



Research Article

Synergistic integration of *Moringa oleifera* and coconut husk for novel wastewater adsorption applications

Dayang Norafizan Awang Chee^{1*}, Claudeareena Garding Malien¹, Nur Afiqah Kamaludin¹, Muhammad Shamil Soffian¹, Faezrul Zackry Abdul Halim¹, and Mohamed Afizal Mohamed Amin²

¹Faculty of Resource Science and Technology, Universiti Malaysia Sarawak, 94300, Kota Samarahan, Sarawak, Malaysia

²Department of Chemical Engineering and Energy Sustainability, Faculty of Engineering, Universiti Malaysia Sarawak, 94300 Kota Samarahan, Sarawak, Malaysia

*Corresponding author: dnorafizan@unimas.my

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Abstract

Moringa oleifera has gained significant attention from researchers due to its efficacy as a natural adsorbent for water treatment, but it alone is unable to remove turbidity, total dissolved solids (TDS), total suspended solids (TSS), and electrical conductivity (EC) effectively from wastewater. This requires chemical alterations using concentrated acid and alum, which is undesirable; thus, this study reported the use of combined *M.oleifera* with coconut husk (CH) as a coagulant for water treatment. The combination of 0.06 g of *M.oleifera* and 0.02 g of CH to form MOCH 3:1, identified as the best optimum composition, was evaluated based on turbidity removal at various pH levels (2, 4, 6, 8, and 10) and temperature (25°C, 40 °C, 60°C, 80°C and 100°C), TDS, TSS, and EC removal from wastewater. The surface properties of these materials were examined through SEM-EDX, FTIR, BET-N₂ adsorption, and zeta potential. The MOCH 3:1 had a synergistic impact, resulting in the highest removal efficiency of over 90% for turbidity, TDS, TSS, and EC from synthetic wastewater at pH 6 and 25°C. This showed that MOCH 3:1 removes wastewater pollutants better than individual coagulants.

Keywords: *Moringa oleifera*, coconut husk, impurities, wastewater, treatment

Introduction

Water is an integral part of life and is pivotal in sustaining human civilisation and the ecosystem. However, the quality of water is increasingly threatened by pollution, rapid population growth, and climate change, which frequently disrupt the availability of clean water [1]. Discharges of untreated wastewater introduce heavy metals, excess nutrients, and suspended solids that cause eutrophication, biodiversity loss, and severe health risks [2]. These challenges place substantial stress on freshwater resources, underscoring the urgent need for efficient and sustainable water treatment methods. Therefore, several water treatment techniques have been employed to remove contaminants from the wastewater, such as physical treatment, chemical treatment, and biological treatment [3]. The application of aluminium sulphate as a primary chemical coagulant in the coagulation-flocculation

process is frequently implemented in water treatment plants to remediate wastewater [4]. This method is unfavourable because the byproduct formed, which is trihalomethanes (THMs), is linked with carcinogenic properties and toxic to both human and aquatic organisms [5]. Bio-based adsorbents present an environmentally sustainable alternative to conventional chemical coagulants due to their biodegradability and natural efficiency [6]. Functional groups such as hydroxyl, carboxyl, and carbonyl facilitate adsorption and charge neutralization, improving contaminant removal from wastewater. Natural adsorbents like *M. oleifera* have gained the highest popularity due to their biodegradable, non-toxic, and locally available properties, reducing environmental impact and operational costs [7].

Even though current studies have focused heavily on the use of *M. oleifera* as a water purification agent,

prior research has indicated that individual *M. oleifera* necessitated certain adjustments to enhance its efficacy in eliminating pollutants from wastewater due to its low porosity [8–10]. Previous studies indicated that specific modifications were necessary for individual *M. oleifera* to effectively eliminate pollutants from wastewater due to its low porosity [11–15]. For instance, the utilisation of hydrochloric acid, sulphuric acid, sodium hydroxide, and potassium hydroxide is used to enhance the removal of low turbidity by using *M. oleifera* but it contravenes the principles of green chemistry [16–19]. Multiple studies have integrated *M. oleifera* seed with aluminium sulphate, and some have modified the seed extract using concentrated sulphuric acid; however, these approaches elevate the acidity of wastewater samples post-treatment, necessitating further measures to achieve neutral pH [20–24]. Plus, the dosage of coagulant must be ascertained to prevent elevated turbidity. Numerous studies advocate for higher dosages to address high turbidity and enhance adsorption, but the organic nature of *M.oleifera* may also promote bacterial growth by serving as a nutrient source [25].

Although numerous studies have linked *M.oleifera* with alum and treated it with strong acids, as mentioned previously, none have integrated coconut husk (CH) to enhance wastewater pollutant removal. CH, composed of cellulose, lignin and tannins, provides high adsorption capacity and acts as a complementary material when integrated with *M.oleifera* [26]. Moreover, CH is a superior wastewater adsorbent, surpassing sugarcane bagasse, palm kernel shell, bamboo shell, and rice husk due to its porous structure, high absorptivity, and strong structural stability [27, 28].

Nasaruddin & Mohamed [23] reported that the greatest removal of total dissolved solids (TDS), total suspended solids (TSS), and electrical conductivity (EC) can be obtained by using CH as it can provide additional sites for the suspended solids. Thus, the synergistic effects from both *M.oleifera* and CH could help to trap more impurities and accelerate the reaction due to their high surface area and more functional groups on the surface of the adsorbent.

Therefore, this study presents a novel approach to increase the removal efficiency of turbidity, total dissolved solids (TDS), total suspended solids (TSS) and electrical conductivity (EC) removal from wastewater by combining *M.oleifera* and CH biosorbents with different dosages while avoiding overdosing. Additionally, zeta potential and surface characteristics of the biosorbents have been examined, emphasising previously uninvestigated dimensions.

Materials and methods

Sample collection and materials

The research was conducted at the Analytical Chemistry Lab, Faculty of Resource Science and Technology (FRST), University Malaysia Sarawak (UNIMAS), using synthetic wastewater prepared in the lab to evaluate the coagulation-flocculation performance of the coagulant, where all analyses were conducted. The solution's pH was determined and regularly adjusted using a pH meter (Model HI9024, HANNA, USA) and pH 4, 7, and 10 standard buffer solutions until it reached a stable value [8]. In addition, the turbidity of the solution was calibrated using 0.5, 10, and 20 NTU standards. The solution was measured using a turbidity meter (Model 0839, Cole-Parmer, USA). The measurement range of this turbidity meter for high turbidity before treatment ranged from 50 NTU to 600 NTU at 660 nm of wavelength, while after treatment using biocoagulants for low turbidity, it ranged from 0 NTU to 20 NTU at 860 nm of wavelength [29]. The initial turbidity of the raw water was assessed, and the average value obtained from these measurements would serve as the starting point. The *M. oleifera* and CH were commercially procured from Babymommom.com.my, a supplier company based in Kuala Lumpur, Malaysia.

Preparation of individual coagulants

A 300-mesh sieve (50 μm) ASTM was used to sieve the seed powder [30, 31]. The preparation of coagulants was conducted following the method proposed by Mohammed et al. [32]. A 0.02 g of seed powder was mixed with 100 mL of 1M sodium chloride (NaCl) salt solution (NaCl, $\geq 99.5\%$ purity, Uni-Chem) to extract the active component from the seed. The solution was stirred for 15 minutes using a magnetic stirrer (Remi, 1 MLH India) at 500 rpm under room conditions. The coagulant salt suspension was filtered using filter paper (Whatman 1). The solution must be freshly prepared to avoid coagulant activity reduction due to microbial decomposition.

Next, the CH was prepared using a method suggested by Sujiono et al. [33]. The sample underwent grinding and sieving until it reached a particle size of 200 mesh. After that, the mixture was heated to 45 °C for 3 hours. Subsequently, the sample was washed with deionized water until the pH level of the solution reached between 6 and 7. The sample was then dried in an oven at 110 °C for 12 hours. A sodium hydroxide (NaOH) activator (NaOH, $\geq 99\%$, Bendosen) was used to activate the sample chemically by a ratio of 1:2 (1 g of CH activated carbon: 2 g of NaOH), which was dissolved with 10 ml of aqueous solution. The sample was dried in an oven to obtain a powder form, and it was then combined with the optimum coagulant

Table 1. Combination of different MOCH ratios used for the coagulation-flocculation process

MOCH ratio	<i>M. oleifera</i> (g)	CH (g)
MOCH 1:3	0.02	0.06
MOCH 3:1	0.06	0.02
MOCH 1:4	0.02	0.08
MOCH 4:1	0.08	0.02
MOCH 4:5	0.08	0.10

Note: MOCH = *M.oleifera*: coconut husk

Table 2. Initial characteristics of synthetic wastewater before the adsorption process

Water sample	Kaolinite Dosage (g)	Turbidity (NTU)	TDS (mg/L)	TSS (mg/L)	EC (µs/cm)
Synthetic turbid wastewater	30	300	2170	8758.5	2440

dosage of *M.oleifera* to treat turbidity, TDS, TSS, and EC. The EC was quantified and recorded in microsiemens per centimetre (µs/cm) using a conductivity meter, while the pH of the solution was determined using a pH meter (EUTECH instruments). The instruments must be calibrated using a standard buffer solution before taking the initial measurement to obtain accurate results.

Preparation of combined coagulants

The three optimal dosages of the individual coagulants, which were determined from prior experiments, were selected and combined through a blending method in a variety of ratios to create distinct *M.oleifera* and coconut husk (MOCH) combinations. The MOCH was prepared by mixing *M.oleifera* and coconut husk in different ratios. For example, MOCH 1:3 (*M. oleifera* : CH = 1:3; 0.02 g : 0.06 g). Other ratios, including MOCH 3:1 (0.06 g : 0.02 g), MOCH 1:4 (0.02 g : 0.08 g), MOCH 4:1 (0.08 g : 0.02 g), and MOCH 4:5 (0.08 g : 0.10 g), were also tested to evaluate turbidity removal performance. The sample designations for these dosages are shown in **Table 1**.

Preparation of synthetic wastewater

Before coagulation-flocculation tests, synthetic wastewater samples were generated by dispersing a pre-prepared suspension of kaolinite particles to regulate turbidity in a specific volume of tap water. The kaolinite particles were chosen based on their surface structure properties, such as hydrophilicity and non-swelling features, which help them disperse quickly in water-based systems. The following method for preparing synthetic turbid wastewater, which mimics the industrial effluent, was conducted and adjusted based on a study by Vigneshwaran et al. [34]. The kaolin powder was dried in a hot-air oven at 105 °C. After that, it was removed from the oven and maintained at room temperature. The original kaolinite suspension was created by dispersing 0.5 g

of kaolinite particles in 1 litre of distilled water to represent 50 NTU. The suspensions were vigorously agitated for about one hour using rapid mechanical stirring at 120 rpm to ensure uniform distribution of the kaolinite clay particles. The mixture was allowed to sit undisturbed for 24 hours to ensure complete hydration of the kaolinite clay. The dispersion stated previously was used to maintain the turbidity of synthetic wastewater samples at a certain level. The turbidity of synthetic wastewater for 100 NTU, 150 NTU, 200 NTU, 250 NTU, 300 NTU, 350 NTU, 400 NTU, 450 NTU, 500 NTU, 550 NTU, and 600 NTU was adjusted according to different kaolinite ranges from 10-60 g. The filtrate produced was utilized as a stock solution in the coagulation experiment. Then, the pH of the kaolin suspension was measured and adjusted using 0.1 M sodium hydroxide (NaOH, ≥99%, Bendosen) and 0.1 M hydrochloric acid (HCl, ≥99%, Sigma-Aldrich) to study the effect of pH on turbidity levels. The turbidity of synthetic wastewater was measured before and after treatment with the coagulants. The initial and final turbidity of the synthetic wastewater samples were assessed before and after treatment, respectively. Table 2 conveys initial turbidity (300 NTU), TDS (2170 mg/L), TSS (8758.5 mg/L), and conductivity (2440 µS/cm) at pH 6 as a baseline for comparison with the final assessment. The difference between the original and final turbidity measurements was then used to calculate turbidity removal efficiency.

Turbidity measurement

A turbidity meter is frequently employed to evaluate the effectiveness of the treatment process by measuring the concentration of suspended solids in water, which is expressed in nephelometric turbidity units (NTU). The turbidity measurement was performed in adherence to the methodology outlined by Cobos et al. [35]. The turbidity meter (Thermo Scientific™ Orion™ AQUAfast AQ3010) was first

calibrated using standard calibration solutions of water before proceeding with the actual measurement. The surface-treated wastewater sample was collected using a dropper after the coagulation-flocculation process. The sample was then transferred to a clean vial until it reached the designated fill line. The vial was pre-cleaned with distilled water and then thoroughly wiped with a lint-free cloth to remove any residues. The vial was positioned in the sample holder of the turbidity meter after the sample was prepared to ensure that it was in correct alignment with the instrument's markings. The vial was subsequently covered to prevent interference from ambient light. Repeated average readings were recorded until a consistent turbidity value was achieved for every 10 minutes. The vial was thoroughly dried and cleansed with distilled water after each measurement to ensure the accuracy of subsequent measurements.

The turbidity removal percentage was then calculated using the Eq. (1).

Turbidity removal (%)

$$= \frac{\text{Turbidity initial} - \text{Turbidity final}}{\text{Turbidity initial}} \times 100\% \quad (1)$$

Surface morphology and chemical characterization method

The scanning electron microscope (SEM) (JEOL JSM-6390LV/LGS) analysis was performed on *M.oleifera*, CH and MOCH to examine their surface morphology and porosity before and after the coagulation-flocculation process. The samples were inserted into the SEM holder before the SEM was turned on. The samples were subsequently placed in a high-pressure chamber with a short working distance, and the electron optical column was differentially pumped to maintain an appropriate vacuum at the electron gun. After that, imaging was obtained.

The following method was conducted based on previous research by Okoya et al. [36]. The Fourier Transform Infrared Spectroscopy-Potassium Bromide (FTIR-KBr) (Thermo Scientific Nicolet iS10) technique was used to analyze the materials. The IR-grade KBr was dried first at 110°C by using a drying oven to avoid moisture interference since the powder was hygroscopic. After that, the sample was mixed with KBr using a mortar to ensure homogeneous distribution. The mixture was then meticulously transferred and formed into a transparent and thin pellet using a hydraulic pressure. Next, the pellet was positioned on a sample holder attached to a recording device and analyzed. The FTIR characterization of *M.oleifera*, CH, and MOCH was performed using

transmittance spectrum data and recorded in the 500-4000 cm⁻¹.

Surface area and pore analyzer method

The adsorbents were analyzed using a surface area and pore size analyzer (Thermo Scientific), followed by the Tokay & Akpinar [37] method. The samples underwent degassing for 3 hours in a vacuum at 105 °C to remove any moisture or adsorbed contaminants on the surface. The pore structures of the sample were analyzed by subjecting it to nitrogen adsorption at a temperature of 77 K after degassing it at a temperature of 105 °C. The surface parameters were measured, such as the BET-specific surface area, micropore volume, and pore size distributions. The BET surface area (S_{BET}) was determined by following Eq. (2).

$$\frac{P}{n^a(P_0 - P)} = \frac{1}{n_m^a} + \left(\frac{C-1}{n_m^a C}\right) \frac{P}{P_0} \quad (2)$$

The variable " n^a " represents the quantity of gas adsorbed at the relative pressure $\frac{P}{P_0}$. The " n_m^a " refers to the monolayer capacity, and " C " is the dimensionless BET constant, linked to the enthalpy of adsorption in the initial adsorbed layer. The macroporous surface area (S_{macro}) and exterior surface area (S_{ext}), along with the micropore volume (V_{mi}), were assessed using the t-plot method. The Barret-Joyner-Halenda (BJH) method was employed to ascertain the particle size distribution (PSD) and the volume of mesopores (V_{me}). The average pore diameter, D_p , was determined using the following Eq. (3).

$$D_p = \frac{4V_T}{S} \quad (3)$$

V_T represents the total quantity of all pores, while S refers to the BET surface area.

Zeta potential analysis

Zeta potential measurements were conducted using a Laser Particle Size Analyser (LPSA-MASTERSIZE 3000) equipped with a zeta potential module. The instrument applies electrophoretic light scattering (ELS), where an electric field is used to induce particle movement, and the mobility of the charged particles is determined from the frequency shift of the scattered laser light, which is then converted into zeta potential values. The following method followed a study by Bezerra et al. [38]. All samples were conducted by analysing 0.05 g of the substance dissolved in 50 ml of an aqueous solution. The pH values of the sample in each flask were modified using a 0.1 M HCl or 0.1 M NaOH solution until the required pH was achieved. A graph displayed the zeta potential values measured in millivolts (mV) with the pH levels and the resulting

curves were used to evaluate the stability and surface charge behaviour of the coagulants.

Coagulation-flocculation process

Each beaker was filled with 200 mL of synthetic wastewater, and the range of turbidity had to be the same before placing the coagulant dose into the respective beakers filled with synthetic wastewater. The synthetic wastewater was treated using different coagulant dosages at 0.02 g to 0.10 g in each beaker. The solution was stirred up at 100 rpm for 1 minute, followed by flocculation at 40 rpm for 10 minutes [39]. After that, the treated solution's turbidity, TDS, TSS and EC were measured using respective instruments. The characteristics of synthetic wastewater samples before and after treatment using *M.oleifera* and CH were recorded. The optimum coagulant dosage from *M.oleifera* and CH was selected based on their best performance in treating the water sample. Then, the efficiency of individual coagulants was evaluated further by comparing it using different combined MOCH ratios.

Total dissolved solids (TDS), total suspended solids (TSS) and electrical conductivity (EC) measurement

Total dissolved solids (TDS) is a measure of the dissolved inorganic and organic substances in milligrams per litre (mg/L) of water that includes calcium, magnesium, bicarbonates, sulphates, which are usually found in natural sources, sewage, industrial wastewater, and chemicals utilised in water treatment [40]. This water quality parameter is usually used to ensure the water's purity and the effectiveness of water treatment systems. The impact of TDS on drinking water is also a crucial factor to consider. Meanwhile, total suspended solids (TSS) are the concentration of suspended particles that dissolve in water samples, which are usually quantified in milligrams per liter (mg/L) [41]. This water quality parameter is used to evaluate the efficacy of wastewater treatment systems because high TSS levels in water samples are primarily associated with the possibility of microbial contamination that can cause challenges in the water purification process, particularly in flocculation and filtration [42]. The effects of TDS and TSS were often studied both before and after the treatment processes.

Total dissolved solids and electrical conductivity analysis

A multifunction PCD 650 waterproof portable meter was used to measure both electrical conductivity (EC) and total dissolved solids (TDS) [30]. The instrument was calibrated using standard solutions before measurement to ensure accuracy. EC values were recorded in $\mu\text{S}/\text{cm}$, while TDS values were expressed directly in mg/L. Before each measurement, the probe

was thoroughly cleaned and carefully immersed in the synthetic wastewater samples. All measurements were conducted at room temperature.

Total suspended solids analysis

The total suspended solids (TSS) measurement was according to the standard method as described in the American Public Health Association (APHA) [43]. The Consult APHA Method 2540 D was used to determine the TSS levels by the use of filtration and gravimetric techniques. The filter paper (Whatman No.1) was weighed using a weighing beam balance (A&D Weighing GX-224A, Japan). The 200 ml water sample underwent filtration using the Whatman No.1 filter paper. Afterward, the filter paper was dried in an oven at 110 °C for 1 hour and its weight was measured again. The TSS concentration in the water sample was expressed in milligrams per litre (mg/L). It was determined by calculating the difference in filter weights before and after treating the synthetic wastewater, as shown Eq. (4).

Total suspended solids,

$$\text{TSS (mg/L)} = \frac{W_F - W_I \times 1000}{\text{Volume of water}} \quad (4)$$

W_F = Final weight (g), W_I = Initial weight (g)

Results and Discussion

Characterization of the biosorbents

The characteristics and properties of biosorbents are essential to analysing their efficacy as adsorbents and enhancing their applications in treating wastewater. Multiple techniques, such as Fourier Transform Infrared Spectroscopy (FTIR), Scanning Electron Microscopy (SEM), Brunauer-Emmett-Teller (BET), and zeta potential measurements, are employed in this work to provide a thorough examination on the biosorbents such as *M. oleifera*, coconut husk (CH), and a combination of *M. oleifera* and coconut husk (MOCH). Each method offers different perspectives on the functional groups, surface area, and surface charge of the biosorbents, which are covered in more detail in the following sections.

FTIR analysis

The *M. oleifera* and CH functional groups were studied using the FTIR characterisation technique. The *M. oleifera*, CH, and MOCH 3:1 revealed the functional group present before and after the coagulation-flocculation process of removing turbidity wastewater. The FTIR analysis demonstrated notable disparities in the functional groups identified within all coagulants after the coagulation-flocculation process. The adsorption process occurred, as evidenced by the discernible shift to the left

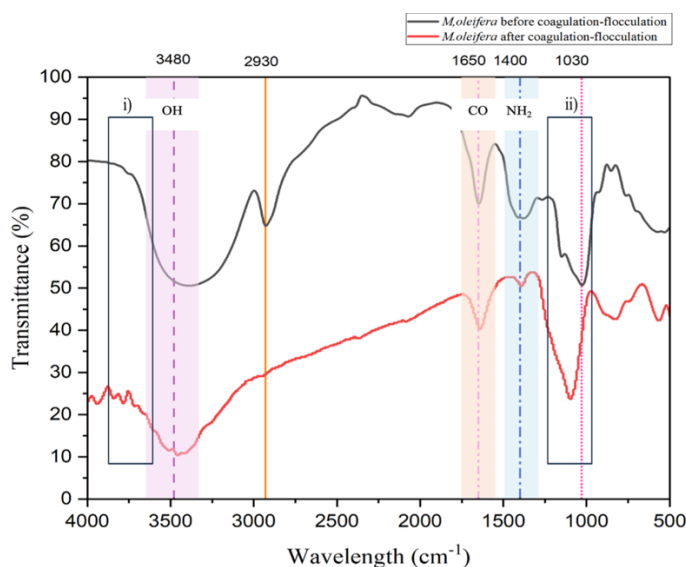


Figure 1 FTIR result of treating synthetic wastewater using *M.oleifera*

following the coagulation process, with a new peak observed between 4000 cm^{-1} and 3500 cm^{-1} as shown by label (i) in all graphs, which confirms the presence of kaolin particles on the surface of coagulants due to the interaction of kaolin with positively charged alumina faces, Al-OH functional groups, interacting with the negative charge of coagulants [44, 45].

The FTIR study of *M.oleifera* before coagulation, as depicted in **Figure 1**, revealed that these functional groups, including the -COO, -OH, and -NH₂ play an essential role in increasing flocculation through neutralisation and bridging mechanisms [46]. The wide band at 3480 cm^{-1} corresponds to the hydroxyl group of *M.oleifera* from protein and there may be a contribution in this region about the N-H stretch of amides [47]. Following the coagulation-flocculation process, the band narrows due to hydrogen bond formation with Al-OH from kaolin [48]. The peak at 2930 cm^{-1} signifies the presence of aliphatic hydrocarbons from the polysaccharides of *M.oleifera*, however, following the coagulation-flocculation process, the peak disappears due to interactions with the hydroxyl groups of kaolin via hydrogen bond, which indicates the successful interaction between *M.oleifera* and the colloidal particle occurred [49]. The peak at 1650 cm^{-1} indicates C=O vibration in the amide, which can form a covalent bond with Al³⁺, acting as a Lewis acid that accepts electrons from the C=O [50]. Additionally, the peak at 1400 cm^{-1} , corresponding to primary amine (NH₂), is capable of forming hydrogen bonds with Al-OH, resulting in a reduction of peak intensity following the coagulation-flocculation process. The peak at 1030 cm^{-1} corresponds to the C-O bond from the pyranose ring in the polysaccharide of *M.oleifera*, which can

interact with the Si-OH group on the kaolin surface via hydrogen bonding, resulting in an increase in shift peak intensity following turbidity removal [51]. Subsequently, two peaks were detected between 500 cm^{-1} and 1000 cm^{-1} after the coagulation-flocculation process, indicating there is interaction between the hydroxyl group of the aromatic lignin from *M.oleifera* and the hydroxyl groups of kaolin, specifically Al-OH and Si-OH, via hydrogen bonding [52].

Other than that, Figure 2 illustrates the FTIR analysis pertaining to the coagulation-flocculation activity of CH. The main characteristic of cellulose, hemicelluloses, and lignin is the presence of functional groups such as C=O, C-H, and OH, which collectively form the primary properties of CH [53]. The absorption band at 3460 cm^{-1} corresponds to the hydroxyl group, OH, from the polysaccharide component of C-H, which can engage in hydrogen bonding with the OH group from kaolin, as the oxygen in Al-OH from kaolin can take hydrogen from the OH group in CH [26]. The minor peak at 2360 cm^{-1} indicates the presence of C≡C in lignocellulosic material, as it exhibits minimal absorption of infrared light but the peak intensity increases due to the generated dipole-dipole interaction between C≡C and Al-OH from kaolin after the coagulation-flocculation process [54]. Additionally, the C=O bond from hemicellulose was detected at 1650 cm^{-1} interacting with Al-OH, hence diminishing the electron density and weakening the dipole moment in the carbonyl group, which results in a reduction of peak intensity [55]. The C-H stretching of the alkyl group in hemicellulose was detected around 1400 cm^{-1} and this alkyl group can form an induced dipole when interacting with Al-OH from kaolin via Van der Waals

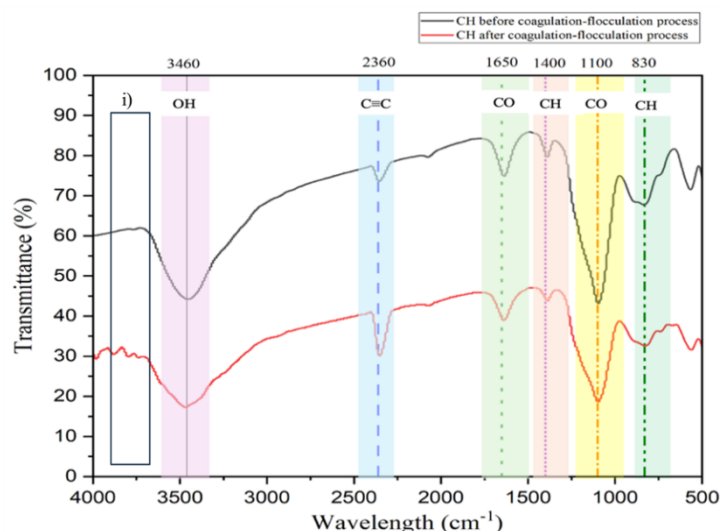


Figure 2 FTIR result of treating synthetic wastewater using CH

forces [56]. The Si-O group in CH exhibited asymmetric bending at 1100 cm^{-1} , which can interact with Al-OH, resulting in hydrogen bonding [57]. The peaks before coagulation-flocculation process at 830 cm^{-1} and 550 cm^{-1} correspond to out-of-plane bending vibrations of C-H in the aromatic rings of lignin in CH [58]. After the turbidity removal process, the decrease in peak intensity of C-H vibration from CH was observed upon interaction with kaolin due to hydrogen bonding [59].

The resultant composite is a combined MOCH 3:1 as depicted in **Figure 3**. The peak at 3400 cm^{-1} due to the presence of hydroxyl group present in the carbohydrates, protein, and lignin from *M. oleifera* and CH. The interaction of kaolin, which may form hydrogen bonds with hydroxyl groups from both *M. oleifera* and CH, causes the peak intensity to increase and the hydroxyl group to broaden after the coagulation-flocculation process [60]. A small peak at 2930 cm^{-1} arises from the combination of *M. oleifera* and CH, attributed to the C-H stretching vibration in polysaccharides of *M. oleifera* that interacts with Si-O-Si from CH, a non-polar siloxane group, via hydrophobic contact but the peak disappears following the interaction with kaolin, indicating that adsorption has occurred. Additionally, a shift peak occurs at 1645 cm^{-1} at the combination of *M. oleifera* and CH, attributed to the presence of C=O from both coagulants, which interact through dipole-dipole interactions, hence reducing the intensity peak relative to each coagulant individually, as previously mentioned [61].

After adsorption, the peak intensity increases due to the rearrangement of molecules, which results in the C=O group becoming more exposed for interaction with kaolin [62]. Furthermore, the peak at 1100 cm^{-1} is attributed to the interaction between the C-O bond of the pyranose ring in *M. oleifera* and the oxygen group of the carbonyl group in CH through dipole-dipole interaction, which diminishes the dipole moment of the C-O bond in the pyranose, resulting in a reduced peak compared to the individual coagulant [63]. The increase in peak intensity following the interaction of kaolin with MOCH 3:1 may be ascribed to the more structured dipole-dipole interactions amongst coagulants facilitated by kaolin.

Other than that, the two peaks found at 600 cm^{-1} and 843 cm^{-1} indicate the interaction between the C-O bond of the pyranose ring in *M. oleifera* and the Si-O bond in CH, facilitated by dipole-dipole interactions, where the reduced peak intensity suggests physical adsorption of C-O groups onto the silica surface of CH [64]. The interaction between kaolin and MOCH 3:1 becomes more ordered after the adsorption, which amplifies the peak because of the synergistic effect from the dipole-dipole interaction of both coagulants. Therefore, the hydroxyl, carbonyl, and amine groups present in the coagulants are responsible for its ability to coagulate and purify water via the adsorption process, confirming their role as excellent contaminants absorbents [65]. These findings proved the potential for an interaction between the coagulants to enhance contaminants removal capability through the synergistic effect of their functional groups.

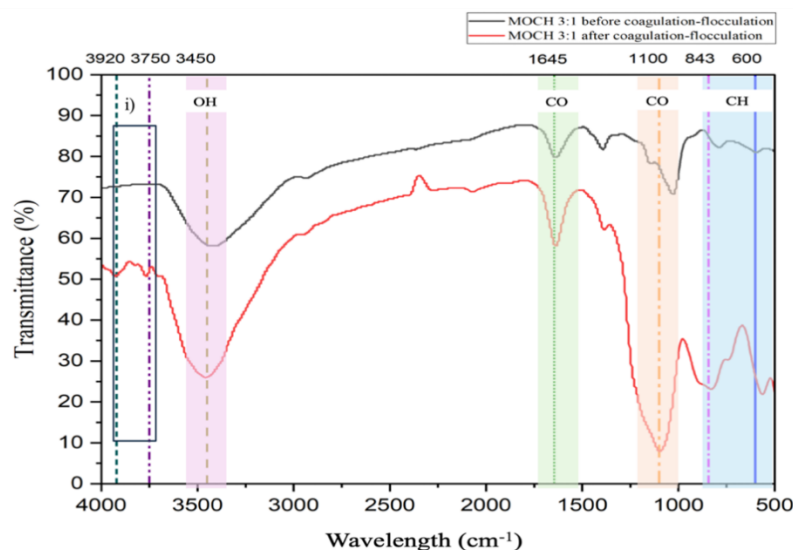


Figure 3. FTIR result of treating the synthetic wastewater using MOCH 3:1

Scanning electron microscopy analysis

Morphological analysis is a vital tool for analysing the surface area and formation of pores on the surface of *M.oleifera* and CH, indicating the activation process's efficacy. The surface morphology of *M. oleifera* and CH was analyzed using SEM images. The *M. oleifera* exhibits a heterogeneous, coarse surface with blocks of various irregular forms and pores of varying diameters before the coagulation process took place, as depicted in **Figure 4**. The results of this study are concurrent with the conclusions made by Nonfodji et al. [60] and Alghamdi et al. [66]. Numerous studies also stated that *M. oleifera* has irregularly formed blocks, many pores of varying sizes, and appears villous, which also plays a vital role in the adsorption process [67–69]. The closed pores in *M. oleifera*'s structure were due to a significant amount of cellulose and lignin in its structure [70]. Nevertheless, other studies have shown contradictory findings in which the natural *M. oleifera* has a smooth surface with a high number of pores [49, 53].

Apart from that, the SEM results before treatment revealed that CH has a rough surface and more pores than *M.oleifera*, which can aid in removing contaminants from the wastewater [71]. Previous studies found that CH is more effective than other plant biomass in water purification due to its dense surfaces with heterogeneous pores, uneven, rough, and irregular shapes [63, 72, 73, 74]. Meanwhile, the SEM result indicated that there is an alteration in physical structure when combining the *M. oleifera* and CH. The MOCH 3:1 exhibits a composite of a coarse texture and villous properties, accompanied by larger pores resulting from the amalgamation of *M. oleifera* and CH. The diverse surface composition of combined MOCH 3:1 offers numerous locations for interparticle

connection, which improves the process of clumping together and aids in the settling of contaminants.

The adsorption of kaolin particles from synthetic wastewater by biosorbents led to significant structural changes in the biosorbents following the coagulation-flocculation process, with a complete coating of kaolinite and a transformation of the surface pores into an irregular and densely packed arrangement, as shown in **Figure 4**. The observed outcomes might be attributed to the interaction of kaolinite and the biosorbent particles, aligning with findings from Vigneshwaran et al. [35]. The SEM results demonstrated that the surface texture for all adsorbents exhibited softer, irregular, and porous after adsorption process, indicating a significant specific surface area aiding in the turbidity removal by bridging coagulants and kaolin particles [73]. Therefore, the SEM surface morphology evaluation and FTIR analysis, as previously described, jointly offer valuable insights into the adsorption capabilities of the biosorbents and their potential applications in water treatment [19].

Surface area and pore analysis

The surface area of biosorbents was estimated using nitrogen adsorption-desorption isotherm measurement (Micromeritics TriStar II 3020). Nitrogen adsorption is a commonly used method to assess the porosity of adsorbents, providing valuable information about their pore structure. The samples were degassed for 3 hours at 150 °C before analysis to remove moisture and impurities without affecting the structure of the sample. **Figure 5** show the BET results for individual coagulants. The morphological features of *M.oleifera* exhibit the smallest pore volume and surface area, with 0.001313 cm³ g⁻¹ and

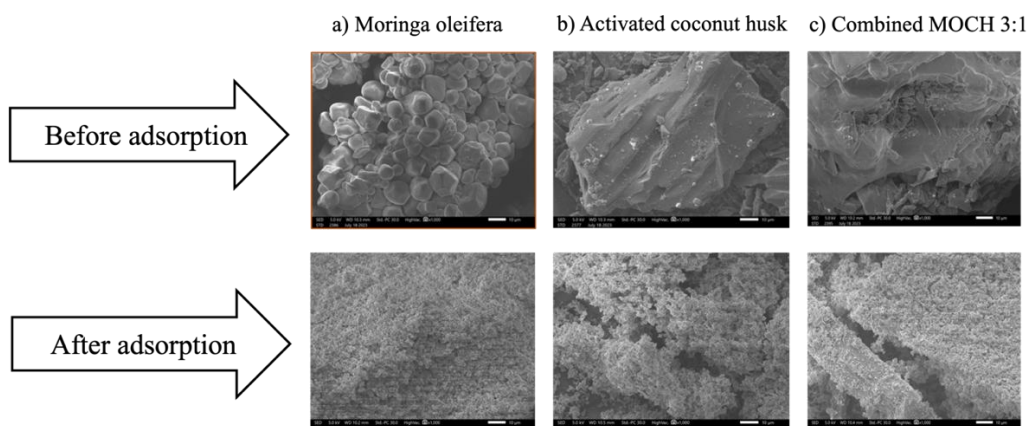


Figure 4. Surface morphology of respective coagulants at x1000 magnifications

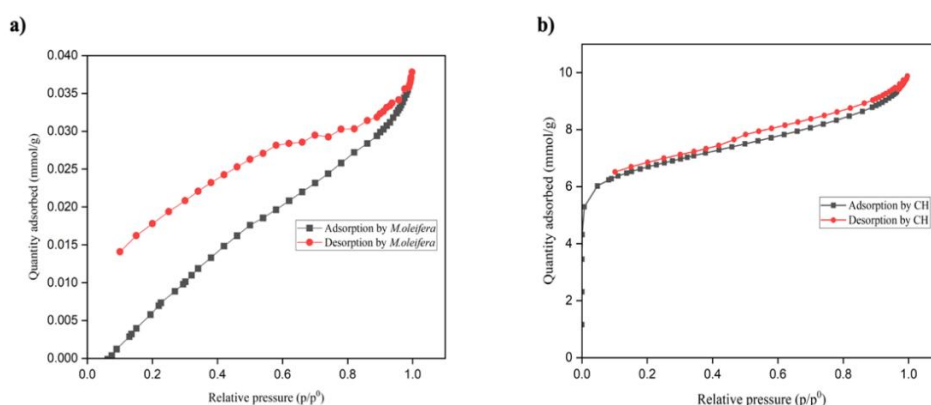


Figure 5. BET analysis of individual coagulants (a) *M.oleifera* (b) CH

0.1273 m² g⁻¹, respectively. The coconut husk (CH) exhibited the greatest pore volume and surface area due to its porous structure, at 0.343164 cm³ g⁻¹ and 552.1280 m² g⁻¹. **Figure 5(a)** demonstrates that the graph is a Type II isotherm, distinguished by a positive curvature of the isotherm. This behavior is generally linked to multilayer adsorption on the non-porous structure of *M.oleifera* and is also ascribed to its comparatively low surface area relative to other biosorbents [75]. This behavior suggests that the adsorbate and adsorbent have weak initial contacts, followed by improved adsorption as multilayer development takes place at greater adsorbate concentrations. Moreover, the graph displayed significant open-loop hysteresis, indicating that certain adsorbates remain firmly attached to the surface due to strong chemical bonding, even at low pressure [76]. Subsequently, a type IV isotherm is initially established in the monolayer at low pressure, followed by the formation of a multilayer, as illustrated in **Figure 5(b)**. This indicates that the contact between the adsorbate and adsorbent occurs via Van der Waals forces during initial adsorption, and

once the surface is saturated, new adsorbate will create a second layer on the initial monolayer [77]. This graph illustrates that the CH exhibits minimal hysteresis, indicating that this process adheres to physisorption, wherein the adsorbate molecules can be readily desorbed due to weak interactions. This suggests that CH possesses a mesoporous structure and implies that the adsorbent demonstrates significant recyclability, rendering it effective for pollutant removal from wastewater throughout numerous cycles.

Next, **Figure 6** illustrates that the MOCH 3:1 adheres to a Type IV isotherm with a moderate open loop hysteresis, indicating its mesoporous nature and a balance between physisorption and chemisorption. Consequently, the narrow necks of the mesopores in the MOCH 3:1 induce medium open loop hysteresis, hindering the escape of certain adsorbates and thereby obstructing the desorption process [78]. In the preliminary adsorption phase, the interaction between MOCH 3:1 and the adsorbate adheres to the Langmuir isotherm, characterized by monolayer adsorption on

the surface, succeeded by multilayer adsorption as pressure increases [79]. On the other hand, the MOCH 3:1 mixture exhibited greater pore volume and surface area compared to *M. oleifera*, with values of 0.010866 cm³ g⁻¹ and 17.1006 m² g⁻¹, respectively. This is because the *M. oleifera* occupied the accessible sites on the CH, resulting in a decreased pore volume and surface area compared to the CH. While MOCH 3:1 exhibited a smaller pore size compared to CH, the biosorbent's contact area is greater due to the presence of more functional groups on the surface that enhance the efficient removal of contaminants from the wastewater. This finding is in line with prior research that has also substantiated this conclusion [80, 81].

This discrepancy can be attributed to activated carbon CH being a porous carbonaceous material with a much larger surface area than *M. oleifera* [81]. Previous studies have shown that unprocessed *M. oleifera* have a specific surface area ranging from 1.2 to 1.52 m² g⁻¹ and a total pore volume ranging from 0.002 to 0.021 cm³ g⁻¹ [38, 70, 82]. In comparison to other biosorbents, such as sugarcane bagasse biomass and aquatic *S. auriculata* plants, *M.oleifera* has a higher total pore volume but a smaller specific surface area, according to the BET data [83]. Apart from that, the BET textural properties of MOCH 3:1 have a high specific surface area of 17.1006 m² g⁻¹, along with a total pore volume of 0.010866 cm³ g⁻¹. In addition, the adsorption rate of an adsorbent is directly influenced by the surface area and pore volume of the adsorbent, in which a more extensive surface area results in a higher rate of adsorption [50]. *M.oleifera* occupies a greater percentage of the available pores in CH, increasing the pore volume and surface area, which accounts for MOCH's increased surface area that allows more adsorption processes to take place than individual *M. oleifera*. Other researchers reported that *M. oleifera* which has been treated with highly concentrated nitric acid (HNO₃), showed that its specific surface area was 5.77 m² g⁻¹, and its total pore

volume was 0.0409 cm³ g⁻¹, which is higher than *M. oleifera* seed that had been exposed to H₂SO₄ with a specific surface area of 1.2 m² g⁻¹ and a total pore capacity of 0.002 cm³ g⁻¹ [70, 84]. These findings revealed that the surface of MOCH 3:1 exhibited an enhanced surface area and a greater number of pores than those documented in prior studies employing concentrated acid treatments, as previously mentioned.

Furthermore, the average pore size of *M. oleifera*, CH, and MOCH 3:1 were 412.555 Å, 24.861 Å, and 25.416 Å, respectively. As per IUPAC, pore sizes are categorized as micropores (less than 20 Å), mesopores (between 20 Å and 500 Å), and macropores (more than 500 Å) [12]. The *M. oleifera*, CH, and MOCH 3:1 are categorised as mesopore-sized, which makes them ideal adsorbents for particle absorption due to their high selectivity and adsorption capacity as mesopores facilitate enhanced accessibility, faster adsorption, and greater capacity for diverse larger contaminants in comparison to micropores [85]. Previous studies reported similar observations [86].

Zeta potential analysis

The zeta potential is correlated with pH variations to understand better the mechanism underlying the coagulation-flocculation capabilities of coagulants, as depicted in **Figure 7**. The zeta potential measurements, which quantify the surface charge, confirmed the existence of negatively charged ionic species on the surface displayed as a function of pH ranging from 2 to 12. The curve demonstrates that the surface of the biosorbents is primarily negatively charged at all pH levels. These numbers show an increase in the negative charges on the surface of the biosorbent towards more alkaline pHs. For instance, the range of measured zeta potential values from pH 2 to 12 by the *M.oleifera* is -3.29 to -18.5, CH is from -0.773 to -29.3, and for MOCH 3:1 is from -0.572 to -20.0.

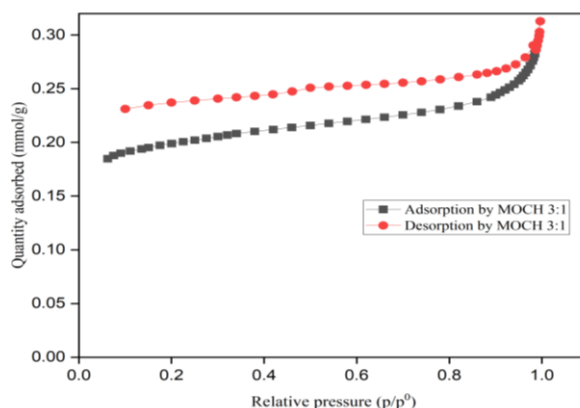


Figure 6. BET analysis of MOCH 3:1

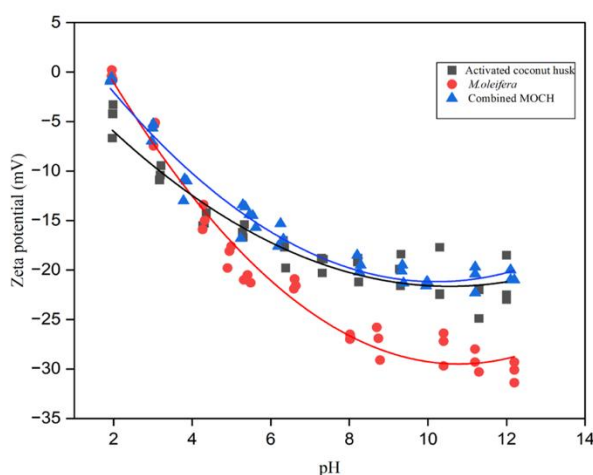


Figure 7. Zeta potential of respective coagulants

Since the kaolin suspension's surface properties are well understood and enable research on flocculation and sedimentation processes, it has been employed for decades as a representative of colloidal systems. The agglomeration of particles is facilitated by the interaction between the positive charges on the basal face and the negative charges on the margins of the heterogeneous kaolin particle surface [87]. Therefore, it may be concluded that the adsorption of turbidity is independent of the electrostatic attraction condition, as the particle has grown even more negatively charged [88]. The ideal coagulation for the biosorbents occurred at pH 2, when minimal repulsive forces were observed, suggesting that charge neutralization is preferred at this pH level. At pH 10, all biosorbents displayed significantly negative zeta potential values, indicating strong repulsion among particles. As a result, no aggregation occurred, and the particles remained uniformly distributed throughout the solution.

Researchers have defined the coagulation mechanism of the active component of *M. oleifera* as adsorption and neutralisation of charges, as well as interparticle bridging [89]. This statement is consistent with researchers attributing that the zeta potential remained negative when using *M. oleifera* [90]. Hence, the occurrence of coagulation through charge neutralisation was improbable [91]. The reduction in turbidity is because of the MOCH 3:1 that entangles suspended solids like kaolin particles in a net-like structure through a sweep coagulation mechanism [92, 93]. This is because the bivalent cations of the kaolin particles may link to the active component of MOCH 3:1, which has a negative charge, forming an insoluble substance. Therefore, the primary mechanism for coagulation is either adsorption or interparticle bridging of destabilised with both mechanisms potentially occurring concurrently.

Turbidity removal performance using individual coagulants

The mixture was stirred for 1 minute at a high speed of 100 rpm to ensure deep penetration of the coagulant protein into the synthetic wastewater, which triggers the creation of small flocs. The stirring speed was operated slowly during the second stage at 40 rpm for 10 minutes to promote flocculation and prevent the irreversible disruption of flocs caused by polymer chain breakage [94].

As a result, coagulant dosages from 0.02 to 0.10 g for *M. oleifera* and CH did not reduce the water turbidity to 5 – 10 NTU within the first 1 hour, as shown in Figure 8a and b. Extending the settling period to 2 hours allowed most dosages to reduce water turbidity to less than 10 NTU. On the other hand, turbidity removal was most effective when 0.06 g of *M. oleifera* and 0.02 g of CH were used at an initial turbidity of 300 NTU with a settling time of 110 minutes and 150 minutes. The individual CH removes kaolin gradually, as it predominantly depends on physical adsorption via silica instead of the charge neutralization procedure utilized by *M. oleifera*. This statement is further corroborated by the BET analysis, which previously indicated that CH adheres to a physisorption process. The CH can eliminate turbidity by a bridging mechanism; however, if floc production is unstable, the adsorbent cannot efficiently remove turbidity. Remarkably, both *M. oleifera* and CH demonstrated exceptional performance by consistently achieving this stringent standard of National Water Quality Standard Malaysia (NWQSM) [95], which dictates turbidity levels below 5 NTU, underlining its effectiveness as a coagulant [96]. A study by Adelodun [97] reported that the turbidity removal increased correspondingly with high *M. oleifera* dosage that allows more formation of flocs through the bridging mechanism. However, it

has been noted that the removal efficiency decreases when using higher dosages of coagulants than their optimal value due to their inability to interact with oppositely charged colloidal particles [27]. This study supports the hypothesis that peak efficiency is reached when the coagulant dosage and turbidity levels are optimal [18].

Turbidity removal performance of different combined MOCH ratios

The optimal coagulant dosage is divided into the corresponding combined MOCH ratio to assess the coagulant combination's efficacy. Five distinct ratios were formed by combining the three optimal dosages of specific coagulants to determine which combination of the MOCH ratio achieves the highest level of turbidity removal at 300 NTU from the synthetic wastewater. The combined coagulant MOCH was prepared in different mixing ratios of *M. oleifera* to coconut husk (MOCH) such as 1:3 (0.02 g : 0.06 g), 3:1 (0.06 g : 0.02 g), 1:4 (0.02 g : 0.08 g), 4:1 (0.08 g : 0.02 g), and 4:5 (0.08 g : 0.10 g). Figure 9 illustrates the turbidity reduction using different combined MOCH ratios.

The outcomes indicated that the MOCH 3:1 combination with the ideal dosage for *M. oleifera* was 0.06 g and CH was 0.02 g exhibited superior performance compared to the other combined MOCH ratios. The MOCH 3:1 facilitates more rapid and stable floc formation than individual coagulants due to its synergistic impact. This is because *M. oleifera* aids in the neutralization process by interacting with positively charged kaolin particles, while the silica

compound in CH contributes to the formation of stronger and more stable floc through a bridging mechanism. Conversely, MOCH 1:3 demonstrates lower turbidity removal than MOCH 3:1, likely due to a higher dosage of CH that impedes adsorbent efficacy [98]. Meanwhile, MOCH 4:5 showed the lowest efficacy in eliminating turbidity, attributed to excessive coagulant dosage leading to reduced removal capacity. This statement was supported by Getahun et al. [99], who stated that excessive use of coagulants could impact turbidity removal because of the electrostatic repulsive interaction between the biosorbents and colloidal particles in the wastewater.

Therefore, excessive amounts of *M. oleifera* or CH can reduce coagulation efficiency, as oversaturation from *M. oleifera* may cause complete occupation of active sites, while an excess of CH can lead to excessive bridging and unstable floc formation due to disrupted particle aggregation. Next, MOCH 1:4 and MOCH 4:1 share almost similar turbidity removal after 1 hour, and both combination ratios can eliminate more turbidity than MOCH 4:5 and MOCH 1:3. This could be ascribed to the balance of optimal coagulant dosage from both *M.oleifera* and CH that facilitate to elimination the contaminants from the synthetic wastewater [97]. During the coagulation-flocculation process, it was noted that MOCH 3:1 consistently showed the most effective pattern for removing turbidity compared to that of other solutions. Thus, MOCH 3:1 provides the optimal combination for eliminating contaminants from the synthetic wastewater.

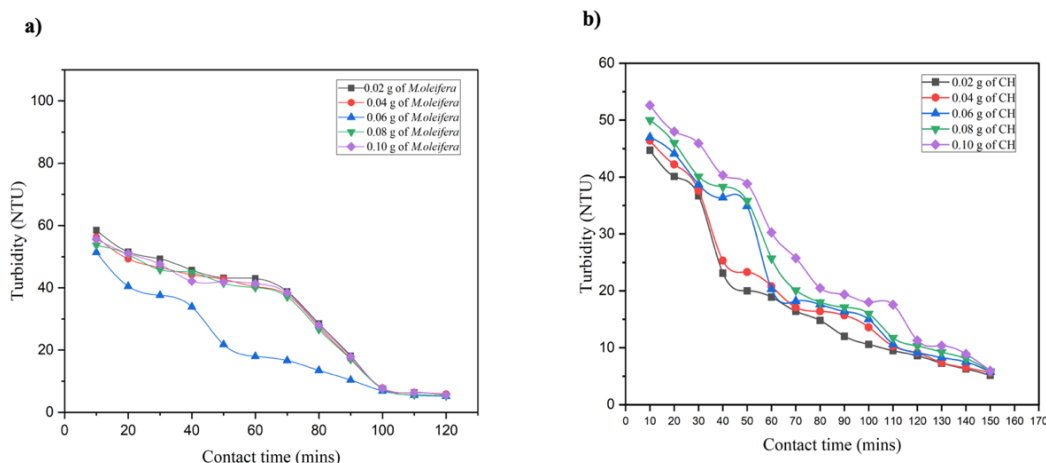


Figure 8. Turbidity reduction of 300 NTU synthetic wastewater using (a) *M.oleifera* (b) CH within 2h settling time at pH 6 and 25 °C

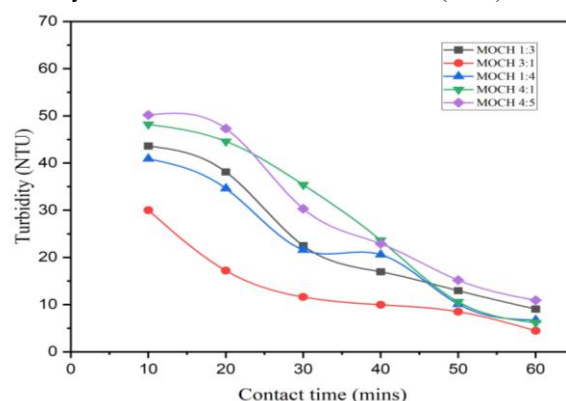


Figure 9. Turbidity reduction of 300 NTU synthetic wastewater using different combined MOCH ratios within 1h settling time at pH 6 and 25 °C

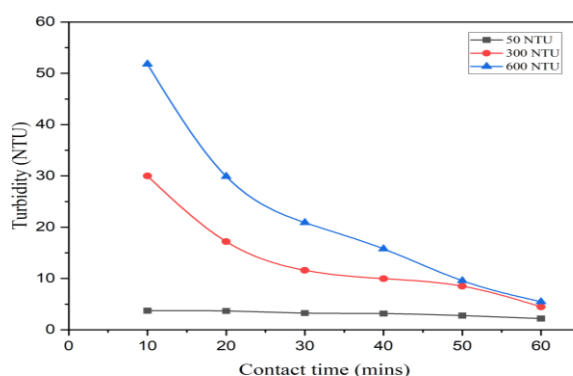


Figure 10. Turbidity reduction of synthetic wastewater with varying initial turbidity levels using MOCH 3:1 (0.06 g of *M.oleifera*: 0.02 g of CH) within 1h settling time at pH 6 and 25 °C

Turbidity removal performance of the combined MOCH 3:1 dosage

The best coagulant dosage from the respective coagulants was chosen in a ratio to compare the performance based on their coagulation efficiency towards turbidity removal. This study used a combined dosage of 0.06 g of *M.oleifera* and 0.02 g of CH with a ratio of 3:1 as a biocoagulant for different initial turbidity levels, which were low turbid wastewater (50 NTU - 200 NTU), medium turbid wastewater (250 NTU - 400 NTU), and high turbid wastewater (450 NTU - 600 NTU). **Figure 10** shows the result for the turbidity removal by MOCH 3:1 at different turbidity levels, which are 50 NTU, 300 NTU, and 600 NTU. The MOCH 3:1 can reduce low turbid wastewater rapidly after 10 minutes from 50 NTU to 3.72 NTU, which has achieved the NWQSM. Next, the MOCH 3:1 can reduce the turbidity to the desired level, which was below 5 NTU within 1 hour at all turbidity levels, as shown in Figure 10. The MOCH 3:1 coagulant demonstrated outstanding performance with efficiency rates of $99\% \pm 0.56$ for high, medium, and low initial turbidity levels in water samples. The MOCH 3:1 exhibits high effective removal at low concentrations of suspended particles

(50 NTU) by neutralizing charges and promoting flocculation, which enhances particle aggregation and elimination through accessible active sites of the coagulants. At high turbidity levels (600 NTU), both coagulants exhibit lower efficacy, likely due to the oversaturation of active sites and inadequate bridging ability to accommodate the increased particle volume. The high particle concentration in the solution may surpass the coagulation mechanism, resulting in diminished stability and the development of larger, less stable flocs, hence decreasing the efficiency of turbidity removal. **Figure 11** represents the comparison between MOCH 3:1 and the best coagulant dosages from *M. oleifera* and CH. This shows that MOCH 3:1 significantly enhanced the reduction of turbidity from the synthetic wastewater compared to the use of respective coagulants. A comparable study conducted by Dandesa et al. [100] revealed a contrasting outcome, as the use of *M. oleifera* alone was ineffective in removing turbidity from low to high levels, thereby failing to meet the NWSQM criteria. Moreover, Vidaurre et al. [101] mentioned that the turbidity removal effectiveness decreases as turbidities rise to 385 NTU, 471 NTU, and 632 NTU when using 100 mg/L of *M. oleifera*.

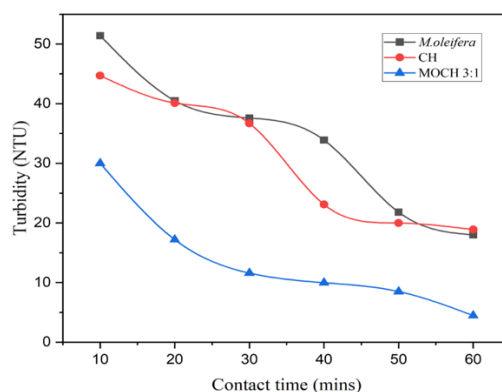


Figure 11. Turbidity reduction of 300 NTU synthetic wastewater using respective coagulants within 1h settling time at pH 6 and 25 °C

According to the study by Rasheed et al. [18], The *M. oleifera* coagulant performed exceptionally well, exhibiting 89%, 80%, and 42% effectiveness rates for water samples with high, medium, and low initial turbidity levels [102]. Besides that, a few studies also reported that turbidity reduction by using *M. oleifera* requires 3 to 4 hours of treatment to achieve the water quality standard [19, 103]. The majority of research findings indicate that the removal efficacy of water turbidity samples with a greater *M. oleifera* dosage is enhanced in the presence of dense suspended solids; however, the efficacy is reduced in low turbidity water [104, 105]. Meanwhile, several studies reported that a high dosage of coagulant, above 5 g, is necessary to treat high turbidity over 150 NTU effectively but high protein content in *M. oleifera* serves as a nutrient source for bacteria, which can promote bacterial growth in water if large dosages are used [106, 107].

The dosage mentioned is significantly higher than the concentrations used in this experiment. This method was advantageous in this study because it only required a small dosage of the biocoagulants to attain the desired turbidity level. Therefore, this study proved that combined MOCH 3:1 achieved the highest turbidity removal at all turbidity levels within one hour compared to the previous study. This is because the heterogeneous surface of MOCH 3:1 structure offers many opportunities for particle bridging and synergistic effect from combined coagulant that help to enhance coagulation-flocculation and speeding up impurity sedimentation by gravitational force [19]. This statement is also supported by Benettayeb et al. [80], which mentioned that the presence of negatively charged groups on the *M. oleifera* surface could lead to electrostatic interactions and hydrogen bonding, allowing the pollutants to be adsorbed followed by the flocculation process.

Effect of temperature on turbidity removal

The impact of temperature on removing turbidity is a critical consideration in water treatment processes. Therefore, it is essential to identify the optimal temperature range for coagulation properties. The most turbidity reduction can be achieved at 50 NTU, followed by 300 NTU, while the lowest turbidity removal can be observed at 600 NTU using combined MOCH 3:1. The trend of turbidity removal decreased as the temperature increased from 25 °C to 100 °C, as shown in **Figure 12**. At 25 °C, the maximum turbidity removal was achieved using MOCH 3:1 at 50 NTU, as the low initial particle concentration enhanced coagulant-particle interactions, promoting efficient flocculation and turbidity elimination. The lowest turbidity removal using MOCH 3:1 was recorded at 600 NTU. This phenomenon can be attributed to particle overcrowding at high turbidity levels, which creates a complex suspension where competition for coagulant binding sites hinders complete neutralization and floc formation, thereby reducing overall removal efficiency.

Overall, the optimum turbidity removal can be observed at room temperature 25°C. This result aligns with findings demonstrating that high turbidity removal can be accomplished at room temperature [108, 109]. Higher temperatures might also affect fluid dynamics and lessen the efficiency of coagulants due to protein denaturation, which could lead to shorter settling times and less effective turbidity removal [108]. On the contrary, another study by Maćczak et al. [109] revealed that elevated temperatures enhance the efficiency of coagulation and flocculation, resulting in improved turbidity removal due to accelerated chemical reactions and more efficient particle aggregation. The results of this study differ from the findings of Elsergany [110], which indicated that the optimal turbidity removal by

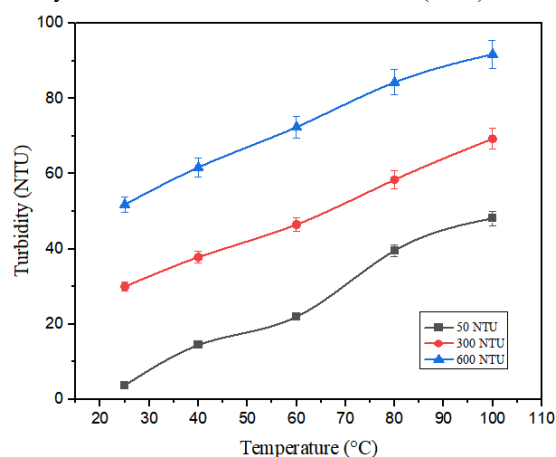


Figure 12. Effect of temperature on turbidity reduction of synthetic wastewater with varying initial turbidity levels using MOCH 3:1 after 1h settling time at pH 6

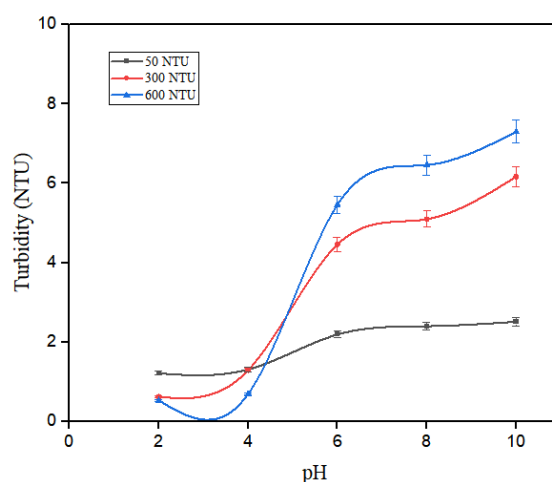


Figure 13. Effect of pH on turbidity reduction of synthetic wastewater with varying initial turbidity levels using MOCH 3:1 after 1h settling time at 25 °C

utilising *M.oleifera* occurs at 60°C while the lowest turbidity removal is observed at room temperature. Therefore, the combined MOCH 3:1 does not necessitate high temperature to achieve optimal turbidity removal, potentially leading to energy savings by eliminating the need for excessive heating.

Effect of pH on turbidity removal

The selection of an appropriate pH for each material can have an impact on the biosorption process since variations in water pH can modify groups and impact the surface charge of biosorbents as well as competition between ions for binding sites, which can either improve or reduce sorption effectiveness [111]. A pattern of increase was observed as the pH levels were raised to 50 NTU, 300 NTU, and 600 NTU by using MOCH 3:1, as illustrated in **Figure 13**.

The interaction between the biocoagulants and the kaolinite surface can be attributed to multiple causes,

leading to improved adsorption. MOCH 3:1 achieved the highest turbidity reduction at high turbidity level (600 NTU) and pH 2, lowering it to 0.53 NTU after 1 hour, while increasing the pH to 10 resulted in the lowest turbidity removal of 7.3 NTU. The positive charge on the kaolinite surface is pH-dependent and intensifies with decreasing pH values because the surface of the kaolinite material possesses active sites that become positively charged at lower pH values due to the presence of $\text{Al}(\text{OH})^{2+}$ species [112]. At pH 2, electrostatic attraction between the positively charged kaolinite surface and the biosorbents enhances adsorption and floc formation. However, at higher pH levels, particularly at pH 10, zeta potential measurements indicate an increase in negative surface charge on both kaolinite and the biosorbents, leading to electrostatic repulsion that hinders aggregation. This combined with increased viscosity and reduced collision frequency, results in weaker flocs and lower

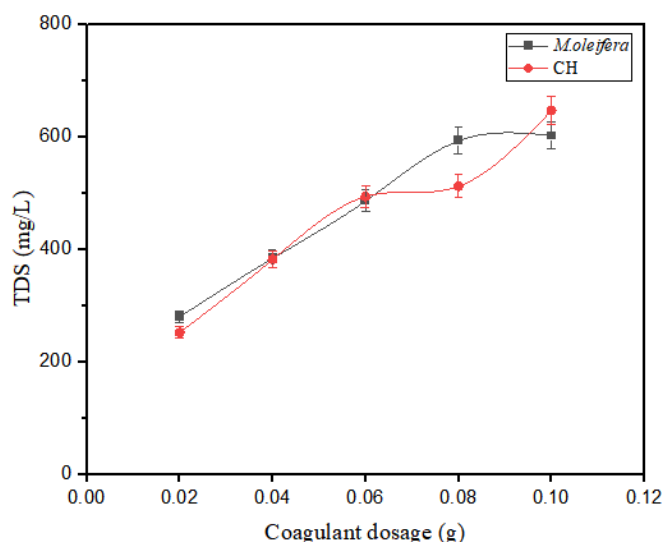


Figure 14. TDS removal at 300 NTU synthetic wastewater using respective coagulant after 1h settling time at pH 6 and 25 °C

turbidity removal efficiency. In contrast, the least turbidity removal was shown by MOCH 3:1 at 50 NTU and pH 2 of synthetic wastewater which was 1.21 NTU, whereas the most significant reduction occurred as increase the pH level to pH 10, reaching 2.51 NTU. This is because the reduced particle concentration in the synthetic wastewater restricted collision frequency, resulting in inadequate floc formation and unsatisfactory turbidity reduction. Nevertheless, MOCH 3:1 exhibited the greatest turbidity reduction of synthetic wastewater at 50 NTU and pH 10 due to diminished competition for flocculation sites at decreasing particle concentrations, facilitating the production of larger and more stable flocs. These flocs demonstrated superior efficacy in sedimentation and turbidity removal.

Lulesa et al. [113] found that the coagulation efficiency of *M.oleifera* could effectively remove turbidity when raising the pH of water from 5 to 11. Additionally, some studies explained that increasing the dosage of *M.oleifera* can raise the water's pH to a level that fulfils government standards [114]. Conversely, several studies reported that the observed drop in water pH aligns with an increase in the concentration of *M.oleifera* dosage [115, 116]. A study by Varsani et al. [40] suggested that a significant decrease in pH was detected in *M.oleifera* coagulant when treated with HCl, with a recorded value of 7.14, followed by NaCl, NaOH, and naturally treated *M.oleifera*, which had pH values of 7.29, 7.9, and 8.01, respectively. However, the combined MOCH 3:1 exhibited a substantial and considerable decrease in pH, transitioning from an alkaline state and acidic condition to a neutral pH (6-7). The pH values

measured fall within the acceptable range (6.5–8.5) as specified by NWQSM for drinking water [96]. Thus, the study demonstrated that combined MOCH 3:1 had improved coagulation-flocculation by destabilising positively charged particles without requiring further pH and dosage adjustment to achieve better turbidity removal. Overall, this method is highly advantageous for its non-toxic nature and cost-effectiveness and aligns with the green chemistry principle to maximise its coagulation efficiency.

Total dissolved solids

The investigation revealed that the initial TDS in untreated water reached a maximum of 2454 mg/L with a turbidity level of 300 NTU. Following treatment with both coagulants, there was a drop in the TDS level at a turbidity level of 300 NTU from 2170 mg/L to 280 mg/L and 252 mg/L by using 0.02 g of *M.oleifera* and CH, respectively. According to WHO standards, water with less than 600 mg/L is considered appropriate, while concentrations above 1000 mg/L indicate unpleasant water in which pre-treatment may be necessary before use [117]. Nonetheless, the increase in coagulant dosage also led to a rise in TDS levels. There is a clear relationship between the amount of coagulant dosages supplied and the increase in TDS. Higher levels of biocoagulants lead to higher total solids, as illustrated in Figure 14. The lower dosage of activated CH and *M.oleifera* reduces the TDS the most after 1 hour. The experiment showed that post-treatment for removing TDS content varied between 252 and 647 mg/L. This observation aligns with the study by Shan et al. [30], which documented a rise in TDS in water when the coagulant dosage increased due to release of soluble organic and inorganic compounds from both coagulants such as

amino acid, sugar, calcium, magnesium and potassium. Kitheka et al. [103] found a notable rise in TDS in water as the concentration of *M. oleifera* increased. This statement is also supported by Vidaurre et al. [101]. Other than that, an increase in CH dosage correlated with a rise in TDS, suggesting that greater dosages facilitated the release of more soluble components into the solution [25]. Thus, the findings are consistent with those obtained in this research.

Figure 15 displays a positive correlation between the TDS content and turbidity levels when MOCH 3:1 was used. The concentration of TDS rises progressively as the turbidity levels increase. The increased TDS values are associated with elevated in turbidity levels caused by high concentrations of sediments in the water samples [116]. It was observed that the highest reduction in TDS was 147 mg/L achieved with the combined MOCH 3:1 coagulant at the lowest turbidity level of 50 NTU. The concentration of TDS rises progressively as the turbidity level increases from medium turbidity to high turbidity level, with TDS values ranging from 195 mg/L (300 NTU) to 414 mg/L (600 NTU). A sudden rise was detected after 400 NTU, indicating that the coagulants may be approaching a threshold beyond which they cease to efficiently aggregate the particles. This occurs because the coagulant becomes oversaturated and begins to release soluble organic compounds such as proteins, polysaccharides, and amino acids, which lead to a rise in TDS concentration [38]. This will result in a greater quantity of soluble compounds remaining in the solution rather than being eliminated. This evidence proved that the MOCH 3:1

produces lower TDS values, even in high turbidity water, making it superior to individuals used alone. Nevertheless, these values still meet the palatable classification since the standard TDS guideline for drinking water based on taste is 1000 mg/l. The combined treatment moderately reduced TDS the most compared to individual treatments at similar levels of 300 NTU. A significant reduction in the TDS in water samples when the combined MOCH 3:1 is used, predominantly in low turbid wastewater. It was observed that the combined MOCH 3:1 was able to give the highest reduction in TDS, from 2170 mg/L to 195 mg/L, compared to both coagulants. This phenomenon can be attributed to the enhanced properties of the MOCH 3:1 surface, which *M. oleifera* effectively neutralizes the charge on the particles, and the extra number of adsorption sites provided by CH enhances the coagulation-flocculation process, facilitating the efficient removal of dissolved solids from wastewater [118]. This implies that the MOCH 3:1 can effectively form larger and denser flocs, thus reducing the TDS content at 300 NTU effectively compared to individual coagulants.

Water properties, treatment agent dosage, and the interaction between *M. oleifera* and CH influence the efficacy of TDS removal. *M.oleifera* can coagulate and flocculate suspended particles and dissolved solids because of its protein content while CH exhibits powerful adsorption properties by efficiently absorbs both organic and inorganic substances, aiding in the elimination of TDS [119]. Thus, combining *Moleifera* and CH can synergistically increase the removal efficiency for a wider variety of pollutants.

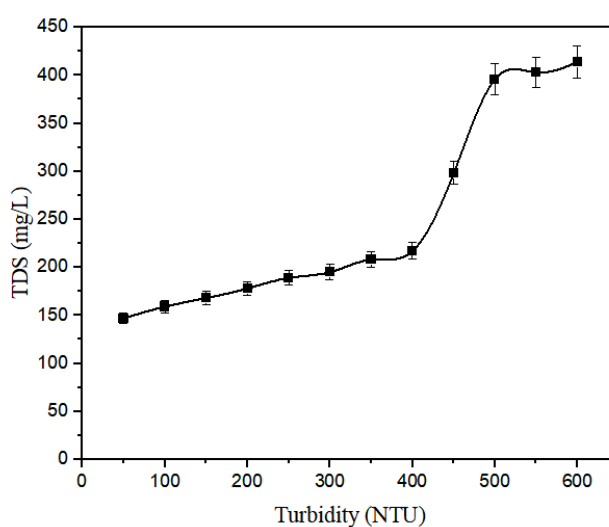


Figure 15. TDS removal at different turbidity levels of synthetic wastewater using MOCH 3:1 after 1h settling time at pH 6 and 25 °C

Total suspended solids

The initial total suspended solids (TSS) in wastewater samples was 2790 mg/L at 300 NTU. Both biocoagulants exhibited a similar increase pattern, with the TSS increasing with a higher coagulant dosage, as shown in **Figure 16**. After 1 hour, the highest TSS content recorded was 26 mg/L, exceeding the acceptable limit of 25 mg/L, when employing 0.10 g of *M.oleifera* and CH, respectively. Meanwhile, the lowest TSS content in synthetic wastewater was found to be 22.7 mg/L when using 0.02 g dosage from each biocoagulant. All dosages of both biocoagulants, except 0.10 g, led to a decrease in the TSS level according to the NWQSM standard, ranging from 22 to 25 mg/L. The maximum turbidity removal effectiveness was achieved with an ideal dose of 0.06 g of *M.oleifera*; nevertheless, a rise in TSS was noted. This process results from the efficient aggregation of particles enabled by the coagulant, resulting in the creation of larger and denser flocs. The presence of bigger aggregated flocs causes the quantifiable TSS to increase even though this process greatly decreases the tiny particles that cause turbidity. At dosages over 0.06 g, TSS levels increased further, likely due to an excess of coagulant surpassing the ideal quantity needed for effective charge neutralization and particle bridging. The surplus coagulant may engage with particles or self-aggregate, therefore leading to an increase in TSS. Conversely, at dosages below 0.06 g, the inadequate coagulant amount did not accomplish complete charge neutralization, leading to smaller, unaggregated particles remaining suspended, which further contributed to increased TSS levels.

The reduction in TSS level was due to the probable mechanisms implicated in this coagulation activity, including the adsorption and neutralization of charges or the adsorption and bridging of destabilized particles [120]. When the coagulant dosages of *M. oleifera* and CH in water samples increased, TSS particles rose. This occurred due to the re-stabilization process triggered by the reversal of the colloidal charge induced by adsorption and potential saturation of the polymer bridge sites in the coagulant [121]. Consequently, the destabilized particles could re-stabilize due to an inadequate number of particles available to construct additional interparticle bridges. Gaayda et al. [94] also noticed this phenomenon while studying the coagulation of synthetic turbid water using extracts derived from *M. oleifera* seeds. Few studies reported that adding a higher quantity of *M.oleifera* seed can significantly increase TSS levels [42]. Conversely, other studies also reported that a decrease in TSS was observed as the content of *M. oleifera* also increased [122, 123].

Next, the efficacy of the combined MOCH 3:1 was evaluated by testing it on wastewater with varying turbidity levels, ranging from low to high, to assess its coagulation efficiency, as depicted in **Figure 17**. The graph illustrates the consistent rise in TSS content as the turbidity level increases. It was noted that the combined MOCH 3:1 consistently obtained the lowest TSS levels, ranging from 7 mg/L to 23.5 mg/L at all turbidity levels, after 1 hour. The minimum TSS concentration was 7 mg/L at a turbidity level of 50 NTU, while the maximum TSS content was 23.5 mg/L at 600 NTU when a MOCH 3:1 was used. The lowest TSS content was attained by MOCH 3:1 at 15.2 mg/L when comparing the TSS removal at 300 NTU with individual coagulants. The MOCH 3:1 effectively removes the maximum TSS at low turbidity due to its ability to immediately interact with accessible particles, resulting in an improved flocculation process.

The synergistic benefits and complementary mechanisms of *M.oleifera* and CH result in reduced TSS in wastewater treatment compared to individuals. This is because activated CH has high porosity properties that can help to facilitate the floc adhesion, leading to the formation of larger and denser flocs, which help it to settle more efficiently by adsorbing a more comprehensive range of dissolved organic and inorganic compounds, including those that *M.oleifera* may not be able to address on its own [124, 125]. Additionally, the TSS content begins to increase beyond 400 NTU, suggesting that larger particles may compete for the available coagulant, resulting in some particles not being sufficiently neutralized and leading to the development of excess floc [126]. It was shown that the combined MOCH 3:1 successfully removed TDS and TSS with more than 95% at all turbidity levels. Therefore, it was proved that combined MOCH 3:1 may lead to a more thorough elimination of suspended particles and other impurities, which will enhance the coagulation and flocculation process.

Electrical conductivity

Electrical conductivity (EC) is a quantitative assessment of the amount of dissolved solids present in water, which can vary significantly across different geographical areas due to variations in the ability of minerals to dissolve [127]. Based on NWQSM, the value of EC below 1000 $\mu\text{S}/\text{cm}$ is acceptable, but elevated conductivity in drinking water may be undesirable for consumers [89]. The coagulants were employed for the treatment of synthetic wastewater with an initial EC value of 2440 $\mu\text{S}/\text{cm}$. All coagulants demonstrated the ability to reduce the EC of synthetic wastewater after 1 hour as shown in **Figure 18**.

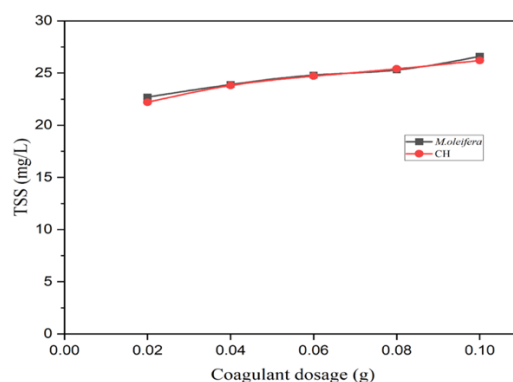


Figure 16. TSS removal at 300 NTU synthetic wastewater using respective coagulant after 1h settling time at pH 6 and 25 °C

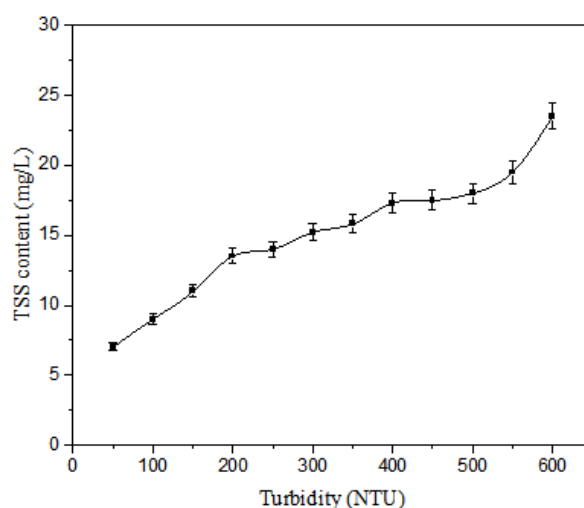


Figure 17. TSS removal at different turbidity levels of synthetic wastewater using MOCH 3:1 after 1h settling time at pH 6 and 25 °C

The water samples treated with different dosages of *M.oleifera* showed some variation from 499 $\mu\text{S}/\text{cm}$ to 519 $\mu\text{S}/\text{cm}$. The *M.oleifera* exhibited a notable reduction in conductivity with a coagulant dosage of 0.02 g. Nonetheless, there was an increase in conductivity as the coagulant dosage increased. The high water conductivity in this study can be attributed to the elevated TDS content from adding coagulants to the wastewater [101]. This may be linked to the release of ions such as K^+ , Ca^{2+} , Mg^{2+} , and COOH^- from *M.oleifera*. High quantity of *M.oleifera* could contribute to high levels of cationic polyelectrolyte found in *M.oleifera* seeds, which resulted in more tremendous dissolved solids and thus increased conductivity [121]. There is a positive correlation between the concentration of *M.oleifera* seed and the conductivity of wastewater [30]. According to Kithika et al. [103], There was a notable rise in the EC of water as the concentration of *M.oleifera* increased due to the presence of anionic polyelectrolyte in the seeds. Other studies also supported this statement that a proportional relationship exists between

conductivity and concentration in water samples treated with *M.oleifera* [126]. In addition, the sodium chloride in the salt extract is responsible for the increased trend in EC because sodium chloride increases the ionic strength of the solution, which in turn enhances the solubility of both coagulants [28, 114]. According to Varsani et al. [40], The active component of *M.oleifera* from the salt solution can cause the dispersion of specific mineral ions and inorganic compounds from the synthetic wastewater into a floc, which is then separated from the water suspension through precipitation, leading to a minor decrease in EC. This aligns with the observation that a decrease in EC was noted after applying 0.06 g of *M.oleifera* and dosages above, indicating that a greater number of ions are adsorbed onto the surplus *M.oleifera* particles in the solution as shown in Figure 18. Conversely, the current findings contradict the study conducted by Thorat et al. [17], which showed that higher concentrations of *M.oleifera* coagulants resulted in a decrease in conductivity values when using a lower dosage than the optimum value.

Next, the CH showed a decreasing pattern followed by a slightly increased EC value when high dosage was used, as shown in **Figure 18**. The values varied from 524 $\mu\text{S}/\text{cm}$ to 432 $\mu\text{S}/\text{cm}$, suggesting a considerable decrease in EC as the dosage of CH increased from 0.02 g of CH to 0.06 g of CH. This occurs because CH functions as a natural ion exchanger, facilitating the adsorption of more ions on its surface, hence reducing the concentration of free ions in the solution [128]. Despite a reduction in EC when utilizing CH with high TDS content, the silica, cellulose, and lignin components of CH exhibited poor ionization, hence elevating the TDS level without increasing the EC itself [129]. However, the EC values rose when the dosage of CH increased from 0.08 g (453 $\mu\text{S}/\text{cm}$) to 0.10 g (471 $\mu\text{S}/\text{cm}$) due to the organic component from the CH dissolving and interacting with the existing ions, thereby increasing the ionic strength.

The combined MOCH 3:1 was tested on the removal of EC from low (50 NTU) to high (600 NTU) turbidity levels as illustrated in **Figure 19**. There was an increasing trend of EC values after 1 hour from a low turbidity level, 50 NTU (184 $\mu\text{S}/\text{cm}$), to a high turbidity level, 600 NTU (488 $\mu\text{S}/\text{cm}$). The combined

MOCH 3:1 exhibited the lowest conductivity value at 300 NTU compared to the individual coagulants, which was 245 $\mu\text{S}/\text{cm}$. The synergistic effect from the combined MOCH 3:1 results in low EC, potentially removing ions by coagulation, flocculation, and adsorption processes. This statement is supported by Tahraoui et al. [125], who stated that the reduction of EC was due to the coagulation-flocculation method using the proper dosage of biosorbents. The MOCH 3:1 successfully eliminates EC at low turbidity levels below 300 NTU by adsorbing more ions into the coagulant and facilitating strong floc formation through the synergistic effects of *M.oleifera* and CH. There was a significant increase in electrical conductivity from 350 to 450 NTU due to high dissociation capacity in water from *M.oleifera* and CH [126]. The combined MOCH 3:1 has attained a threshold of 350 NTU, resulting in the release of excessive soluble ions from the coagulant in the solution as turbidity levels rise. Hence, the conductivity of water treated with biosorbents, as reported in this work, falls significantly below the permitted conductivity limit (1000 $\mu\text{S}/\text{cm}$) for drinking water according to NWQSM standards.

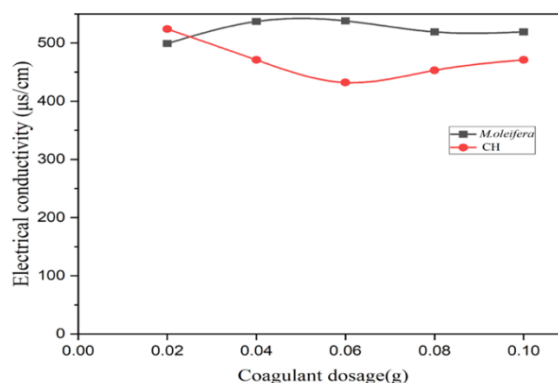


Figure 18. EC removal at 300 NTU synthetic wastewater using respective coagulant after 1h settling time at pH 6 and 25 °C

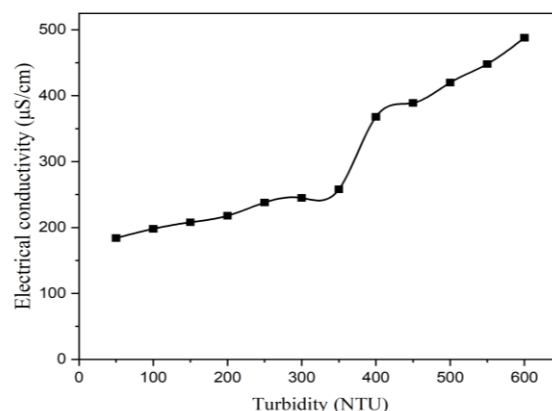


Figure 19. EC removal at different turbidity levels of synthetic wastewater using MOCH 3:1 after 1h settling time at pH 6 and 25 °C

Table 4. Summary of treating synthetic water with respective biosorbents

Type of Coagulants	Turbidity (NTU)	TSS (mg/L)	TDS (mg/L)	EC ($\mu\text{S/cm}$)	Settling time (mins)
0.06 g of <i>M.oleifera</i>	5.48	24.8	486	538	110
0.02 g of CH	5.18	22.2	494	524	140
MOCH 3:1	4.46	15.2	195	245	60

Overall, MOCH 3:1 enhanced the elimination of turbidity, TSS, TDS, and EC within 1 hour, compared to the individual coagulants, from the synthetic wastewater with a turbidity value of 300 NTU and pH 6 at 25°C, as shown in **Table 4**. This enhancement provides numerous benefits in terms of treatment efficiency, cost-effectiveness, and environmental sustainability. Improved removal efficiency results in clearer water with lower levels of contaminants, ensuring more constant compliance with regulatory criteria. The utilization of MOCH 3:1 can reduce the dependence on larger quantities of individual coagulants and minimise the agricultural wastes in landfills. The benefits of combining both composite powders underscore their high potential in enhancing wastewater treatment procedures and promoting more eco-friendly water management practices.

Conclusions

In conclusion, the surface characteristics of *M.oleifera*, coconut husk (CH) and MOCH 3:1 have been analysed using SEM, FTIR, BET, and zeta potential. The SEM images show that pollutants from wastewater accumulate on the coagulants' surfaces, leading to increased softness and contact between the biosorbents and kaolin particles. This is due to the presence of hydroxyl, amide and carbonyl groups on the surface of biosorbents, which interact with the kaolin particles through hydrogen bonding. The MOCH 3:1 composite has a larger surface area and pore volume than the individual *M.oleifera*, while CH has the highest surface area and pore volume. All biosorbents showed negative surface charges at all pH levels, indicating electrostatic attraction with oppositely charged particles. This study also evaluated the effectiveness of individual coagulants and combined MOCH 3:1 in eliminating turbidity based on pH and temperature, TDS, TSS, and EC. The most suitable for individual coagulant dosages for *M.oleifera* and CH were 0.06 g and 0.02 g, but these coagulants required 110 and 150 minutes, respectively to achieve the turbidity standard. The turbidity removal (99.46%) was achieved using MOCH 3:1, reducing it to below 5 NTU at all turbidity levels within 1 hour at 25 °C and pH 6. The findings suggest that the MOCH 3:1 does not require pH adjustment and high temperature to maximize the turbidity removal. In addition, the highest removal rate for TDS contents (195 mg/L) and TSS contents (15.2 mg/L)

from synthetic wastewater was demonstrated using MOCH 3:1 at 300 NTU compared to *M.oleifera* and CH. The most reduction EC with less than 500 $\mu\text{S/cm}$ was shown by MOCH 3:1. Therefore, the results suggest that incorporating CH can improve the efficiency of *M.oleifera* in wastewater treatment.

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