# Integration of Copperas and *Moringa oleifera* Seeds as Hybrid Coagulant for Turbidity and Ammonia Removal from Aquaculture Wastewater

**Sofiah Hamzah[1], Nazaitulshila Rasit[1*], Nurul Aqilah Mohamad[1], Mohammad Hakim Che Harun[1], Alyza Azzura Abd Rahman Azmi[1,2], Nur Hanis Hayati Hairom[3], Ahmad Ariff Fahmi Mustofa[4], Mohd Salleh Amri Zahid[4], Norhafiza Ilyana Yatim[5], and Nor Azman Kasan[5]**

[1]*Environmental Sustainable Material Research Interest Group, Faculty of Ocean Engineering, Technology, and Informatics, Universiti Malaysia Terengganu, Kuala Nerus 21030, Malaysia*

[2]*Faculty of Science and Marine Environment, Universiti Malaysia Terengganu, Kuala Nerus 21030, Malaysia*

[3]*Faculty of Engineering Technology, Universiti Tun Hussein Onn Malaysia, Hab Pendidikan Tinggi Pagoh, Km 1, Jalan Panchor, Muar 84600, Malaysia*

[4]*Venator Asia Sdn. Bhd., Teluk Kalung, Kemaman 24007, Malaysia*

[5]*Higher Institution Centre of Excellence (HICoE), Institute of Tropical Aquaculture and Fisheries, Universiti Malaysia Terengganu, Kuala Nerus 21030, Malaysia*

*** Corresponding author:**

*email: nazaitulshila@umt.edu.my*

*Received: April 4, 2023*
*Accepted: September 12, 2023*

**DOI:** 10.22146/ijc.83634

**Abstract:** *The rapid development of the aquaculture industry has contributed to the high amount of nutrients in wastewater that subsequently led to eutrophication and deterioration of water quality. Aquaculture wastewater consists of uneaten fish feed, fecal and other excretion or residue of chemicals used. Thus, this study aimed to evaluate the performance of hybrid coagulants of* Moringa oleifera *(MO) and copperas for aquaculture wastewater treatment. In this present study, different formulations of MO and copperas were explored in the coagulation treatment of aquaculture wastewater using a jar test experiment. The FTIR and SEM analysis are used to determine the morphology and surface of MO. This study focuses on the effect of coagulant aids formulation, coagulant dosage, the effect of initial pH and coagulation time on turbidity and ammonia removal in the coagulation of aquaculture wastewater. The finding shows that the highest removal of turbidity and ammonia was obtained with the use of 80% MO and 20% copperas at the condition of initial pH of 6 at 20 min of coagulation time, with the highest percentage removal of 66% and 91%, respectively. The coagulation isotherm of hybrid coagulant 80:20 is well described with the Freundlich isotherm model which describes the surface heterogeneity.*

**Keywords:** *aquaculture wastewater; coagulation-flocculation; copperas;* Moringa oleifera

## INTRODUCTION

The aquaculture industry is one of the fastest-growing industries all around the world. However, the long-term sustainability of the aquatic environment has raised worries over the environmental impact of this imperative sector because of its negative impact on aquatic ecology and systems. This is due to aquaculture intensification requiring the use of highly nutritious feeds and other compounds that produce waste that is hard to curb and is harmful to aquatic life in most cases [1]. Most aquaculture systems produce great amounts of wastewater containing compounds such as suspended solids, nitrogen, and phosphorus [2]. Any discharge of wastewater into aquatic ecosystems in the form of effluents may lead to changes in the receiving environment. High organic load in wastewater from aquaculture will contribute to the eutrophication of receiving water bodies, which causes a lot of havoc in aquatic ecosystems on biodiversity. The aquaculture sector contains a complex mixture of pollutants.

The most common toxic pollutants are ammonia, nitrogen, phosphorus, pathogens, and ammoniacal nitrogen. Ammonia is a colorless gas with a strong scent which contains compounds of nitrogen and hydrogen [3]. Ammonia in water is converted to nitrite and nitrate through the nitrification process by two bacteria, *Nitrosomonas* and *Nitrobacter* [4]. When the levels of nitrogenous compounds (ammonia, nitrite, and nitrate) exceed the prescribed limits can affect aquatic life. One of the main reasons for unexplained losses in hatcheries is the toxicity of ammonia. Although the ammonia molecule is an essential nutrient, excessive amounts of ammonia may accumulate and alter the organism's metabolism or raise the pH level in the body.

Because hazardous contaminants and other factors affecting water quality have a direct impact on the effectiveness of feed and growth rates for fish health and survival. Due to the evident cause of environmental climate change, these characteristics have not remained constant for a while now. There is a need for improvement in the development and transfer of technology in catfish breeding and hatchery to stabilize the supply of fish.

Treatment of wastewater for pollutant removal is very crucial since it can cause a threat to potable drinking water sources, fisheries, and recreational water bodies. Membrane technology [5-6], liquid membrane [7], and adsorption [8-11] are a few of the treatment techniques and technologies utilized to treat wastewater. However, certain technologies already in use still struggle to comply with the standard of discharge, the restrictions that have been set up, difficulty in scaling up, and come at a high cost. Several studies have been conducted for efficient systems for wastewater treatment, including constructed wetland technology [12], the use of fish meal in removing heavy metals [13], microalgae [14], and others [15]. It is believed that wetland was one of the most cost-effective and well-known treatment methods that have been established [16].

Another method for treating aquaculture wastewater is coagulation-flocculation which helps to remove suspended solids, turbidity, and other inorganic compounds that can disturb the water quality and water ecosystem. Coagulation-flocculation is considered an excellent pre-treatment method for wastewater since there is low energy required, the cost is reasonable, and it is easy to operate. The removal mechanism is primarily caused by cationic hydrolysis products' ability to charge-neutralize negatively charged colloids, which facilitates the formation of microflocs by encouraging early colloidal particle aggregation [17]. Two types of coagulants used for the removal of pollutants are either natural coagulants or chemical coagulants. Using chemical coagulants such as aluminum sulphate (alum) is a very common treatment method that is used mainly in the water treatment process. Inorganic coagulants such as alum, ferrous sulphate, and ferric chloride has been used to recover pollutant removal through coagulation-flocculation and was demonstrated successfully efficient in achieving the aims. However, excessive use of chemical coagulants may be unacceptable for aquaculture purposes as they have negative effects on aquatic life.

It has been demonstrated that a natural coagulant like *Moringa oleifera* (MO) can be utilized to treat effluent from aquaculture mainly to remove ammoniacal nitrogen, phosphorus, and turbidity [18-20]. Treatment sequence of wastewater using MO seeds by coagulation–flocculation to sedimentation-filtration showed maximum turbidity and elimination of suspended materials. Powdered seeds produce less volume of sludge and promote the removal of chemical oxygen demand (COD) [21]. The seeds have successful properties of coagulation that were verified in laboratory studies. The seeds do not hurt humans or animals. It is also very effective in reducing the presence of microorganisms in raw water. The physicochemical treatment shows encouraging pollutant mitigation efficiency. These may be due to relatively active flocs produced at the relatively high dosage of seeds. The extreme flocs are more likely to resist the hydraulic shear forces encountered during filtration.

Water handled with MO seeds is therefore better compared to alum. The seeds did not affect the water's pH value significantly, but they served as a pH corrector. For comparison, when treated with alum, the pH value decreased to 4.2. It has shown that MO seeds will be more economical and environmentally important for wastewater treatment than aluminum or other chemicals. A by-product of the titanium industry, copperas is chemically known as ferrous sulfate. By lowering the electrostatic surface charge in an acidic or alkaline pH range, the presence of this hydrolyzed metal species can encourage the agglomeration of particles in wastewater. Copperas and MO seeds have lately gained interest in water and wastewater treatment technologies due to their exceptional efficacy and cost-effectiveness. Hybrid components are novel materials with a high potential to treat wastewater when compared to inorganic coagulants and organic flocculants, respectively [22]. There have recently been many previous works of coagulation/flocculation techniques for treating water and wastewater. However, the hybrid materials chosen in this study have not been extensively compared and discussed.

The ultimate objective of this present study is to determine the efficiency of hybrid coagulants (copperas and MO seed) for aquaculture wastewater treatment. Copperas and MO were characterized in terms of morphology and chemical composition, using scanning electron microscopy (SEM), Fourier transform infrared (FTIR), and X-ray diffraction (XRD). The removal of turbidity and ammonia with this hybrid coagulant is being studied at various formulations, dosages, and initial pH levels. The Freundlich and Langmuir isotherm models were utilized in a coagulation equilibrium investigation, and ANOVA was used to statistically validate the data.

## ■ EXPERIMENTAL SECTION

### Materials

The aquaculture wastewater used in this study was obtained from the discharged point of a catfish pond in UMT Hatchery. Aquaculture effluent that was discharged was collected in 40 L plastic containers in an acceptable amount. Before being taken to the lab, the containers were labelled and sealed appropriately. Prior to use, this wastewater was maintained below 4 °C to prevent sample degradation brought on by microbial activity.

Venator Asia Sdn. Bhd., Teluk Kalung, Terengganu, Malaysia, provided the copperas while the analytical grade of calcium hydroxide ($Ca(OH)_2$), sodium hydroxide (NaOH), and hydrochloric acid (HCl) were acquired from Sigma-Aldrich in the United States.

### Instrumentation

A JEOL JSM-6360 LA model SEM was used to analyze the surface morphology of copperas. The sample was transferred to the microscope and viewed at 2,000× magnification after sputtering with gold. FTIR analysis was used to identify the functional groups found in copperas (Shimadzu/IRTracer-100 model). In a sample holder, an FTIR scan was carried out at frequencies ranging from 4,000 to 400 $cm^{-1}$ with a spectral resolution of 4 $cm^{-1}$.

### Procedure

#### *Preparation of hybrid coagulant solution copperas/***Moringa oleifera**

MO was rinsed with distilled water slowly and dried at 35 °C in an oven, then grounded to obtain a fine powder. The MO powder was dissolved in distilled water to a final concentration of 1000 mg/L and mixed for 30 min using a stirrer to get a uniform solution and the extract was then passed through a muslin cloth. This solution was prepared freshly to avoid the reduction in its coagulation efficiency, and it was kept in a cool place at around 20 °C to avoid viscosity and pH changes. The prepared MO solution was integrated with 1000 ppm copperas solution based on different ratios (100% MO, 100% copperas, 50:50, 80:20 and 20:80 MO:copperas) to form hybrid coagulant.

#### *Coagulation-flocculation process (jar test experiments)*

To coagulate aquaculture effluent, flocculation tests were performed using a standard jar-test apparatus with six paddle rotors. For each test run, a 1 L beaker containing 500 mL of wastewater was used. The ambient temperature for all tests was between 24 and 28 °C. Using either an acid (1.0 M HCl) or a base (1.0 M NaOH), the pH value was changed to the desired value. Different coagulant dosages (40–200 mg/L) and the sample's initial pH (3–8) were used in the jar test. The aquaculture wastewater sample was subjected to 150 rpm of rapid mixing for 10 min, then 20 min of slow mixing at 25 rpm. After coagulation, samples of the treated wastewater were taken for analysis from a depth of 2 cm. Eq. (1) was used to calculate the turbidity and ammonia removal efficiency;

$$\text{Removal } (\%) = \frac{C_i - C_f}{C_i} \times 100\% \quad (1)$$

where $C_i$ and $C_f$ are the initial and final readings, respectively, of turbidity and ammonia.

### *Determination of aquaculture wastewater characteristics*

The characterization of aquaculture wastewater involves two methods: field measurement and laboratory experiments. The tests involved were pH, turbidity, and ammoniacal nitrogen (HACH Method 10031). The pH and turbidity were tested using pH and turbidity meters (Orion AQ3010), respectively, while ammoniacal nitrogen was tested using DR900. All testing was performed in triplicates and the results were presented as the average of three values.

### *Coagulation equilibrium*

The isotherm model, which can also explain the interaction between the coagulant and the sample of aquaculture effluent, can describe coagulation equilibrium. In order to assess the efficacy of two isotherm models, the Freundlich and Langmuir models were used to determine whether coagulation results from multilayer or monolayer development of the coagulant particles. By setting the coagulant and its dosage for wastewater samples at the optimum condition for turbidity reduction, the adsorption experiments were carried out.

Surface heterogeneity of the adsorbates is introduced by the Freundlich isotherm model with a variable energy level distribution of adsorption [23-24]. This model shows whether the coagulant particles have adsorbed the adsorbent in a multilayer formation [23,25]. According to Eq. (2), this isotherm model is an empirical representation of the Langmuir isotherm model.

$$q_e = K_f \left(C_e\right)^{1/n} \quad (2)$$

The linear form of the Freundlich isotherm is presented in Eq. (3):

$$\log\log q_e = \log K_f + \frac{1}{n}\log C_e \quad (3)$$

where $q_e$ is the coagulation adsorption capacity (mg/mg), $K_f$ is the Freundlich affinity coefficient (L/mg), $C_e$ is the equilibrium concentrations of turbidity in aquaculture wastewater, and n is the exponential constant of the Freundlich model. Hussain et al. [25] stated that the adsorption intensity, $q_e$, usually decreases as the values of 1/n increase and decreases as $K_f$ decreases. Lin et al. [24] mentioned that favorable adsorption would occur if the value of 1/n is less than 1.

The surface homogeneity of the adsorbent serves as the foundation for the Langmuir isotherm model [23]. This model explains how a monolayer form on the adsorbate's adsorbent surface [24-26]. According to Nhut et al. [18], this model also demonstrates that once the adsorbent is linked to the particular adsorbent site, no further adsorbent molecule can occupy the adsorbent site. Once a sorbate molecule resides at a location, no more adsorptions will take place there. The Langmuir isotherm model suggests that the adsorbed molecules do not interact with one another in order to explain adsorption in an aqueous solution [22]. The Langmuir isotherm model was determined as Eq. 4.

$$q_e = \frac{\left(abC\right)_e}{\left(1 + aC\right)_e} \quad (4)$$

The linear form of the Langmuir isotherm is presented in Eq. (5):

$$\frac{1}{q_e} = \frac{1}{\left(abC\right)_e} + \frac{1}{b} \quad (5)$$

where a is the Langmuir constant and b is the maximum coagulation value

### *Statistical analysis*

An important step in carrying out various statistical procedures is statistical data analysis. ANOVA is a statistical technique for testing a hypothesis about the model's input parameters by breaking down the total variation in a set of data into components linked to certain sources of variation. ANOVA's null hypothesis states that there are no significant differences between the groups that were chosen, but the alternative hypothesis holds that there are at least one or more significant differences between the groups that were chosen [22].

## ■ RESULTS AND DISCUSSION

### Characteristics and Properties of Hybrid Coagulant

Fig. 1 shows the surface morphology of MO and copperas viewed under SEM. The morphology of MO (Fig. 1(a)) shows a heterogeneous and relatively porous matrix. This structure facilitates ion adsorption processes due to the interstices and, more importantly, the presence of the seed protein component. The coagulant surface tends to be irregular and rough at some points and at others smooth. Small pores were found around the edges, indicating the possibility of sorption at a smaller magnitude. Thus, based on these characteristics, it can be concluded that this material has an adequate morphological profile for retaining metal ions.

Copperas morphology shows that this compound has much asperity and is more coarsely grained, creating a large contact area [22]. The depicted image also shows a compact gel network structure which is more favorable for coagulating colloidal particles and forming bridge aggregation between flocs than a branched structure [27].

Using FTIR analysis, it was possible to determine the chemical composition and functional group of copperas and MO, as shown in Fig. 2. Generally, all spectra

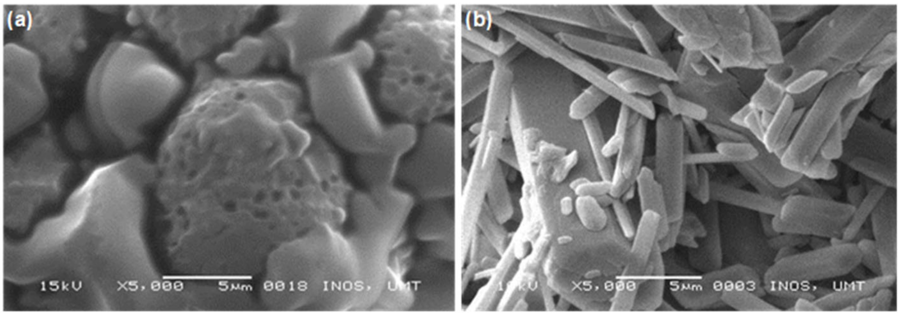


**Fig 1.** Morphology of (a) MO and (b) copperas

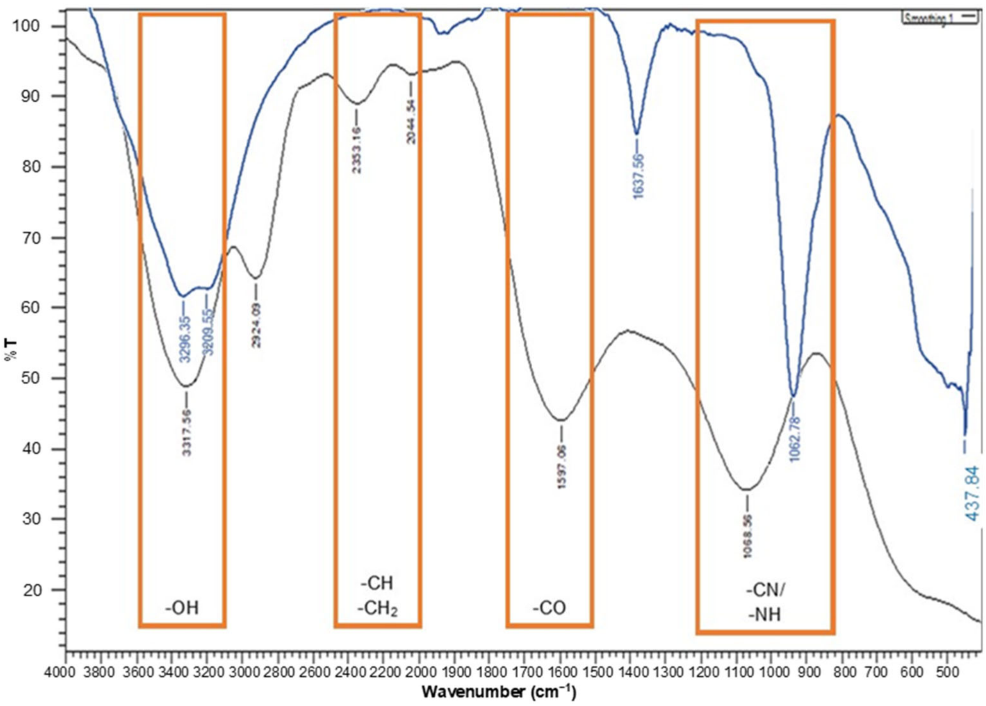


**Fig 2.** FTIR analysis of MO (black line) and copperas (blue line) [22]

display the same profile, which is a broad band attributed to O–H stretching with a center at 3,317 $cm^{-1}$. This functional group is mostly found in the fatty acid and protein structures of MO seeds. The N–H stretching of amide groups also contributes to this region because of the high protein content in seeds. The C–H of the $CH_2$ group found in fatty acids is stretched symmetrically and asymmetrically, resulting in peaks at 2,353 and 2,044 $cm^{-1}$, respectively. Three strong bands with the C–O bond stretching are located between 1,597 and 1,400 $cm^{-1}$. The fatty acid and protein structures contained the carbonyl group. It is possible to attribute the presence of a peak at 1,068 $cm^{-1}$ to C–N stretching or N–H deformation. The existence of this band demonstrates the MO seeds' protein structure. In the treatment of contaminated water, the high protein content of MO is functional as an electrical cationic.

### Physical and Chemical Characteristics of Aquaculture Wastewater

The initial parameters of aquaculture effluent that came from a catfish pond are shown in Table 1. According to high levels of turbidity and ammonia, the aquaculture effluent needed additional treatment before being discharged to a neighboring watercourse. However, research on the technology used to remove ammonia from aquaculture wastewater is relatively limited. Since the coagulant and coagulant aids employed in this study have strong removal capacities for this type of pollutant, ammonia removal is one of the parameters that has been focused on in the present study.

### Coagulation of Aquaculture Wastewater Using Hybrid Coagulant of MO and Copperas

Coagulation of aquaculture wastewater was initially done using different formulations of hybrid coagulant MO/copperas at different dosages varied from 40 to 200 mg/L with 40 mg/L intervals. Other parameters were kept constant, with rapid mixing at 150 rpm for 10 min, slow mixing at 25 rpm for 20 min, and settling down for 30 min.

### Effect of Hybrid Coagulant Formula and Coagulant Dosage

The coagulants used were native MO, native copperas and the integrated MO and copperas at different formulations (50:50, 80:20, 20:80). Fig. 3(a) and (b) display the results for this stage of the coagulation study.

**Table 1.** Physico-chemical characteristics for examined aquaculture wastewater

| Parameter | Value |
|---|---|
| Turbidity | 77.0 NTU |
| Ammonia | 2.3 mg/L |
| pH | 6.0 |

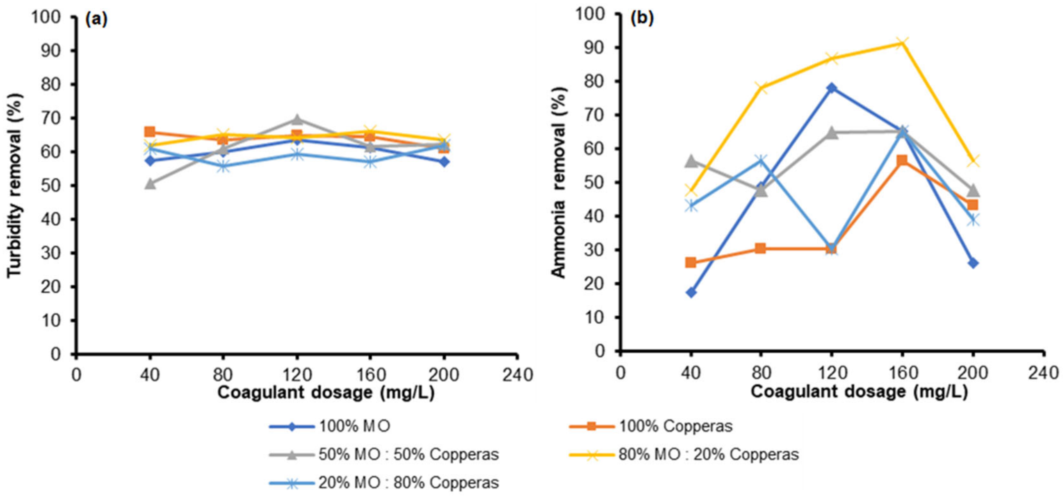


**Fig 3.** Effects of coagulant formulation and dosage on turbidity removal in aquaculture wastewater treatment (Experimental condition: pH 6, ambient temperature)

From observation, the best removal was obtained by coagulant formulation of 80% MO and 20% copperas at 160 mg/L coagulant dosage. At this condition, optimum turbidity and ammonia removal reached 66% and 91%, respectively. The use of 80% MO as a coagulant provides high cationic polyelectrolytes of protein, which destabilizes the particle charge in wastewater and eventually forms a bridge between the particles. These bridging particles then formed a large aggregate and precipitated during the sedimentation process [28]. The addition of 20% copperas during coagulation results in the formation of corresponding gel such as hydroxides and some positively charged mononuclear and polynuclear species. The positively charged $Fe^{2+}$ interacts with negatively charged colloidal particles present in the wastewater by charge neutralization mechanism and when these hydroxides and complexed hydroxides are settled under gravity, they sweep away and precipitate remaining uncharged or charged colloidal particles from the wastewater. However, at higher dosage of coagulant (more than 160 mg/L) the coagulation activity decreased due to the repulsion between the same charge from the coagulant protein of MO and $Fe^{2+}$ species. The excessive cationic polyelectrolytes were also cause destabilization of aggregates and deflocculation [29].

## Effect of Initial pH Wastewater

In this experiment, the pH of aquaculture wastewater was adjusted from pH 3 to 8 at fixed coagulant dosages 160 mg/L of 80:20 MO:copperas. The initial solution of pH was adjusted by $H_2SO_4$ and NaOH. The pH is one of the most important parameters affecting any wastewater treatment process. According to the result, pH 6 is identified as the optimum pH due to the highest removal efficiency of turbidity and ammonia. As seen from Fig. 4, the percentage removal efficiency of turbidity and ammonia had reached the highest removal of pollutants which was 66 and 91%, respectively. This dependence is closely related to the acid-base properties of various functional groups on the adsorbent surfaces [30]. The literature shows that an aqueous heterogeneous mixture of MO seeds presents various functional groups, mainly amino and acid groups. These groups are capable of interacting with metal ions, which is dependent on the pH. An increase in metal adsorption with increasing pH values can be explained by rivalry between the proton and metal ions for the same functional groups and a decrease in the positive surface charge resulting in a higher electrostatic attraction between the surface and the metal. The pH value of the point of zero charge for the proposed

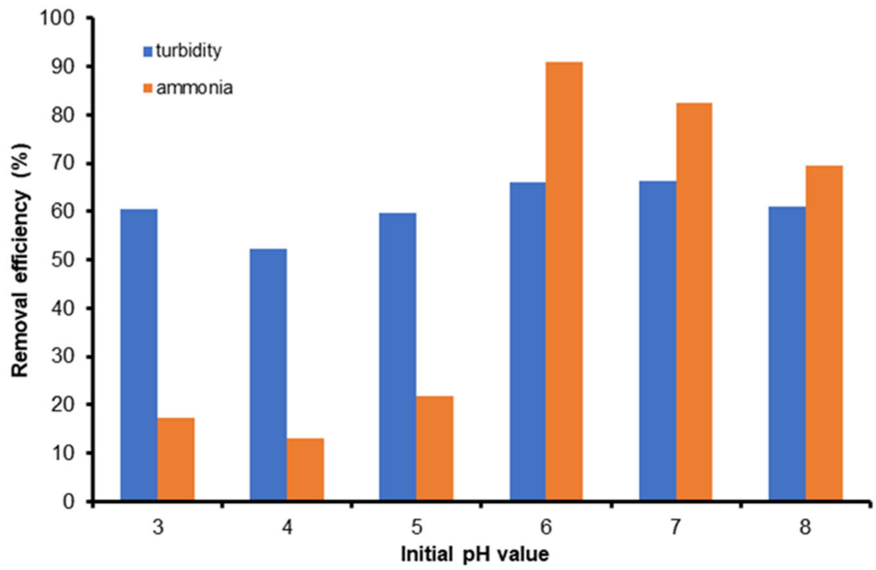


**Fig 4.** Effects of initial pH on turbidity and ammonia removal in aquaculture wastewater treatment (Experimental conditions: 160 mg/L of 80:20 MO:copperas at ambient temperature)

adsorbent was within the range of 6 to 7. Thus, above this pH range, the surface of the sorbent will be negatively charged and will adsorb positively charged species.

### Effect of Coagulation Time

Coagulation time is one of the crucial parameters in the coagulation/flocculation process to ensure that the coagulant is mixed properly in order to treat the sample at the optimal time [31]. This hybrid coagulant showed a good performance as the coagulation time was up to 45 min with the turbidity removal about 80%. For ammonia removal, 80% MO with 20% copperas gave a good performance at a lower coagulation time with 91% removal as represented in Fig. 5. Increasing the coagulation time resulted in poor removal of ammonia since the pollutant and the coagulant used had a low collision between them hence lowering down the formation of floc during sedimentation [32].

### ANOVA

ANOVA is a one-way test used in this study to validate the experimental data for the effect of coagulant dosage, initial pH and coagulation time for the best coagulant which was 80% MO with 20% copperas as tabulated in Tables 1-3. The analysis was accomplished by employing the final value of turbidity and ammonia. The findings showed that there was a significant difference (p-value < 0.05) in all parameters studied, indicating that

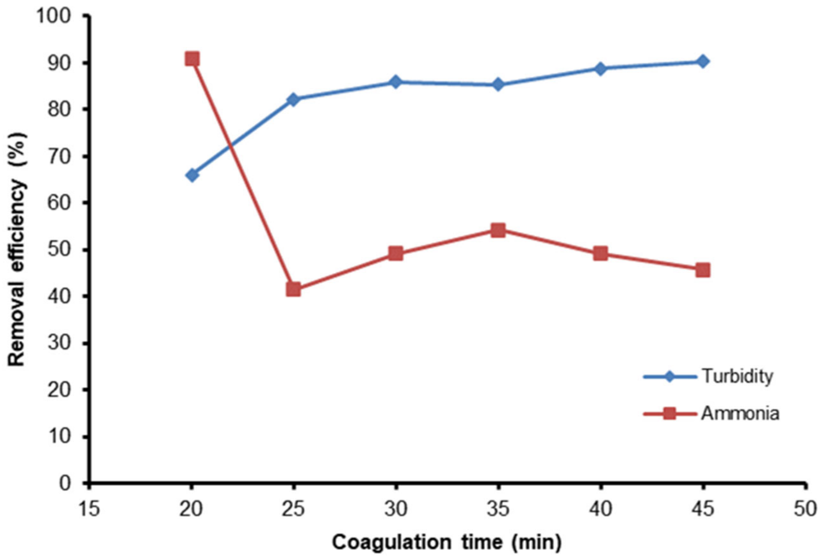


**Fig 5.** Effects of coagulation time on turbidity and ammonia removal in aquaculture wastewater treatment (Experimental conditions: 160 mg/L of 80:20 MO:copperas at ambient temperature and pH 6)

**Table 1.** ANOVA test of 80% of MO and 20% of copperas at different coagulant dosage for both turbidity and ammonia removal

| Source of variation | Sum of square (SS) | Degree of freedom (DF) | Mean square (MS) | F-test | p-value |
|---|---|---|---|---|---|
| | | | Turbidity removal | | |
| Treatment | 21372.130 | 1 | 21372.130 | 10.682 | 0.011385 |
| Error | 16006.090 | 8 | 2000.762 | | |
| Sum | 37378.220 | 9 | | | |
| | | | Ammonia removal | | |
| Treatment | 35617.020 | 1 | 35617.020 | 17.808 | 0.002916 |
| Error | 16000.770 | 8 | 2000.097 | | |
| Sum | 51617.800 | 9 | | | |

**Table 2.** ANOVA test of 80% of MO and 20% of copperas at different initial pH value for both turbidity and ammonia removal

| Source of variation | Sum of square (SS) | Degree of freedom (DF) | Mean square (MS) | F-test | p-value |
|---|---|---|---|---|---|
| | | | Turbidity removal | | |
| Treatment | 1948.2010 | 1 | 1948.201 | 253.0514 | 1.99E-08 |
| Error | 76.9883 | 10 | 7.698833 | | |
| Sum | 2025.1890 | 11 | | | |
| | | | Ammonia removal | | |
| Treatment | 51.6675 | 1 | 51.6675 | 26.1541 | 0.000455 |
| Error | 19.7550 | 10 | 1.9755 | | |
| Sum | 71.4225 | 11 | | | |

**Table 3.** ANOVA test of 80% of MO and 20% of copperas at different coagulation time for both turbidity and ammonia removal

| Source of Variation | Sum of square (SS) | Degree of freedom (DF) | Mean square (MS) | F-test | p-value |
|---|---|---|---|---|---|
| | | | Turbidity removal | | |
| Treatment | 1176.1200 | 1 | 1176.1200 | 17.5159 | 0.001872 |
| Error | 671.4600 | 10 | 67.1460 | | |
| Sum | 1847.5800 | 11 | | | |
| | | | Ammonia removal | | |
| Treatment | 709.9408 | 1 | 709.9408 | 9.0269 | 0.013242 |
| Error | 786.4683 | 10 | 78.6468 | | |
| Sum | 1496.4090 | 11 | | | |

**Table 4.** Isotherm model for aquaculture wastewater treated with 80:20 MO:copperas

| Isotherm model | Unit | 80:20 (MO:copperas) |
|---|---|---|
| Langmuir | a | 18.93706 |
| | b | 0.033385 |
| | $R^2$ | 0.499216 |
| Freundlich | n | 0.933595 |
| | $K_f$ | 0.717376 |
| | $R^2$ | 0.992175 |

the null hypothesis should be rejected which assumes the dosage and initial pH (treatment) were equal for all the parameter removals. This means that the data have a significant difference between the means of groups.

## Coagulation Equilibrium

Coagulation equilibrium studies were to evaluate their performance and determine whether coagulation occurs due to multilayer or monolayer formation of the coagulant particles in describing the interaction between the used coagulant and the aquaculture wastewater sample. The adsorption experiments were conducted by fixing the coagulant and its dosage for wastewater samples at the optimum condition for ammonia removal.

Table 4 shows the coagulation equilibrium studies performed by using both Langmuir and Freundlich isotherm models. As tabulated in Table 4, the Langmuir isotherm model shows the a and b values are positive and stipulated that the 80:20 MO:copperas based on monolayer adsorption supported by Hussain et al. [25]. Fig. 6 represents the linear form of both isotherm models, Langmuir and Freundlich. The coagulation-adsorption analysis revealed that 80:20 MO:copperas showed monolayer formation of the coagulant, which is well-described in the Freundlich Isotherm model, which describes surface heterogeneity.

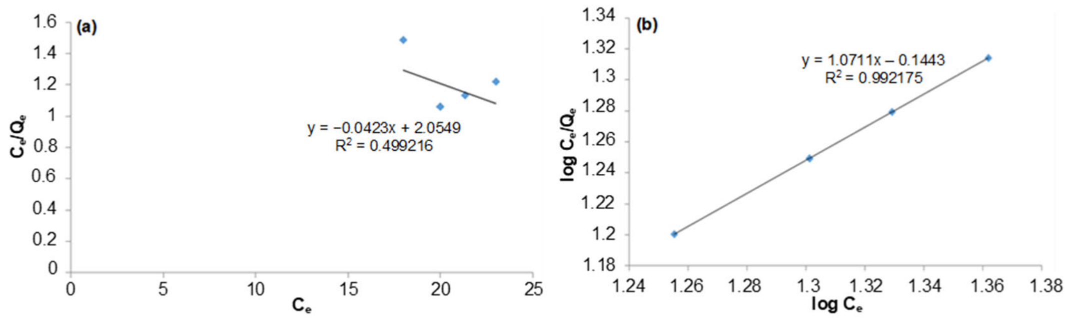


**Fig 6.** (a) Langmuir and (b) Freundlich Isotherm model for 80:2 MO:copperas in treating aquaculture wastewater

## CONCLUSION

Overall results showed that for the aquaculture wastewater sample employing 80:20 MO:copperas as a hybrid coagulant with the optimal conditions for pollutant removal in aquaculture wastewater were 160 mg/L of coagulant dose, pH 6, and 20 min of coagulation time. This ratio of MO:copperas shows monolayer development of the coagulant, which is well-described in the Freundlich isotherm model, according to the coagulation-adsorption analysis. These results could be a baseline research for creating a pilot plant for effective hatchery wastewater treatment as well as important information for the aquaculture sector in implementing efficient chemical treatment.

## ACKNOWLEDGMENTS

The authors would like to thank Universiti Malaysia Terengganu for the funding of TAPE-RG (VOT 55325) and Venator Asia Sdn. Bhd. for the for the contribution and support of this project.

## AUTHOR CONTRIBUTIONS

Sofiah Hamzah, Nurul Aqilah Mohamad, and Mohammad Hakim Che Harun conducted the experiment. Alyza Azzura Abd Rahman Azmi, Nur Hanis Hayati Hairom, Ahmad Ariff Fahmi Mustoffa, and Mohd Salleh Amri Zahid conducted the calculations. Norhafiza Ilyana Yatim revised the literature review and analysis section. Nor Azman contributed to the analysis section and proofread the manuscript. Sofiah Hamzah and Nazaitulshila Rasit wrote and revised the overall manuscript prior to publication. All authors agreed to the final version of this manuscript.

## REFERENCES

[1] Omitoyin, B.O., Ajani, E.K., Okeleye, O.I., Akpoilih, B.U., and Ogunjobi, A.A., 2017, Biological treatments of fish farm effluent and its reuse in the culture of Nile tilapia (*Oreochromis niloticus*), *J. Aquacult. Res. Dev.*, 8 (2), 1000469.

[2] Turcios, A.E., and Papenbrock, J., 2014, Sustainable treatment of aquaculture effluents—What can we learn from the past for the future?, *Sustainability*, 6 (2), 836–856.

[3] Li, Z., Wang, C., Qiu, J., Ma, Y., Wang, C., Sun, X., Li, K., Ning, P., and Wang, F., 2023, Advances in selective catalytic oxidation of ammonia ($NH_3$-SCO): A review of catalyst structure-activity relationship and design principles, *Chin. Chem. Lett.*, 35 (1), 108432.

[4] Romano, A., Ortiz, I., and Urtiaga, A.M., 2021, Comprehensive kinetics of electrochemically assisted ammonia removal in marine aquaculture recirculating systems, *J. Electroanal. Chem.*, 897, 115619.

[5] Ali, A., Awang, M., Mat, R., Johari, A., Kamaruddin, M.J., and Wan Sulaiman, W.R., 2014, Influence of hydrophilic polymer on pure water permeation, permeability coefficient, and porosity of polysulfone blend membranes, *Adv. Mater. Res.*, 931-932, 168–172.

[6] Rakcho, Y., Mouiya, M., Bouazizi, A., Abouliatim, Y., Sehaqui, H., Mansouri, S., Benhammou, A., Hannache, H., Alami, J., and Abourriche, A., 2023, Treatment of seawater and wastewater using a novel low-cost ceramic membrane fabricated with red clay and tea waste, *Arabian J. Chem.*, 16 (11), 105277.

[7] Othman, N., Chui Heng, L., Mohamed Noah, N.F., Zing Yi, O., Jusoh, N., Nasruddin, N.A., Ali, N., and Hamzah, S., 2015, Removal of phenol from wastewater by supported liquid membrane process, *Jurnal Teknologi*, 74 (7), 117–121.

[8] Mohd Azmi, L.H., Williams, D.R., and Ladewig, B.P., 2021, Polymer-assisted modification of metal-organic framework MIL-96 (Al): Influence of HPAM concentration on particle size, crystal morphology and removal of harmful environmental pollutant PFOA, *Chemosphere*, 262, 128072.

[9] Mustapha, S., Shuaib, D.T., Ndamitso, M.M., Etsuyankpa, M.B., Sumaila, A., Mohammed, U.M., and Nasirudeen, M.B., 2019, Adsorption isotherm, kinetic and thermodynamic studies for the removal of Pb(II), Cd(II), Zn(II) and Cu(II) ions from aqueous solutions using *Albizia lebbeck* pods, *Appl. Water Sci.*, 9 (6), 142.

[10] Hamzah, S., Yatim, N.I., Alias, M., and Ali, A., 2019, Extraction of hydroxyapatite from fish scales and its integration with rice husk for ammonia removal in aquaculture wastewater, *Indones. J. Chem.*, 19 (4), 1019–1030.

[11] Ali, A., Ing, A.W.C., Abdullah, W.R.W., Hamzah, S., and Azaman, F., 2020, Preparation of high-performance adsorbent from low-cost agricultural waste (peanut husk) using full factorial design: Application to dye removal, *Biointerface Res. Appl. Chem.*, 10 (6), 6619–6628.

[12] Wu, H., Fan, J., Zhang, J., Ngo, H.H., Guo, W., Liang, S., Hu, Z., and Liu, H., 2015, Strategies and techniques to enhance constructed wetland performance for sustainable wastewater treatment, *Environ. Sci. Pollut. Res.*, 22 (19), 14637–14650.

[13] Zhang, S., Ding, J., Razanajatovo, R.M., Jiang, H., Zou, H., and Zhu, W., 2019, Interactive effects of polystyrene microplastics and roxithromycin on bioaccumulation and biochemical status in the freshwater fish red tilapia (*Oreochromis niloticus*), *Sci. Total Environ.*, 648, 1431–1439.

[14] Han, P., Lu, Q., Fan, L., and Zhou, W., 2019, A Review on the use of microalgae for sustainable aquaculture, *Appl. Sci.*, 9 (11), 2377.

[15] Chatla, D., Padmavathi, P., and Srinu, G., 2020, "Wastewater Treatment Techniques for Sustainable Aquaculture" in *Waste Management as Economic Industry Towards Circular Economy*, Eds. Ghosh, S.K., Springer Singapore, Singapore, 159–166.

[16] Waly, M.M., Ahmed, T., Abunada, Z., Mickovski, S.B., and Thomson, C., 2022, Constructed wetland for sustainable and low-cost wastewater treatment: Review article, *Land*, 11 (9), 1388.

[17] Gibson, T.F., Watanabe, W.O., Losordo, T.M., Whitehead, R.F., and Carroll, P.M., 2020, Evaluation of chemical polymers as coagulation aids to remove suspended solids from marine finfish recirculating aquaculture system discharge using a geotextile bag, *Aquacult. Eng.*, 90, 102065.

[18] Nhut, H.T., Hung, N.T.Q., Lap, B.Q., Han, L.T.N., Tri, T.Q., Bang, N.H.K., Hiep, N.T., and Ky, N.M., 2021, Use of *Moringa oleifera* seeds powder as bio-coagulants for the surface water treatment, *Int. J. Environ. Sci. Technol.*, 18 (8), 2173–2180.

[19] Karnena, M.K., Konni, M., and Dwarapureddi, B.K., 2022, Blend of natural coagulants as a sustainable solution for challenges of pollution from aquaculture wastewater, *Appl. Water Sci.*, 12 (3), 47.

[20] Aziz Zaid, A.Q., and Ghazali, S., 2019, Preliminary investigation of water treatment using *Moringa oleifera* seeds powder as natural coagulant: A case study of Belat River, Malaysia, *Int. J. Eng. Sci.*, 8 (2), 79–85.

[21] Bhuptawat, H., Folkard, G.K., and Chaudhari, S., 2007, Innovative physico-chemical treatment of wastewater incorporating *Moringa oleifera* seed coagulant, *J. Hazard. Mater.*, 142 (1-2), 477–482.

[22] Mohamad, N.A., Hamzah, S., Che Harun, M.H., Ali, A., Rasit, N., Awang, M., Wan Abd. Rahman, W.R., Azmi, A.A.A.R., Abu Habib, A.A., Amri Zahid, M.S., Fahmi Mustofa, A.A., Latfi, S.A., Mohd Aripin, S., and Saad, R., 2021, Integration of copperas and calcium hydroxide as a chemical coagulant and coagulant aid for efficient treatment of palm oil mill effluent, *Chemosphere*, 281, 130873.

[23] Hossain, M.S., Omar, F., Asis, A.J., Bachmann, R.T.,

Islam Sarker, M.Z., and Ab Kadir, M.O., 2019, Effective treatment of palm oil mill effluent using $FeSO_4 \cdot 7H_2O$ waste from titanium oxide industry: Coagulation adsorption isotherm and kinetics studies, *J. Cleaner Prod.*, 219, 86–98.

[24] Lin, J., Zhan, Y., and Zhu, Z., 2011, Adsorption characteristics of copper(II) ions from aqueous solution onto humic acid-immobilized surfactant-modified zeolite, *Colloids Surf., A*, 384 (1-3), 9–16.

[25] Hussain, S., van Leeuwen, J., Chow, C., Beecham, S., Kamruzzaman, M., Wang, D., Drikas, M., and Aryal, R., 2013, Removal of organic contaminants from river and reservoir waters by three different aluminum-based metal salts: Coagulation adsorption and kinetics studies, *Chem. Eng. J.*, 225, 394–405.

[26] Gök, Ö., Özcan, A., Erdem, B., and Özcan, A.S., 2008, Prediction of the kinetics, equilibrium and thermodynamic parameters of adsorption of copper (II) ions onto 8-hydroxy quinoline immobilized bentonite, *Colloids Surf., A*, 317 (1-3), 174–185.

[27] Zhu, G., Zheng, H., Zhang, Z., Tshukudu, T., Zhang, P., and Xiang, X., 2011, Characterization and coagulation–flocculation behavior of polymeric aluminum ferric sulfate (PAFS), *Chem. Eng. J.*, 178, 50–59.

[28] Lee, C.S., Robinson, J., and Chong, M.F., 2014, A review on application of flocculants in wastewater treatment, *Process Saf. Environ. Prot.*, 92 (6), 489–508.

[29] Ostolska, I., and Wiśniewska, M., 2014, Application of the zeta potential measurements to explanation of colloidal $Cr_2O_3$ stability mechanism in the presence of the ionic polyamino acids, *Colloid. Polym. Sci.*, 292 (10), 2453–2464.

[30] Wang, L.L., Wang, L.F., Ren, X.M., Ye, X.D., Li, W.W., Yuan, S.J., Sun, M., Sheng, G.P., Yu, H.Q., and Wang, X.K., 2012, pH Dependence of structure and surface properties of microbial EPS, *Environ. Sci. Technol.*, 46 (2), 737–744.

[31] Mohamad, N.A., Hamzah, S., Che Harun, M.H., Ali, A., Rasit, N., Awang, M., Wan Abdul Rahman, W.R., Abd. Rahman Azmi, A.A., Zahid, M.S.A., Mustofa, A.A.F., Ahmed Latfi, S., Mohd Aripin, S., and Saad, R., 2021, Copperas as iron-based coagulant for water and wastewater treatment: A review, *Biointerface Res. Appl. Chem.*, 12 (3), 4155–4176.

[32] Ngteni, R., Hossain, M.S., Ab Kadir, M.O., Asis, A.J., and Tajudin, Z., 2020, Kinetics and isotherm modeling for the treatment of rubber processing effluent using iron(II) sulphate waste as a coagulant, *Water*, 12 (6), 1747.