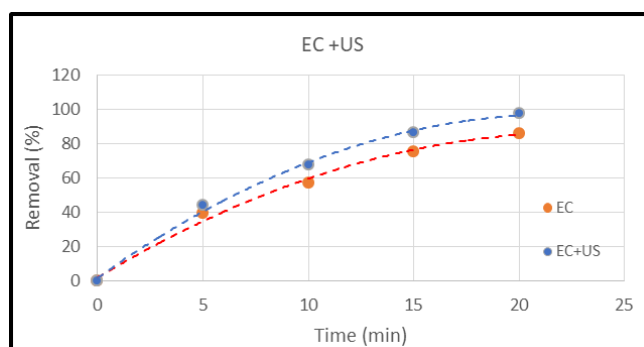
Figure 5. Effect of ultrasonic field on COD removal.

5. Conclusions

The present investigation was devoted to validate the application of US irradiation as a pre-treatment stage before the EC treatment can remarkably enhance the remediation of water from OM. Generally, the outcomes of this study indicated that though the EC technique demonstrated good efficiency in the remediation water from OM, the US irradiation still needed to attain efficient removal of OM. The efficiency of the new US-EC technique depends on the pH of solution, CD and ID. According to the obtained results, the efficiency of this technique could be improved by increasing the CD, decreasing the ID or maintain neutral pH value. At the optimum operation conditions, which are CD of $4.0 \, \mathrm{mA. \, cm^{-2}}$, pH of 7.0 and ID of 5mm, the US-EC technique removed about 98% of the OM in water. Although this study covered key aspects in the performance of the US-EC technique, more studies are still needed to validate the performance of this technique. For example, more studies could be done to check the effects of other factors, such as the treatment time of both US and EC, on the removal of the OM.

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