Application of hybrid electrocoagulation–filtration methods in the pretreatment of marine aquaculture wastewater

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ABSTRACT

The aim of this study was to provide technical means and data support for enhancing the filtration pretreatment capacity of a recirculating aquaculture system. A continuous flow electrocoagulation (EC)–filtration system was designed and its application in the pretreatment of marine aquaculture wastewater was studied. The influences of anode combination modes, hydraulic retention times (HRTs) of the EC reactor and filter pore sizes on the water treatment capacity were investigated. Results showed that EC could significantly enhance the treatment efficiency of the filtration equipment used in subsequent steps. Al-Fe electrodes used as anode led to better processing capacity of this system, and the optimum anode was 3Al + Fe. With the increase of HRT and decrease of filter pore size, the enhanced effect of the EC process on the filter was more obvious. When the current density was 19.22 A/m², the anode was 3Al + Fe, the HRT was 4.5 min and the filter pore size was 45 μm, the removal efficiency of the system for *Vibrio*, chemical oxygen demand, total ammonia nitrogen, nitrite nitrogen (NO₂⁻-N), nitrate nitrogen (NO₃⁻-N) and total nitrogen was 69.55 ± 0.93%, 48.99 ± 1.42%, 57.06 ± 1.28%, 34.09 ± 2.27%, 18.47 ± 1.88% and 55.26 ± 1.42%, respectively, and the energy consumption was (26.25 ± 4.95) × 10⁻³ kWh/m³.

Key words | anode combination mode, EC–filtration system, energy consumption, recirculating aquaculture system, water treatment

HIGHLIGHTS

- The combination technology of EC and filtration was used for the pretreatment of aquaculture wastewater.
- The filtration capacity of physical filtration equipment was improved through the EC–floculation.
- Al-Fe combined anode was used to improve the treatment capacity of the EC–filtration system.

INTRODUCTION

As an environmentally friendly and healthy aquaculture technique, recirculating aquaculture has attracted increasing attention, and the continuous improvement and development of recirculating aquaculture system (RAS) procedures are now established practices (Ben-Asher & Lahav 2019). Generally, in the RAS, the effluent of the aquaculture tank is initially channeled through solid–liquid separation equipment to remove the majority of suspended particulate
matter, and subsequently through equipment that provides biological oxidation, oxygenation, disinfection and other processes, in order to achieve recycling (Van Rijn 2013). The pretreatment of aquaculture wastewater by solid-liquid separation equipment greatly reduces the processing load sustained by subsequent water treatment units and it reduces the overall energy consumption of the system (Xiao et al. 2019). In many RASs, a microscreen drum filter is used because of its advantages such as high applicability, reduced space requirements, large water treatment capacity and limited head loss (Ebeling et al. 2006; Xiao et al. 2019). The removal rate of suspended particles depends on the pore size of filtration membranes, and the smaller the pore size is, the higher the removal rate is (Xiao et al. 2019). Nevertheless, because of the need to ensure the smooth circulation of water in RASs and because of process and cost limitations, it is difficult to improve the physical filtration efficiency by reducing the filter pore size continuously, thus hampering the pursuit of higher filtration levels of RASs.

One traditional method to enhance the efficiency of sewage filtration, involves adding chemical flocculants such as aluminum (Al) or iron (Fe) salts to the sewage ahead of filtering (Kimura et al. 2014). Ebeling et al. (2006) used chemical flocculants (alum) to improve the filtering capacity of the belt filter system on the tail water of the RAS. Electrocoagulation (EC) is also a common method for enhancing solid-liquid separation of sewage, in which the sacrificial anode releases metal cations with flocculation characteristics under the action of an external electric field, then forms flocculants to absorb pollutants in the water (Heffron et al. 2019). In addition, the EC process produces active oxidants, such as reactive oxygen species and free chlorine, which can degrade chemical oxygen demand (Moussa et al. 2017; Al-Qodah et al. 2019), color (Medel et al. 2019), microorganisms (Heffron et al. 2019), ammonia and nitrite (Mook et al. 2012), etc. Compared with traditional chemical flocculation, EC has the advantages of causing less secondary pollution and less sludge output and having more controllability; also it involves easier operation requirements (Linares-Hernández et al. 2009; Moussa et al. 2017). At present, EC-filtration technology has been successfully applied to surface water purification (Zhao et al. 2014; Chellam & Sari 2016), industrial wastewater treatment (García-Morales et al. 2018) and in other fields.

The size, structure and strength of flocs produced by EC have a significant impact on the subsequent filtration and separation processes (Rong et al. 2013). The properties of flocs are related to many factors, such as anode material (Govindan et al. 2014), current density and pH (Hu et al. 2017) and hydraulic conditions (Rong et al. 2013). Among these factors, the anode material and EC treatment time seem particularly important, because they determine the type and quantity of flocculants (Moussa et al. 2017). Al and Fe are the most common anode materials (Nidheesh & Singh 2017). During the EC process, flocules produced by Al anode have a large surface area, strong adsorption capacity, but slow complexation speed and low structural strength, compared with Fe anode-produced flocules (Moussa et al. 2017). In recent years, an increasing number of studies have focused on the Al-Fe composite electrodes. Linares-Hernández et al. (2009) evaluated the removal efficiency of organic pollutants in highly complex industrial wastewater by EC with Al, Fe and Al + Fe as sacrificial anodes, respectively, and found that the optimum practice was the use of Al + Fe. Aoudj et al. (2015) studied the effect of anode materials on the pretreatment of acidic semiconductor effluents by EC-electroflotation, and found that a hybrid Fe-Al anode performs better than Fe or Al anodes in terms of simultaneous removal of Cr(VI), fluoride and turbidity.

Through the flocculation initiated by EC processes, the small suspended particles in the wastewater coagulate into larger particles, improving filtration accuracy without changing the original filtration equipment and showing that EC-filtration technology has a great potential and application for sewage treatment procedures (Xie et al. 2017). However, the application of EC technology in aquaculture wastewater is rare, and the application of EC-filtration technology in aquaculture has not been reported. The main objective of the present research was to investigate the pretreatment effect of EC-filtration technology on aquaculture wastewater, which is expected to provide some reference for the future improvement of the filtration capacity of the RAS.

MATERIALS AND METHODS

Experimental set-up

In this experiment, a continuous flow EC-filtration system with a laboratory scale was designed, consisting of three sections: an EC reactor, a mixed flocculator and filtration equipment. The EC reactor consisted of a DC power supply (SS-3030KD, Guangdong, China), electrodes and an electrolytic cell. The voltage and current regulation ranges of the DC power supply were 0–30 V and 0–30 A, respectively. The effective volume of the EC reactor was 5 L, in which nine groups of electrodes were arranged in parallel: four groups of anodes were composed of Al plate
or Fe plate electrodes, and five groups of cathodes contained titanium (Ti) plate electrodes. The distance between the electrodes was 1.5 cm. Each electrode plate was 20 cm in length, 5 cm in width and 0.5 cm in thickness, the under-water length of electrodes was 15 cm and the effective working area was 0.016 m². The mixed flocculator was arranged vertically and its effective volume was about 1.5 L. It was located between the EC reactor and the filtration equipment, adapted to an upward flow design. The bottom of the flocculator was equipped with a micro-aeration system and the introduced gas volume (1.0 L/min) served the purpose of disturbing the water body to avoid floc deposition. The function of the mixed flocculator was to improve the contact frequency between the flocculants and the suspended particles, thereby improving the efficiency of flocculation and adsorption. After the flocculation treatment by EC, the aquaculture wastewater was finally filtered. The filtration equipment used was a micro-screen drum filter whose filter pore size is generally between 50 μm and 75 μm (Xiao et al. 2019). In this trial, the microscreen drum filter was 10 cm in diameter and 15 cm in length. The filter screen was made of stainless steel and the effective area was 0.031 m². The schematic diagram of the EC–filtration system is shown in Figure 1.

### Aquaculture wastewater

The wastewater used in the experiment originated from effluents of the aquaculture pond used for the Litopenaeus vannamei RAS (Dalian Huixin Titanium Equipment Development Co., Ltd, Dalian, China). The water quality parameters are shown in Table 1.

![The schematic diagram of the EC–filtration system](image)

**Figure 1 |** The schematic diagram of the EC–filtration system: 1. Water inlet. 2. Liquid flowmeter. 3. DC power supply. 4. Electrodes. 5. EC reactor. 6. Sewage outlet. 7. Airstone. 8. Mixed flocculator. 9. Microscreen drum filter. 10. Air pump. 11. Water outlet.

### Experimental design

In this experiment, five different anode combinations (Al-Al-Al, Al-Fe-Al-Al, Al-Fe-Al-Fe, Fe-Al-Fe-Fe, and Fe-Fe-Fe-Fe), three different EC reactor hydraulic retention times (HRTs) (1.5 min, 3.0 min, and 4.5 min) and four filtration pore diameters (75 μm, 63 μm, 54 μm, and 45 μm) were used. A control group was set up without EC treatment and only relying on filtration equipment. The current density of the EC reactor was 19.22 A/m². EC operation parameters are shown in Table 2. Different HRTs can be obtained by controlling the liquid flowmeter and by modifying the inlet water flows. When the HRT of the EC reactor was 1.5 min, 3.0 min, and 4.5 min, the inlet water flow was about 200 L/h, 100 L/h, and 67 L/h, respectively. The experiment was aimed at measuring the removal effect on the total number of Vibrio, chemical oxygen demand (CODMn), total ammonia nitrogen (TAN), nitrite nitrogen (NO2-N), nitrate nitrogen (NO3-N) and total nitrogen (TN) in aquaculture wastewater through

### Table 1 | Characteristics of aquaculture wastewater

| Water quality indexes | Average value with standard deviation |
|-----------------------|---------------------------------------|
| pH                    | 7.12 ± 0.01                           |
| Salinity              | 15 ± 0.03                             |
| Conductivity (μS/cm)  | 36.35 ± 0.53                          |
| CODMn (mg/L)          | 12.41 ± 0.44                          |
| TAN (mg/L)            | 1.63 ± 0.02                           |
| NO2-N (mg/L)          | 0.44 ± 0.01                           |
| NO3-N (mg/L)          | 14.94 ± 0.07                          |
| TN (mg/L)             | 97.92 ± 0.96                          |
| Total number of Vibrio (CFU/mL) | 22,110 ± 311 |

### Table 2 | The operation parameters of EC

| Anode combination modes | Current (A) | Supply voltage (V) | Total resistance (Ω) | Power (10⁻³ kW) |
|-------------------------|-------------|--------------------|----------------------|-----------------|
| Al-Al-Al-Al            | 1.23 ± 0.01 | 1.32 ± 0.03        | 1.05 ± 0.05          | 1.62 ± 0.09     |
| Al-Fe-Al-Al (3Al + Fe) | 1.23 ± 0.01 | 1.42 ± 0.03        | 1.22 ± 0.02          | 1.75 ± 0.33     |
| Al-Fe-Al-Fe (2Al + 2Fe)| 1.24 ± 0.01 | 1.68 ± 0.02        | 1.36 ± 0.01          | 2.09 ± 0.04     |
| Fe-Al-Fe-Fe (Al + 3Fe)| 1.23 ± 0.01 | 2.13 ± 0.02        | 1.72 ± 0.02          | 2.62 ± 0.03     |
| Fe-Fe-Fe-Fe (4Fe)      | 1.22 ± 0.01 | 3.44 ± 0.02        | 2.81 ± 0.02          | 4.20 ± 0.06     |
the use of an EC–filtration system with different combinations of anodes, different HRTs and variable size of filtration pores. Samples were collected at the water inlet of the system to measure the initial water quality index. After the aquaculture wastewater was treated in the EC reactor, samples were collected in the mixed flocculator to further test their water quality index. Based on the change in water quality detected, it was possible to evaluate the treatment capacity of the EC reactor under different test conditions. Finally, the aquaculture wastewater post-EC treatment was filtered through filtration equipment in order to complete the pretreatment procedure. It was then sampled to evaluate the treatment capacity of the whole EC–filtration system for aquaculture wastewater. Water samples were taken three times consecutively to reduce the experimental error. Also, power supply data were recorded every 20 seconds and adjusted, to avoid increasing the test error.

Analysis method

The total number of *Vibrio* was detected immediately using the plate counting method (TCBS plate) after the water sample was collected (Pang et al. 2006). For the detection method of CODMn followed Zhen et al. (2016). And TAN, NO2-N, NO3-N and TN were measured using standard methods (APHA 1998). Salinity, pH and conductivity were measured by a multi-parameter water quality analyzer (YSI-556, USA). Reported results are based on mean and standard deviation (mean ± SD).

Removal efficiency (RE, %) of pollutants was calculated using Equation (1):

\[
RE(\%) = \frac{C_0 - C_1}{C_0} \times 100
\]

where \(C_0\) is the initial concentration of pollutants (mg/L) and \(C_1\) is the concentration of pollutants after treatment (mg/L).

The EC reactor’s energy consumption \((W, \text{kWh/m}^3)\) was calculated using Equation (2):

\[
W = \frac{P \cdot t}{V}
\]

where \(P\) is the power of the EC reactor (kW), \(t\) is the treatment time (h) and \(V\) is the volume of the EC reactor (m\(^3\)).

In this experiment, the energy efficiency ratio (EER, %) was used to evaluate the practicability of different composite anodes in wastewater treatment; the calculation formula is shown in Equation (3).

\[
EER(\%) = \frac{W_{(HRT,i)} - W_{\text{no-EC}}}{RE_{(HRT,i)} - RE_{\text{no-EC}}} \times 100
\]

where \(EER(\%)\) is the ratio between the energy increase and the efficiency increase of pollutant removal, HRT is the hydraulic retention time of the EC reactor, \(i\) is the anode combination mode and \(W_{(HRT,i)}\) (kWh/m\(^3\)) is the energy consumed by the EC reactor when a certain HRT and anode combination is \(i\). \(W_{\text{no-EC}}\) (kWh/m\(^3\)) is the energy consumption of the control group. \(RE_{(HRT,i)}(\%)\) is the pollutant removal efficiency when a particular HRT and anode combination is \(i\). \(RE_{\text{no-EC}}(\%)\) is the pollutant removal efficiency of the control group.

RESULTS AND DISCUSSION

The removal effect of the EC–filtration system on the total number of *Vibrio*

In production, RAS uses ozone and ultraviolet synergistic sterilization to control pathogenic bacteria, which usually requires massive energy consumption (Van Rijn 2013; Xiao et al. 2019). Although ozone has strong bactericidal ability, it will threaten the health of cultured organisms, so the use of ozone in RAS needs to be strictly controlled (Xiao et al. 2019). The ultraviolet lamp needs regular maintenance to ensure work efficiency, which increases the cost of disinfection (Xiao et al. 2019). EC–filtration technology, as a pretreatment method of aquaculture water, can eliminate pathogenic bacteria while removing residual bait and feces; its application in RAS can reduce the work intensity and energy consumption of subsequent ozone and ultraviolet sterilization equipment. The EC–filtration system removes *Vibrio* based on two factors. (1) The role of electrodes, electric field and oxidants. During EC, the electrode adsorbs and kills a part of bacteria, and the electric field destroys the cell membrane of germs (Ghernaout et al. 2019). In addition, EC also produces active oxidizing substances with a bactericidal function, such as -OH, O\(^{2-}\) and Cl\(_2\) (Ghernaout et al. 2019). (2) The role of electrical neutralization/flocculation and physical filtration. Charged ions generated by EC can neutralize the surface charge of pathogenic bacteria, reduce electrostatic repulsion and form flocs (Chellam & Sari 2016). In addition, the floculants produced by EC can actively absorb and flocculate the bacteria (Chellam & Sari 2016). These floculants are eventually removed by physical filtration.

Figure 2(a) shows the effect of different composite anodes and HRTs on the removal of *Vibrio* in the EC reactor. When the HRT was 1.5 min and anode combination was 4Al, 3Al + Fe, 2Al + 2Fe, Al + 3Fe, and 4Fe, the removal efficiency of
the EC reactor for *Vibrio* was 6.98 ± 1.90%, 8.19 ± 1.61%, 12.56 ± 1.13%, 15.27 ± 1.13%, and 17.23 ± 0.98%, respectively; and when the HRT increased to 4.5 min, the removal efficiency increased to 37.07 ± 1.40%, 38.79 ± 0.74%, 41.80 ± 1.82%, 45.42 ± 1.49%, and 45.88 ± 0.56%, respectively. It follows that the EC reactor had a significant removal effect on the total number of *Vibrio*. Also, without a solid–liquid separation, the removal efficiency increased with the addition of the iron electrode proportion in the combined anode and with the increase of HRT. Furthermore, the experiment proved that the sterilization of Fe as anode during EC was more effective than that of Al anode, which is consistent with results reported by Ndjomgoue-Yossa et al. (2015). This was due to the fact that Fe(II) was produced when Fe was used as anode, which could kill the bacteria in aquaculture water, while Al(III) did not have bactericidal ability (Moussa et al. 2017). Also, the longer HRT meant that more oxidants were released by the EC reactor into the wastewater, resulting in a better bactericidal effect.

Figure 2(b1)–2(b3) show the effects of different composite anodes and filter pore sizes on the *Vibrio* removal by EC–filtration when the HRT of the EC reactor was 1.5 min, 3.0 min, and 4.5 min, respectively. Removal efficiency for *Vibrio* by EC–filtration showed different trends with the addition of the Fe electrode proportion in composite anodes. This was determined by the two sterilization procedures characterizing the EC–filtration system. When the electrode, electric field and electro-oxidation played a major role, the removal efficiency for *Vibrio* increased with the addition of the Fe electrode proportion in composite anodes. When the electric neutralization/flocculation–filtration became predominant, the removal efficiency increased initially and then decreased with the addition of the Fe electrode proportion in combined anodes. This was due to the fact that the flocculants produced by the Al–Fe combined anodes in the EC process were superior to those of the single Al or Fe electrode in terms of structure and strength; also they showed a better filtration performance (Aoudj et al. 2015). The results showed that the control group could also reduce the total number of *Vibrio*. When the HRT was 4.5 min and the filter pore sizes were 75 µm, 63 µm, 54 µm, and 45 µm, the removal efficiency of the
filtration equipment for Vibrio was 5.2 ± 1.84%, 12.26 ± 0.98%, 15.05 ± 0.85%, and 21.36 ± 2.75%, respectively. Previous research has shown that protozoa and pathogenic bacteria in the wastewater can be decreased after filtration pretreatment, further reducing the energy required for the subsequent use of equipment for sterilization and disinfection procedures (Chellam & Sari 2016). A portion of microorganisms in the aquaculture wastewater attaches to the suspended particles, which act as a substrate for microbial growth (Wold et al. 2014). As the particulates are filtered by the filtration equipment, the pathogenic bacteria attached to them will be removed. Compared to the control group, the removal efficiency of experimental groups for Vibrio was obviously greater. The longer the HRT and the smaller the filtration pore diameter were, the better the removal effect on the total number of Vibrio was. In this experiment, the removal efficiency of the EC–filtration system was the highest when the EC reactor HRT was 3.31 times greater than the results seen in the control experiment, the removal efficiency on the total number of Vibrio was obviously greater. The longer the HRT and the smaller the reactive site anodes and Al anodes. Linares-Hernández et al. (2009) evaluated the impact of different anode materials on the removal of COD from industrial wastewater in the EC process, and reached the same conclusion. In this experiment, when the anode combination was 4Fe and HRT was 4.5 min, the COD removed by the EC reactor through electro-oxidation was the greatest. The highest removal rate was 22.97 ± 0.30% and the energy consumption was (65.00 ± 0.90) × 10⁻³ kWh/m³.

**Figure 3(c)** shows the effect of different composite anodes and filter pore sizes on CODMn removal in the EC–filtration system when the HRT of the EC reactor was 1.5 min, 3.0 min, and 4.5 min, respectively. In the control group, when the HRT was 4.5 min and the filter pore sizes were 75 μm, 63 μm, 54 μm, and 45 μm, the removal efficiency of CODMn was 2.26 ± 0.56%, 4.67 ± 0.44%, 7.09 ± 0.32%, and 9.85 ± 0.49%, respectively. The removal efficiency of CODMn in aquaculture wastewater by physical filtration depended on filtration pore size. In general, the aperture size of the RAS filtration equipment is 120–300 mesh (about 125–54 μm), and the most used is 200 mesh (about 75 μm) (Xiao et al. 2019). Only relying on traditional physical filtration, the experiment yielded a relatively low removal efficiency, which was a challenge for other water treatment units of the RAS. The removal efficiency of CODMn through filtration equipment depends on the size of organic particles in the water when the filtration aperture is constant; and the larger the particle size, the higher the removal efficiency is. In the EC–filtration system, flocculants can be released into aquaculture wastewater by the EC reactor, which can increase the size of organic particles, thus improving the efficiency of the subsequent physical filtration (Ben-Sasson et al. 2013). When the HRT was 4.5 min and the filter pore sizes were 75 μm, 63 μm, 54 μm, and 45 μm, the highest removal rates of CODMn by EC–filtration (with anode combination 2Al + 2Fe) were 33.28 ± 0.50%, 41.02 ± 0.95%, 46.49 ± 0.85%, and 53.02 ± 0.74%, respectively. Removal efficiency increased by 14.73 times, 8.78 times, 6.56 times, and 5.39 times compared to the control group. Therefore, the EC–filtration system has an excellent removal effect on CODMn in aquaculture wastewater, and without changing the original filter aperture, EC can immensely enhance the capacity of the physical filtration equipment to remove CODMn. Sanyal et al. (2015) had achieved positive results in the treatment of high-strength wastewater by EC and polyelectrolyte multilayer.
membranes. Zhao et al. (2014) treated water produced by EC pretreatment before reverse osmosis membrane filtration and the removal rate of COD reached 66.64%. Figure 3(d1)–3(d3) also show that the removal efficiency of CODMn by EC–filtration initially increased and then decreased with the addition of the Fe electrode proportion to composite anodes. When the filter pore sizes were 75 μm, 63 μm and 54 μm, the EC–filtration system with 2Al + 2Fe as anode had the best removal effect on CODMn, followed by the system with Al + 3Fe, 4Fe, 3Al + Fe, and 4Al as anodes, respectively. With the decrease in filter aperture size, the advantage of the Al-Fe combined anode was more and more evident. When the filter aperture was 45 μm, the removal efficiency with the Al-Fe composite anode was better than that with either Al or Fe as anode. It was proved that the performance of the combined electrode was better than that of the single electrode in terms of flocculation.

Effects of the EC–filtration system on TAN, NO2−N, NO3−N and TN removal

EC–filtration technology can remove TAN, NO2−N and NO3−N at the same time as removing residual bait and feces (Mook et al. 2012; Nazlabadi et al. 2019). Applying EC as a pretreatment process can not only reduce the load of subsequent biological oxidation equipment (such as moving bed biofilter, fixed film aerobic bioreactor), but also improve the activity of microorganisms (Al-Qodah et al. 2019), thereby improving the nitrogen removal capacity of RAS and reducing the threat of nitrogen pollutants to cultured organisms.

The experiment showed that the removal efficiency of TAN and NO2−N increased with the increase of HRT and the addition of the Fe electrode proportion to composite anodes, as shown in Figure 4(e1) and 4(e2). As the HRT and the proportion of Fe electrode in combined anodes increased,
the amount of oxidants released by the EC reactor into the water also increased, resulting in the enhancement of TAN and NO$_2$-N removal efficiency. When the HRT was 4.5 min and the anode was 4Fe, the EC reactor had the best removal effect on TAN and NO$_2$-N, which reached 63.19 ± 1.69% and 67.42 ± 3.47%, respectively, and the energy consumption was (63.00 ± 0.90) × 10$^{-3}$ kWh/m$^3$. Under the same conditions, when the anode was 4Al, the removal rates of TAN and NO$_2$-N were 51.53 ± 1.28% and 24.24 ± 3.47%, respectively, and the energy consumption was (24.30 ± 1.35) × 10$^{-3}$ kWh/m$^3$. It follows that, compared with Al, using Fe as the sacrificial anode of the EC reactor was better for the treatment of TAN and NO$_2$-N but it involved higher energy consumption.

A portion of TAN and NO$_2$-N in the aquaculture wastewater is converted into NO$_3$-N during EC processes (Mook et al. 2012). It is not accurate to discuss the effect of HRTs and combined anodes on the removal of NO$_3$-N in the EC reactor only based on the change of NO$_3$-N concentration in the inlet and outlet water. It is necessary to refer to the change in TN concentration in the process of EC. Figure 4(e3) and 4(e4) show the removal effects of EC on NO$_3$-N and TN under different HRTs and combined anodes. The removal rates of NO$_3$-N and TN increased with a longer HRT in the EC reactor. Furthermore, Figure 4(e3) shows that the removal rate of NO$_3$-N decreased with the addition of the Fe electrode proportion to composite anodes, while there was no significant difference in TN removal (Figure 4(e4)). This was due to the fact that the removal rates of TAN and NO$_2$-N in the EC reactor increase with the adding of the Fe electrode proportion to the composite anode, which led to the increase in the total concentration of NO$_3$-N. However, the anode combination had no obvious effect on TN removal rate, which proved that the anode material had no effect on the electroreduction of the EC reactor.

The suspended solids in the aquaculture water contain a certain amount of TN, accounting for 10–40% of total TN, which can be removed by solid–liquid separation (Van Rijn 2015). Therefore, in addition to electroreduction, the EC–filtration system can also remove TN from aquaculture wastewater by flocculation–filtration. Figure 5 shows the effects of the EC–filtration system on the removal of TN. When the HRT was 4.5 min and the filter pore sizes were...
75 μm, 63 μm, 54 μm, and 45 μm, the TN removal efficiencies in the control group (no-EC) were $2.91 \pm 0.18\%$, $6.87 \pm 0.55\%$, $9.90 \pm 0.95\%$, and $12.90 \pm 1.53\%$, respectively. The TN removal efficiency of the filtration system clearly increased after EC treatment, and the higher the HRT of the EC reactor, the greater its enhancement effect on the subsequent use of filtration equipment. When the HRT was 4.5 min and the filter pore sizes were 75 μm, 63 μm, 54 μm, and 45 μm, the highest removal rates of TN by EC–filtration system (2Al + 2Fe as anode) were $27.40 \pm 0.81\%$, $38.76 \pm 0.98\%$, $50.01 \pm 1.54\%$, and $58.90 \pm 1.96\%$, respectively. The removal rate was 9.42 times, 5.64 times, 5.05 times and 4.57 times that of the control group, respectively, and energy consumption was $(31.35 \pm 0.6) \times 10^{-3} \text{kWh/m}^3$. Results show that the EC–filtration system has an extremely good removal effect on TN in aquaculture water. The TN removal efficiency of the EC–filtration system increased initially and then decreased with the addition of the Fe electrode proportion to composite anodes. When the anode composite was 2Al + 2Fe, the removal efficiency was the highest, followed by 3Al + Fe, Al + 3Fe, 4Al, and 4Fe, respectively. This proved that the flocculation produced by the anode composed of Al and Fe in the EC process was stronger than that of a single Al or Fe anode, and that the flocculation of the Al anode was better than that of the Fe anode (Aoudj et al. 2015).

**Energy consumption analysis**

In this paper, the application of composite anodes in the pretreatment of aquaculture wastewater through the EC–filtration system was evaluated in terms of electric energy loss. From Table 3, results show that during the EC process, the energy consumption of the system increased with the addition of the Fe electrode proportion to the composite anodes and with the increase in the HRT of the EC reactor.
The study found that the electro-oxidation of the EC reactor was stronger when Fe was used as anode than when Al was used; it also found that the treatment efficiency in terms of COD$_{Mn}$, TAN and NO$_2$-N removal was higher. When Al was used as sacrificial anode, the adsorption/flocculation capacity of the EC reactor was greater than when Fe was used, and the removal efficiency for Vibrio and TN was higher. However, under the same operating conditions, the energy consumption of the EC reactor with the anode combination of 4Fe was 2.59 times greater than that of the EC reactor with the anode combination of 4Al. By utilizing Al-Fe composite anodes, the running energy consumption of the EC reactor was 4.5 min, the filter pore size was 45 $\mu$m and the anode combination was 3Al + Fe, the removal efficiency for Vibrio, COD$_{Mn}$, TAN, NO$_2$-N, NO$_3$-N and TN was 69.55 $\pm$ 0.93%, 48.99 $\pm$ 1.42%, 57.06 $\pm$ 1.28%, 34.09 $\pm$ 2.27%, 18.47 $\pm$ 1.88%, and 55.26 $\pm$ 1.42%, respectively, and the energy consumption of the EC reactor was $(26.25 \pm 4.95) \times 10^{-3}$ kWh/m$^3$.

EC can improve the treatment capacity of the filtration system, thereby reducing the load of subsequent water treatment units. However, the impact of the EC-filtration system on the scale and energy consumption of subsequent water treatment (biological oxidation, disinfection, etc.) units needs further study. Moreover, the impact of metal ions and active oxidants produced by EC on the cultured organisms also needs to be investigated.

### CONCLUSIONS

In this experiment, EC-filtration technology was used to pretreat aquaculture wastewater, and the effects of anode combinations, EC reactor HRTs and filter pore sizes on pollutants removal were studied. The main findings are listed as follows.

1. EC process could enhance the capacity of the subsequent physical filtration equipment. The EC-filtration system was found effective for the removal of Vibrio, COD$_{Mn}$, TAN, NO$_2$-N, NO$_3$-N and TN in aquaculture wastewater, and the highest removal efficiency was 70.75 $\pm$ 2.46%,
53.02 ± 0.74%, 63.19 ± 1.69%, 67.42 ± 3.47%, 19.28 ± 0.11% and 58.90 ± 1.96%, respectively.

(2) Compared with the Al electrode, when the Fe electrode was used as anode, the EC–filtration system had a stronger electro-oxidation capacity, which was conducive to the removal of TAN and NO2–N. However, the flocculation effect was relatively weaker and the energy consumption was increased by 2.59 times.

(3) In comparison with the single Al or Fe anode, the Al-Fe combined anode had more advantages. Considering both removal efficiency and energy consumption, the optimum anode combination for the EC–filtration system was 3Al + Fe. When the anode was 3Al + Fe, the HRT was 4.5 min and the filter pore size was 45 μm, the EER of the total number of Vibrio, CODmn and TN removed by the EC–filtration system was 54.47%, 67.03% and 61.97%, respectively.

(4) With the increase of HRT and the decrease of filtration pore size, the enhanced effect of the EC process on the filtration equipment was more obvious. When the HRT was 4.5 min and the filter pore size was 45 μm, the maximum removal efficiency for Vibrio, CODmn and TN increased by 3.31, 5.39 and 4.57 times compared with the control group, respectively.

**REFERENCES**

Al-Qodah, Z., Al-Qudah, Y. & Omar, W. 2019 On the performance of electrocoagulation-assisted biological treatment processes: a review on the state of the art. Environmental Science and Pollution Research 1–25. https://doi.org/10.1007/s11356-019-06053-6.

Aoudj, S., Khelifa, A., Drouiche, N., Belkada, R. & Miroud, D. 2015 Simultaneous removal of chromium(VI) and fluoride by electrocoagulation–electroflotation: application of a hybrid Fe-Al anode. Chemical Engineering Journal 267, 153–162. https://doi.org/10.1016/j.cej.2014.12.081.

APHA 1998 Standard Methods for the Examination of Water and Wastewater. American Public Health Association/American Water Works Association/Water Environment Federation, Washington DC, USA.

Ben-Asher, R. & Lahav, O. 2019 Minimization of THM formation in seawater-fed recirculating aquaculture systems operated with electrochemical NH3 removal. Aquaculture 502, 162–175. https://doi.org/10.1016/j.aquaculture.2018.12.025.

Ben-Sasson, M., Zidon, Y., Calvo, R. & Adin, A. 2015 Enhanced removal of natural organic matter by hybrid process of electrocoagulation and dead-end microfiltration. Chemical Engineering Journal 232, 338–345. https://doi.org/10.1016/j.cej.2013.07.101.

Chellam, S. & Sari, M. A. 2016 Aluminum electrocoagulation as pretreatment during microfiltration of surface water containing NOM: a review of fouling, NOM, DBP, and virus control. Journal of Hazardous Materials 304, 490–501. https://doi.org/10.1016/j.jhazmat.2015.10.054.

Ebeling, J. M., Welsh, C. F. & Rishel, K. L. 2006 Performance evaluation of an inclined belt filter using coagulation/flocculation aids for the removal of suspended solids and phosphorus from microscreen backwash effluent. Aquacultural Engineering 35 (1), 61–77. https://doi.org/10.1016/j.aquaeng.2005.08.006.

García-Morales, M. A., Juárez, J. C. G., Martínez-Gallegos, S., Roa-Morales, G., Peralta, E., del Campo López, E. M., Barrera-Díaz, C., Miranda, V. M. & Blancas, T. T. 2018 Pretreatment of real wastewater from the chocolate manufacturing industry through an integrated process of electrocoagulation and sand filtration. International Journal of Photoenergy 2018, 1–7. https://doi.org/10.1155/2018/2146751.

Ghernaout, D., Touahnia, M. & Aichouni, M. 2019 Disinfecting water: electrocoagulation as an efficient process. Applied Engineering 3, 1–12. https://doi.org/10.11648/j.aee.20190301.11.

Govindan, K., Oren, Y. & Noel, M. 2014 Effect of dye molecules and electrode material on the settling behavior of flocs in an electrocoagulation induced settling tank reactor (EISTR). Separation and Purification Technology 133, 396–406. https://doi.org/10.1016/j.seppur.2014.04.046.

Heffron, J., McDermid, B., Maher, E., McNamara, P. J. & Mayer, B. K. 2019 Mechanisms of virus mitigation and suitability of bacteriophages as surrogates in drinking water treatment by iron electrocoagulation. Water Research 163, 114877. http://doi.org/10.1016/j.watres.2019.114877.
Mook, W. T., Chakrabarti, M. H., Aroua, M. K., Khan, G. M. A., Medel, A., Ramírez, J. A., Cárdenas, J., Sirés, I. & Meas, Y. Linares-Hernández, I., Barrera-Díaz, C., Roa-Morales, G., Bilyeu, B. Kimura, K., Tanaka, K. & Watanabe, Y. Ndjomgoue-Yossa, A. C., Nanseu-Njiki, C. P., Kengne, I. M. & Nazlabadi, E., Moghaddam, M. R. A. & Karamati-Niaragh, E. Moussa, D. T., El-Naas, M. H., Nasser, M. & Al-Marri, M. J. Mook, W. T., Chakrabarti, M. H., Aroua, M. K., Khan, G. M. A., Ali, B. S., Islam, M. S. & Hassan, M. A. Medel, A., Ramírez, J. A., Cárdenas, J., Sirés, I. & Meas, Y. 2019 Evaluating the electrochemical and photoelectrochemical production of hydroxyl radical during electrocoagulation process. Separation and Purification Technology 208, 59–67. https://doi.org/10.1016/j.seppur.2018.05.021. Mook, W. T., Chakrabarti, M. H., Aroua, M. K., Khan, G. M. A., Ali, B. S., Islam, M. S. & Hassan, M. A. 2012 Removal of total ammonia nitrogen (TAN), nitrate and total organic carbon (TOC) from aquaculture wastewater using electrochemical technology: a review. Desalination 285, 1–13. https://doi.org/10.1016/j.desal.2011.09.029. Moussa, D. T., El-Naas, M. H., Nasser, M. & Al-Marri, M. J. 2017 A comprehensive review of electrocoagulation for water treatment: potentials and challenges. Journal of Environmental Management 186, 24–41. https://doi.org/10.1016/j.jenvman.2016.10.032. Nazlabadi, E., Moghaddam, M. R. A. & Karamati-Niaragh, E. 2019 Simultaneous removal of nitrate and nitrite using electrocoagulation/floatation (ECF): a new multi-response optimization approach. Journal of Environmental Management 250, 109489. https://doi.org/10.1016/j.jenvman.2019.109489. Ndjomgoue-Yossa, A. C., Nanseu-Njiki, C. P., Kengne, I. M. & Ngameni, E. 2015 Effect of electrode material and supporting electrolyte on the treatment of water containing Escherichia coli by electrocoagulation. International Journal of Environmental Science and Technology 12 (6), 2103–2110. https://doi.org/10.1007/s13762-014-0609-9.

Hu, C., Sun, J., Wang, S., Liu, R., Liu, H. & Qu, J. 2017 Enhanced efficiency in HA removal by electrocoagulation through optimizing flocs properties: role of current density and pH. Separation and Purification Technology 175, 248–254. https://doi.org/10.1016/j.seppur.2016.11.036. Kimura, K., Tanaka, K. & Watanabe, Y. 2014 Microfiltration of different surface waters with/without coagulation: clear correlations between membrane fouling and hydrophilic biopolymers. Water Research 49, 434–443. https://doi.org/10.1016/j.watres.2013.10.030. Lakshmi, P. M. & Sivashanmugam, P. 2015 Treatment of oil tanning effluent by electrocoagulation: influence of ultrasound and hybrid electrode on COD removal. Separation and Purification Technology 116, 378–384. https://doi.org/10.1016/j.seppur.2013.05.026. Linares-Hernández, I., Barrera-Díaz, C., Ros-Morales, G., Bilyeu, B. & Urena-Nunez, F. 2009 Influence of the anodic material on electrocoagulation performance. Chemical Engineering Journal 148 (1), 97–105. https://doi.org/10.1016/j.cej.2008.08.007. Medel, A., Ramírez, J. A., Cárdenas, J., Sirés, I. & Meas, Y. 2019 Evaluating the electrochemical and photoelectrochemical production of hydroxyl radical during electrocoagulation process. Separation and Purification Technology 208, 59–67. https://doi.org/10.1016/j.seppur.2018.05.021. Van Rijn, J. 2013 Waste treatment in recirculating aquaculture systems. Aquacultural Engineering 53, 49–56. https://doi.org/10.1016/j.aquaeng.2012.11.010. Wold, P. A., Holan, A. B., Ofie, G., Attrimadali, K., Bakke, I., Vadstein, O. & Leiknes, T. O. 2014 Effects of membrane filtration on bacterial number and microbial diversity in marine recirculating aquaculture system (RAS) for Atlantic cod (Gadus morhua L.) production. Aquaculture 422, 69–77. https://doi.org/10.1016/j.aquaculture.2013.11.019. Xiao, R., Wei, Y., An, D., Li, D., Ta, X., Wu, Y. & Ren, Q. 2019 A review on the research status and development trend of equipment in water treatment processes of recirculating aquaculture systems. Reviews in Aquaculture 11 (3), 863–895. https://doi.org/10.1111/raq.12270. Xie, S., Yuan, S., Liao, P., Tong, M., Gan, Y. & Wang, Y. 2017 Iron-anode enhanced sand filter for arsenic removal from tube well water. Environmental Science & Technology 51 (2), 889–896. https://doi.org/10.1021/acs.est.6b04387. Zhao, S., Huang, G., Cheng, G., Wang, Y. & Fu, H. 2014 Hardness, COD and turbidity removals from produced water by electrocoagulation pretreatment prior to reverse osmosis membranes. Desalination 344, 454–462. https://doi.org/10.1016/j.desal.2014.04.014.

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