



Faculty of Resource Science and Technology

Preparation and Characterization of *Moringa oleifera* and Coconut Husk Hybrid Coagulant as Bioadsorbent for Wastewater Treatment Application

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Preparation and Characterization of *Moringa oleifera* and Coconut Husk Hybrid Coagulant
as Bioadsorbent for Wastewater Treatment Application

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A thesis submitted
in fulfilment of the requirements for the degree of Master of Science
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DECLARATION

I declare that the work in this thesis was carried out in accordance with the regulations of University Malaysia Sarawak. Except where due acknowledgements have been made, the work is that of the author alone. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

A handwritten signature in black ink, consisting of several loops and strokes, positioned above a horizontal dotted line.

Signature

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ABSTRACT

Moringa oleifera has gained significant attention from researchers due to its efficacy as a natural adsorbent for removing pollutants from wastewater. However, this particular *M.oleifera* is incapable of effectively removing turbidity, total dissolved solids (TDS), total suspended solids (TSS), and electrical conductivity (EC) from wastewater. This necessitates chemical modifications that use concentrated acid and alum, which is not a favourable combination. Thus, this study reported the potential use of combined *M.oleifera* with activated coconut husk (CH) for water treatment. The effectiveness of using *M.oleifera*, CH, and a combination of 0.06 g of *M.oleifera* and 0.02 g of CH to form MOCH 3:1, were 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). The TDS, TSS, and EC reduction from synthetic wastewater were also studied using respective coagulants. The surface properties of these materials were examined through the utilization of Fourier Transform Infrared (FTIR) Spectroscopy, Scanning Electron Microscope (SEM), Brunauer-Emmett-Teller-Nitrogen adsorption (BET-N₂), and zeta potential. The MOCH 3:1 agreed with the Langmuir, indicating monolayer formation and pseudo-second-order kinetic models, suggesting chemisorption, with R² value of 0.99. 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 finding suggests that MOCH 3:1 has superior potential for wastewater contaminant removal than individual coagulants.

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

***Penyediaan dan Pencirian Koagulan Hibrid Moringa oleifera dan Sabut Kelapa
Sebagai Bioadsorben Untuk Aplikasi Rawatan Air Kumbahan***

ABSTRAK

Moringa oleifera telah menarik perhatian yang signifikan daripada para penyelidik kerana keberkesannya sebagai adsorben semula jadi untuk menyingkirkan bahan pencemar daripada sisa kumbahan. Namun begitu, M. oleifera tidak mampu menghilangkan kekeruhan, jumlah pepejal terlarut (TDS), jumlah pepejal terampai (TSS), dan kekonduksian elektrik (EC) daripada air kumbahan dengan berkesan. Ini memerlukan pengubahsuaian kimia seperti asid pekat dan alum, yang bukan kombinasi yang baik. Oleh itu, kajian ini melaporkan potensi gabungan antara M. oleifera dan sabut kelapa (CH) untuk rawatan air. Keberkesanan penggunaan M. oleifera, CH, serta gabungan 0.06 g M. oleifera dan 0.02 g CH untuk membentuk MOCH 3:1, dinilai berdasarkan penyingkiran kekeruhan pada pelbagai tahap pH (2, 4, 6, 8 dan 10) dan suhu (25 °C, 40 °C, 60 °C, 80 °C dan 100 °C). Pengurangan TDS, TSS, dan EC daripada air sisa sintetik juga dikaji menggunakan koagulan tersendiri. Sifat permukaan koagulan telah dikaji menggunakan Spektrometer Infra Merah (FTIR), Pengimbas Mikroskop Elektron (SEM), Brunauer-Emmett-Teller-Nitrogen (BET-N₂), dan potensi zeta. MOCH 3:1 mematuhi model Langmuir (monolayer) dan model kinetik pseudo-tertib kedua (kemisorpsi) dengan nilai R² sebanyak 0.99. MOCH 3:1 mempunyai kesan sinergistik dengan penyingkiran tertinggi melebihi 90% untuk kekeruhan, TDS, TSS dan EC daripada air sisa sintetik pada pH 6 dan 25°C. Kajian mencadangkan bahawa MOCH 3:1 mempunyai potensi cemerlang untuk menyingkirkan bahan pencemar daripada sisa kumbahan berbanding dengan koagulan individu.

Kata kunci: Moringa oleifera, sabut kelapa, kekotoran, air sisa, rawatan

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LIST OF ABBREVIATIONS

ANOVA	One-way Analysis of Variance
APHA	American Public Health Association
BOD	Biochemical Oxygen Demand
CH	Coconut husk
COD	Chemical Oxygen Demand
DOE	Department of Environment
DO	Dissolved Oxygen
EC	Electrical conductivity
EDTA	Ethylenediaminetetraacetic
FMO	Functionalized <i>Moringa oleifera</i>
FTIR	Fourier Transform Infrared Spectroscopy
MF	Microfiltration
MOCH	Combination of <i>Moringa oleifera</i> and CH
NWQSM	National Water Quality Standard Malaysia
RO	Reverse Osmosis
SDG	Sustainable Development Goals
SEM	Scanning Electron Microscopy
TDS	Total Dissolved Solids
TSS	Total Suspended Solids
THM	Trihalomethanes
WHO	World Health Organisation

CHAPTER 1

INTRODUCTION

1.1 Background

Water is an integral part of life and is pivotal in sustaining human civilisation and the ecosystem. Human societies utilise a massive amount of water for their daily activities, but they also discharge contaminants back into the environment, which alters the origin of water quality characteristics. The world's water system will soon approach a critical threshold with over 40% of the world's population is anticipated to experience water scarcity in the upcoming decades due to the proliferation of human consumption that is forcing the demand for water resources (Bartolomeu et al., 2018). Plus, only a small part of the water fraction is suitable for direct human use, with groundwater accounting for 96.8%, while 3.18% and 0.02% come from lakes and river water, respectively. In contrast, most unconventional water sources are desalinated and treated wastewater (Alsharhan & Rizk, 2020). As a result, 700 million people have insufficient access to clean drinking water, and it is estimated that the upward trend of population size to 2.7 billion people will face a water crisis by 2050, if this matter continually becomes a subject of ignorance for all parties (Musie & Gonfa, 2023; Shemer et al., 2023). Therefore, there are several water treatment technologies have been employed to treat contaminants from the wastewater, including physical treatment, chemical treatment and biological treatment (Ahmed et al., 2021). The implementation of wastewater treatment techniques is dependent upon acceptable effluent standards, operational costs, and environmental compatibility, with recent studies concentrating on enhancing treatment efficacy through a combination of established

techniques such as coagulation-flocculation and sedimentation (Mansour et al., 2024), ion-exchange (Jasim & Ajjam, 2024), adsorption (Satyam & Patra, 2024), electrolysis (Shah et al., 2024), membrane separation (Sravan et al., 2024), reverse osmosis (Tayeh, 2024) and ultrafiltration (Alomar et al., 2023). Nonetheless, most of these methods have presented some significant drawbacks, including high sludge generation that necessitates additional treatment, low removal efficiency, and high energy requirements (Oladimeji et al., 2024). The application of aluminium sulphate as a primary coagulant in the coagulation-flocculation process is frequently implemented in water treatment plants to remediate wastewater (Iwuozor, 2019). However, 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 (Jesus et al., 2024). Thus, the use of natural coagulants have gained significant attention as viable alternatives to replace conventional coagulants. The application of *M.oleifera* has gained the highest popularity as a natural coagulant due to its excellent coagulant properties, being biodegradable, non-toxic, locally available and cost-effective in treating wastewater treatment while minimizing environmental impact (Andrade et al., 2021). These benefits arise from its capacity to hinder soil and water acidification, reduce toxicity to aquatic organisms, and produce less sludge than aluminium sulphate in wastewater treatment (Desta & Bote, 2021). Comparatively, the combination of coagulation-flocculation and adsorption process are usually employed simultaneously to increase the efficiency of *M.oleifera* as coagulant in treating wastewater since it is inexpensive, convenient and easy to perform (Zahmatkesh et al., 2024). The biosorption which is a specific type of adsorption, has recently gained recognition as an environmentally sustainable and efficient alternative technology, involving a physiochemical process whereby pollutants from wastewater are adsorbed onto the surface of a biosorbent

(biomaterial) through mechanisms such as Van der Waals forces, covalent bonds, or other residual interactions (Shrestha et al., 2021). Various waste materials were reportedly converted into biosorbents for pollutants removal from wastewater such as green peas waste (Holliday et al., 2024), peanut shells (Lara et al., 2023), banana peel (Tjahjanti et al., 2021), chitosan (Crini et al., 2019) and rice husk (Shamsollahi & Partovinia, 2019). Nevertheless, the study results indicated that these adsorbents have constrained adsorption efficiencies due to their low surface area and poor regeneration capacities, hence restricts their applicability in large-scale wastewater treatment (Kainth et al., 2024). In this study, coconut husk (CH) has been chosen as an additional adsorbent to be combined with *M.oleifera*. Among all other agricultural wastes, CH has outperformed the performance of sugarcane bagasse, palm kernel shell, bamboo shell, and rice husk as an excellent adsorbent for treating wastewater (Boadu et al., 2019; Odisu et al., 2019). This is because CH has beneficial properties, such as a highly porous structure and strong absorptivity attributed to its high lignin and cellulose content, making it an exceptional as an additional bio-adsorbent for wastewater pollutants (James & Yadav, 2021)

1.2 Problem Statement

Although *M.oleifera* exhibits significant potential in wastewater treatment, nonetheless it possesses certain adjustments to enhance its coagulation efficiency such as coagulant dosage, combination with other coagulants and stirring speed. For instance, Ribeiro et al. (2019) asserted that the use of individual *M.oleifera* is unable to treat low turbidity effectively. This might be due to the low porosity of individual *M.oleifera*, which hinders it from removing the turbidity efficiently (Fereja et al., 2020; Gaayda et al., 2022). Thus, a study by Varsani et al. (2022), highlighted that removing low turbidity could be increased when treating the natural *M.oleifera* with hydrochloric acid. Numerous studies

have combined *M.oleifera* with other coagulants, such as aluminium sulphate and made modifications by using concentrated sulphuric acid, but these methods still yielded low efficiency in removing the turbidity (Freitas et al., 2016; Valverde et al., 2018). Plus, the activation of *M.oleifera* adsorbent using hydrochloric acid, sulphuric acid, sodium hydroxide, and potassium hydroxide is inappropriate due to its dependence on non-renewable resources and the production of hazardous waste and byproducts, which contradicts the fundamental principles of green chemistry that emphasise waste reduction, toxicity mitigation, and the utilisation of renewable materials (Asadu et al., 2021). Furthermore, it is necessary to determine the appropriate coagulant dosage, as an excessive amount might lead to elevated turbidity levels (Rasheed et al., 2023). In addition, the coagulation-flocculation process often necessitates an initial high stirring speed, then followed by a reduction to a low stirring speed for optimal coagulation (Novita et al., 2019). Nonetheless, prior research indicated that excessively high or low stirring speeds could reduce efficiency by causing particles to restabilise, resulting to weaker flocs and poor contaminants removal (Elemile et al., 2021).

Hence, the incorporation of CH as a biosorbent will enhance removal efficiency by targeting a broader range of contaminants, overcoming the limitations of *Moringa oleifera*, which alone is less effective in removing certain impurities. According to Varsani et al. (2022), *M.oleifera* cannot efficiently remove total dissolved solids (TDS), total suspended solids (TSS), electrical conductivity (EC) unless it is treated with concentrated acid and alkaline chemicals to enhance its removal capability. However, Nasaruddin & Mohamed (2021) reported that the greatest removal of TDS, TSS and EC can be achieved by using CH due to its high surface and excellent ion exchange capacity. Meanwhile, Sidik et al. (2022)

discovered that individual CH can achieve a maximum turbidity removal of up to 67% after 2 hours. The removal of turbidity can be maximised with the help of *M.oleifera* by effectively eliminating high turbidity levels and accelerating the reaction time (Gaikwad & Munavalli, 2019; Bouchareb et al., 2021). Since high removal of turbidity, TDS, TSS, and EC are crucial to increase water purification and are safe for human consumption, the combination of CH and *M.oleifera* will enhance each other's removal capabilities. Therefore, the combination of *M.oleifera* with CH will effectively replace concentrated acidic chemicals and alum sulphate, promoting stable floc formation during the coagulation-flocculation process at an optimal stirring speed and improving the removal of various contaminants from wastewater.

1.3 Significance of Study

This study maximises the effectiveness of *M.oleifera* by incorporating the substituting coconut husk (CH) for alum and other concentrated acidic chemicals. This work aims to enhance the effectiveness of *M.oleifera* by substituting CH for alum and other highly concentrated acidic compounds. This is because majority of studies have compared the effectiveness of *M.oleifera* and alum in wastewater treatment; however, there have been relatively few that have compared *M.oleifera* and other plant-based coagulants. Despite other seed proteins might be utilised similarly, a thorough analysis of over a hundred natural coagulants has clarified that the most preferred and effective natural coagulant for water treatment is *M.oleifera* plant (Kansal & Kumari, 2014; Muthuraman & Sasikala, 2014; Rosmawanie et al., 2018; Nouhi et al., 2019; Nisar et al., 2020; Okoya et al., 2020). Although *M.oleifera* has been proved to be a good adsorbent, the removal efficiency still has to be optimised. The performance of *M.oleifera* might be enhanced by adding other powerful

adsorbents, particularly CH. Instead of viewing CH materials as simply waste, converting them into natural adsorbent offers a twofold advantage. To date, no studies have been made regarding the implementation combination of *M.oleifera* and activated CH have been made. Therefore, this study suggests a novel method that utilises the possibilities for reusing and scaling up the biowastes into adsorbents. This method also provide high availability sources material and cost-effectiveness (Sidik et al., 2022). Moreover, high volume sewage sludge generated from the chlorination process can have adverse effects on the environment and human health (Xia et al., 2020; Zhao et al., 2021). Meanwhile, the effluent treated using *M.oleifera* is non-hazardous and has a smaller volume is produced compared to alum which makes it suitable for agricultural purposes and safe for environment (Hendrawati et al., 2016; Sulaiman et al., 2017; Nhut et al., 2021). The use of alum could lower the pH level, requiring the use of alkalinity to restore pH levels back to neutral, whereas *M.oleifera* does not alkalization for the effluent discharge (Andrade et al., 2021). This indicates that greater cost savings and faster. In this regard, the composite materials have addressed improved the turbidity, TDS, TSS, and EC removal and help speed up the settling time process compared to previous studies.

1.4 Research Objectives

The project carries the following objectives:

1. To analyse the characteristics of *M.oleifera*, coconut husk (CH) and different combined *M.oleifera* and coconut husk (MOCH) using scanning electron microscope (SEM), Fourier Transform Infrared Spectroscopy (FTIR), Brunauer-Emmett-Teller (BET) and zeta potential.

2. To evaluate the effect of different dosages of individual coagulant and different combined MOCH in wastewater samples by measuring removal of total dissolved solids (TDS), total suspended solids (TSS), electrical conductivity (EC), and turbidity based on different pH and temperature using respective instruments.
3. To elucidate the adsorption mechanism of the coagulants based on Pseudo-first order-kinetic model, Pseudo-second-order kinetic model, Langmuir model and Freundlich model.

1.5 Research Scope

The areas of research have been identified and are presented below.

Scope for objective 1: Analysing the characteristics of *M.oleifera*, coconut husk (CH) and combined *M.oleifera* and coconut husk (MOCH)

- a) Investigating the characteristics of *M.oleifera*, CH and combined MOCH 3:1 through the utilisation of Fourier Transform Infrared (FTIR) Spectroscopy, Scanning Electron Microscope (SEM), Brunauer-Emmett-Teller (BET) and zeta potential analysis.
- b) The study emphasises SEM, FTIR, BET, and zeta potential analyses, noting that characteristics like chemical stability, biodegradability, and long-term efficacy in wastewater treatment will not be evaluated at this stage, while the precision of these analyses is significantly dependent upon the uniformity of sample preparation.

Scope for objective 2: Comparing and assessing the impact of different dosages of individual coagulants and combined MOCH 3:1 for the removal of total dissolved solids (TDS), (TSS), turbidity and electrical conductivity (EC) from wastewater

- a) Examining the efficiency of individual dosage *M.oleifera* and CH in wastewater treatment by measuring TDS, TSS, turbidity and EC using respective instruments. The ideal dosage of coagulants may fluctuate depending on the composition of wastewater, and incorrect dosages might lead to inefficient removal of TDS, TSS, turbidity, and EC from wastewater.
- b) Selecting the optimal dosage from individual coagulants to create a combined MOCH ratio, such as MOCH 1:3, MOCH 3:1, MOCH 1:4, MOCH 4:1 and MOCH 4:5.
- c) Evaluating the turbidity removal performance using the optimised dosage, specifically MOCH 3:1 at different turbidity levels (50 NTU to 600 NTU) to determine its optimum performance as coagulation efficiency may vary depending on the turbidity levels.
- d) Optimizing the turbidity removal efficiency of MOCH 3:1 at different pH levels (2, 4, 6, 8 and 10) and temperatures (20 °C, 40 °C, 60 °C, 80 °C and 100 °C). Fluctuations in pH and temperature in natural wastewater can affect the efficacy of coagulants in eliminating TSS, turbidity, and EC.

Scope for objective 3: Investigating the adsorption process for the purpose of removing turbidity using MOCH 3:1

- a) Evaluating the adsorption performance of MOCH 3:1 in eliminating turbidity using Langmuir, Freundlich, Pseudo-first and Pseudo-second order kinetic models.

CHAPTER 2

LITERATURE REVIEW

2.1 Water crisis

Rapid increases in population growth, climate change, urbanisation, developing energy requirements, and changes in socioeconomic status have caused further water quality degradation (Mishra et al., 2021). Despite the existence of contemporary treatment methods, a significant number of individuals, particularly those in the least developed nations, still do not have access to clean water (Rahim et al., 2024). Likewise, Malaysia, although rich in water resources, is not exempt from the water crisis. Recent interruptions in water supply have not only threatened property values but also resulted in significant financial losses for businesses (Raihan et al., 2020). Water disruption is mostly caused by pollution, rapid population growth, and climate change, all of which increase the demand for water supply (Rashid et al., 2021). Recently, water crisis has emerged in several states of Malaysia notably in Selangor, affecting more than 8.32 million residents which has been caused by frequent shutdowns of the water treatment plant due to violations in the quality of the raw water (Loi et al., 2022). In Malaysia, the primary sources of clean water are surface water and groundwater; nonetheless, the Department of Environment (DOE) Malaysia (2024) reported that 94.13% of rivers are categorized as Class II and Class III, reflecting substantial pollution levels that considerably contribute to water stress in the nation. Furthermore, the access to clean water supply continues to be a significant barrier for indigenous groups in Malaysia still devoid access to clean piped water (Pimid et al., 2022; Goodson et al., 2023). As a result of this predicament, they have been compelled to depend on rainwater and river water for

their daily necessities. Consequently, individuals are susceptible to numerous waterborne diseases because they are dependent on untreated surface water or polluted wells (Balasooriya et al., 2023). Moreover, improper effluent disposal from industrial and domestic activities has placed unprecedented pressure on human health and aquatic ecosystems. The wastewater discharges mostly contain toxic heavy metals, high suspended solids and nutrient pollutant loads can bring adverse health effects such as cancer (Akpore et al., 2014), organ failure in humans and animals or even resulting in death due to exposure to these highly toxic substances (Uppal et al., 2019). This has resulted in an escalation in the pressure on freshwater sources, which further depletion of scarce water resources, with less than 10% of groundwater remaining in Malaysia (Segaran et al., 2022). The national economy could be significantly affected by the reduction of commercial fishing-related businesses and increased water purification costs due to the occurrence of algal bloom resulting from excessive nutrients present in the untreated wastewater effluent (Béchar, 2020; Liu et al., 2023). When eutrophication occurs, the depletion of dissolved oxygen concentration will occur, resulting in massive death of aquatic organisms and eventually leading to extinction over time (Haque et al., 2023). This condition led to a reduced supply of clean water due to the poor quality of the water (Nishat et al., 2023). As efficient water treatment is a fundamental prerequisite for humans and the environment, these challenges require constructive efforts by implementing low-cost yet trusted and excellent water treatment techniques to accelerate the pace of achieving water security as mentioned in sustainable development goals (SDG) by the year 2030. Water quality standards are guidelines established by water management authorities to ensure that the community's water supply is safe and clean based on the limits and criteria for permissible contaminants in water. As it is vital to ensure that future generations have access to potable water, the

quality of water-treated *M.oleifera* must be compared to DOE (2024) to verify that the water fulfils the necessary quality criteria, as indicated in Table 2.1.

Table 2.1: Characteristics of water quality standard

No.	Physicochemical parameters	Unit	Class I	Reference
1	Turbidity	NTU	5	.
2	Conductivity	μS/cm	1000	
3	pH	-	6.5-8.5	Department of Environment (DOE) Malaysia, 2024
4	TDS	mg/L	500	
5	TSS	mg/L	25	

2.2 Wastewater treatment technologies

Water crisis has become major concern due to improper disposal from various sectors such as industries, agriculture and households (Roshan & Kumar, 2020). Since access to safe and pure water resources is becoming limited in this era of realignment, thus there is a high demand for new and improved methods for water purification. Every water purification technique has benefits and limitations in the scope of environmental impacts, total operational costs, pre-treatment needed, and by-products from treated wastewater (Saleh et al., 2020). Several wastewater management techniques have been employed for water purification process such as biological treatment (slow sand filtration), chemical treatment (coagulation), and physical treatment (membrane separation) (Sravan et al., 2024). Slow sand filtration, primarily a biological process removes contaminants through combination of physical straining and microbiological activity (Bai et al., 2024). However, this method is less effective in treating high turbidity, heavy metals, colour removal and prone to clogging from excessive algae which reduced its filtration efficiency (Abdiyev et al., 2023). Other than that, electrocoagulation (EC) is a type of chemical treatment that employs an electric current to dissolve sacrificial electrodes, resulting in the generation of metal ions that interact with water pollutants, prompting their coagulation and subsequent removal (Mao et al., 2023). EC has several drawbacks, just like other treatment methods, such as high electricity consumption, electrode passivation and production of secondary contaminants in complex wastewater (Akter et al., 2022). In addition, the advanced oxidation process (AOPs) is one of the chemical treatments that utilizes hydroxyl radicals to strongly oxidize complex contaminants, thereby altering their nature and reducing their toxicity by destroying their structure (Kumari & Kumar, 2023). The widespread adoption of AOPs is hindered by their high operating costs, substantial energy consumption, and production of secondary by-

products that are frequently more toxic than the original pollutants, even though they are effective in eliminating persistent organic pollutants (Gopalakrishnan et al., 2023). Other method that used is ultrafiltration in which this physical treatment exhibits high efficiency in retaining microorganisms, but it experiences reduced membrane flux and shortened membrane lifespan as a result of membrane fouling induced by natural organic debris (Alomar et al., 2023). Nonetheless, these conventional methods encounter constraints caused by elevated expenses, inefficiency, and ancillary pollutants. Apart from that, traditional water purification techniques, including aluminium sulphate (alum), calcium hypochlorite, and chlorination, are commonly utilized in wastewater treatment but are progressively viewed as unfavorable due to huge operational and maintenance expenses, along with the generation of hazardous by-products such as trihalomethanes during the process, which are recognized for their carcinogenic characteristics (Popescu et al., 2017; Tak & Vellanki, 2018 ; Aboagye et al., 2021). In Malaysia, more than two million tonnes of aluminium sludge resulting from the coagulation-flocculation process are produced annually from water treatment plant and usually disposed to landfill which require more lands to dump the alum sludge. Moreover, the use of highly concentrated used of sulphuric acid is used to recover the alum sludge which not align with green chemistry principle (Basri et al., 2019). Other than that, previous method used adsorbents such as activated carbon, clays, zeolite, polyaniline and Zn-Al magnetite nano MgO to remediate water pollution (Ahmad et al., 2022). Nevertheless, the removal efficiency could be increased when high concentration of nitric acid is used and modifying the adsorbent with heavy metals as chelating agent. Since all methods above are expensive, require high energy and producing unwanted products, hence it is crucial to employ the use of low-cost adsorbents, environmental friendly and readily available (James & Yadav, 2021).

2.3 Application of biosorbents in wastewater treatment plant

Biosorbents are derived from biomaterials, mostly utilising accessible waste biomass from agricultural or other industrial sectors, either in their natural state or with enhanced pollutant removal capabilities (Madeła & Skuza, 2021). Recent advancements in adsorption technology have concentrated on employing agricultural waste as cost-effective, biodegradable biosorbents, offering an environmentally friendly option that aligns with the broader focus on minimizing waste generation and reusing unavoidable waste through a closed-loop system (Satyam & Patra, 2024). Moreover, the implementation of agricultural waste has gained popularity in recent years driven by the urgent necessity to alleviate environmental consequences (Rao & Rathod, 2019). Diverse agricultural wastes such as cassava peels, banana peels, pineapple peels, rice husks, cucumber peels, garlic and straw ash are recognised for their capacity to eliminate contaminants from water in an environmentally sustainable and economical way, particularly as substitutes for conventional methods such as chlorination and aluminium sulphate (Kumar et al., 2020; Sijabat et al., 2020; Zhang et al., 2021; Senanu et al., 2023). The properties of the biosorbent considerably influence the process of adsorption which involves the adhesion of adsorbates to the surface of the adsorbent that help to remove contaminants from wastewater (Tsoutsas et al., 2024). The main constituents of these biosorbents are cellulose, hemicelluloses, and lignin and might possess functional groups such as hydroxyl, aldehyde, carbonyl, carboxyl, phenolic, and ether groups, that selectively interact with wastewater contaminants (Karić et al., 2022). Since the biosorption capabilities of raw agricultural wastes are typically lower compared to synthetic commercial adsorbents, thus numerous adsorbents have been tailored as hybrid materials to enhance their selectivity for the removal of turbidity, total dissolved solids, total suspended solids, heavy metals, and other pollutants (Tang, 2024). Among other plant-based

coagulants, *M. oleifera* stands out as a highly effective biosorbent due to its high content of natural coagulant proteins, excellent antimicrobial agent, and abundance of functional groups to interact with wastewater pollutants via the mechanism of charge neutralization and adsorption (Nimesha et al., 2022). Nonetheless, the efficacy of *M. oleifera* alone may occasionally be limited, leading to poor reduction of turbidity, total dissolved solids, total suspended solids, and electrical conductivity, thereby requiring the integration with alum, or employing chemical modification with concentrated acid chemicals to boost its performance (Pandey & Khan, 2022; Varsani et al., 2022). While high dosages of *M.oleifera* can effectively remove high turbidity levels, they might not be as effective at reducing low turbidity levels (Amin et al., 2022). Conversely, dosage optimization is essential because smaller dosages are better suited for preventing microbiological growth after the coagulation-flocculation process (Shah et al., 2024). The performance of *M.oleifera* at lower dosages can be enhanced by substituting or combining it with another agricultural waste, particularly coconut husk (CH), which increase adsorption capacity and facilitates more efficient water treatment. This substitution is advantageous due to the inexpensive cost, widespread availability, and exceptional capacity of CH to effectively eliminate different contaminants from wastewater by providing additional sites, owing to its lignocellulosic composition (Varsani et al., 2022). In fact, the abundance of coconut farms in Malaysia contributes to a substantial quantity of coconut waste, with over 80,000 tones discarded annually, making it the leading contributor to the entire volume of agricultural waste (Rosli & Jumali, 2022). Hence, studies should be conducted to evaluate the efficacy of these combinations of adsorbents in enhancing water quality.

2.3.1 Characteristics of *Moringa oleifera*

As pressure is rising on nations to incorporate more innovative and sustainable water treatment technology for a green future, the practice of using natural plant materials as coagulants has garnered significant interest (Manasa & Mehta, 2020; Saeed et al., 2022). Malaysia has been recognised as having one of the richest biodiversity in the world, and the *M.oleifera* tree grows well in this country, thus making it easy to find and nurture (Demies et al., 2019). *M.oleifera* is as a tropical multipurpose tree that may grow up to 12 meters tall and is well-adapted to semi-arid regions (Ali & Seng, 2018). This tree belongs to the native plant of India, which is well-known for its high nutritional value and is widely used as a traditional medicine in most countries (Singh et al., 2020). This readily available natural product offers a cheap method since this crop has been spread worldwide in northern Europe, Asia, Africa, and Madagascar (Trigo et al., 2020). The leaves of *M.oleifera* have significant potential in treating various diseases such as cancer, diabetes, malaria, fever and HIV/AIDS (Ortega & Campos, 2018). On the other hand, *M.oleifera* seeds exhibited excellent antimicrobial activity against gram-positive and gram-negative bacteria (Jagaba et al., 2021; Mehwish et al., 2021). *M.oleifera* seed provides an environmentally friendly green approach to phycoremediation of wastewater by providing cost-effective technology and excellent microalgal biomass harvesting efficiency compared to alum, which brings major human health risk implications (Hasan et al., 2021). Figure 2.1 summarises the benefits use of *M.oleifera* in many applications. The plantation of *M.oleifera* trees could benefit to the country as it improves the economic sector by generating more employment for the local population and promoting carbon capture near the city.

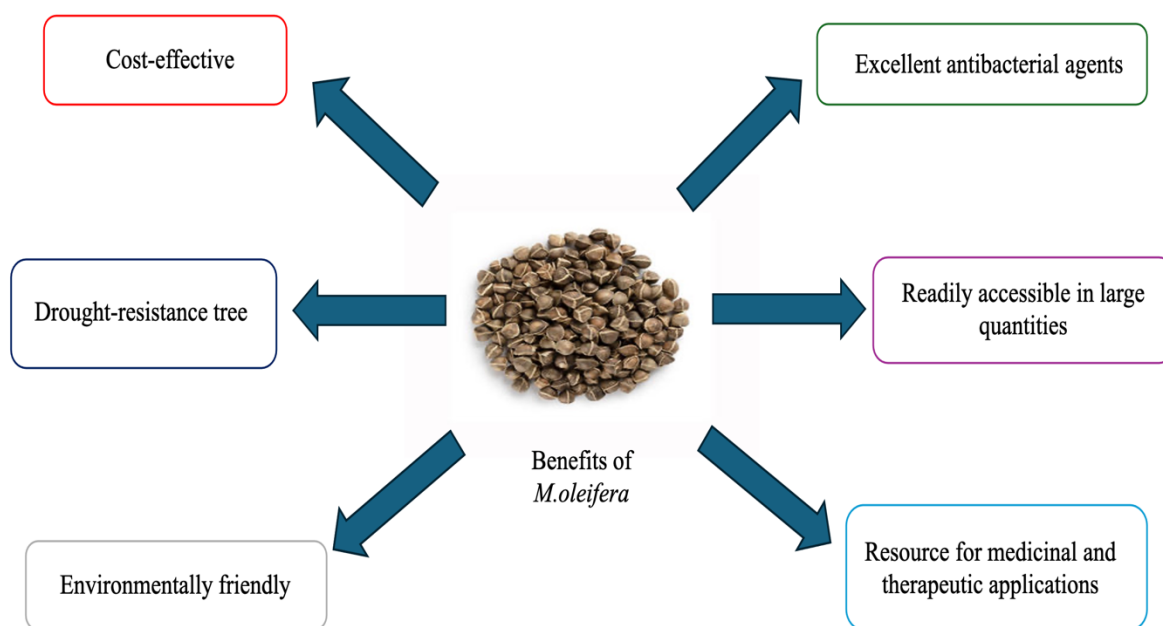


Figure 2.1: Summary of advantages using *M.oleifera* in various fields. Illustration adapted with permission from Sulaiman et al. (2017)

The seeds of the plant have unique coagulant properties that allow them to be used in water sanitation and contain natural ingredients that are harmless to humans. The seeds of *M.oleifera* have been discovered to possess bioactive compounds, including 4-(α -L-rhamnosyloxy) benzyl isothiocyanate, pterygospermin, and morigyne, as shown in Figure 2.2 (Idris et al., 2016; Choudhary & Neogi, 2017; Delelegn et al., 2018; Williams, 2019). One of the compounds in the *M.oleifera* identified as having a notable role in combating bacterial infections is 4-(α -L-rhamnosyloxy) benzyl isothiocyanate. The removal of harmful bacteria from water is crucial because they are usually correlated with pulmonary diseases, skin diseases, gastroenteritis and urinary tract infections (Romeo et al., 2018; Taylor & Unakal, 2019). Furthermore, *M.oleifera* seeds are a rich source of additional bioactive compounds, including 4-hydroxyphenyl acetonitrile, 4-hydroxyphenyl-acetamide, 4-(α -l-rhamnopyranosyloxy)-benzyl glucosinolate, Roridin E, Veridiflorol, 9-octadecanoic, 2-O-

(6-o oleoyl-beta-D-glucopyranosyl)-beta-sitosterol, and beta-sitosterol-3-O-beta-D-glucopyranoside (Anzano et al., 2021). The seeds' potential as natural remedies is enhanced by the presence of 4-(-l-rhamnopyranosyloxy)-benzylglucosinolate and Roridin E, which have anti-inflammatory and immunomodulatory properties (Rani et al., 2018). In addition, the presence of Veridiflorol, 9-octadecanoic, 2-O-(6-o oleoyl-beta-D-glucopyranosyl)-beta-sitosterol, and beta-sitosterol-3-O-beta-D in *M.oleifera* seeds has been associated with numerous health benefits, such as cholesterol-lowering properties and potential antioxidant effects (Bhattacharya et al., 2018). These diverse bioactive compounds in *M.oleifera* seeds demonstrate their value as a natural resource for medicinal and therapeutic applications, paving the way for developing new pharmaceutical agents and dietary supplements. Besides that, the functional groups in the bioactive compounds of *M.oleifera* responsible for the coagulation-flocculation process include hydroxyl, carboxyl, and carbonyl groups, which engage with contaminants via mechanisms such as electrostatic attraction, hydrogen bonding, complexation, and Van der Waals forces (Benettayeb et al., 2022). These characteristics not only demonstrate *M.oleifera's* promise as an economical and environmentally benign wastewater treatment method but also enhance human health. Overall, employing *M.oleifera* as a natural coagulant is straightforward and provides greater advantages than synthetic coagulants, owing to its natural properties and effectiveness against waterborne bacterial diseases (Hassan et al., 2021).

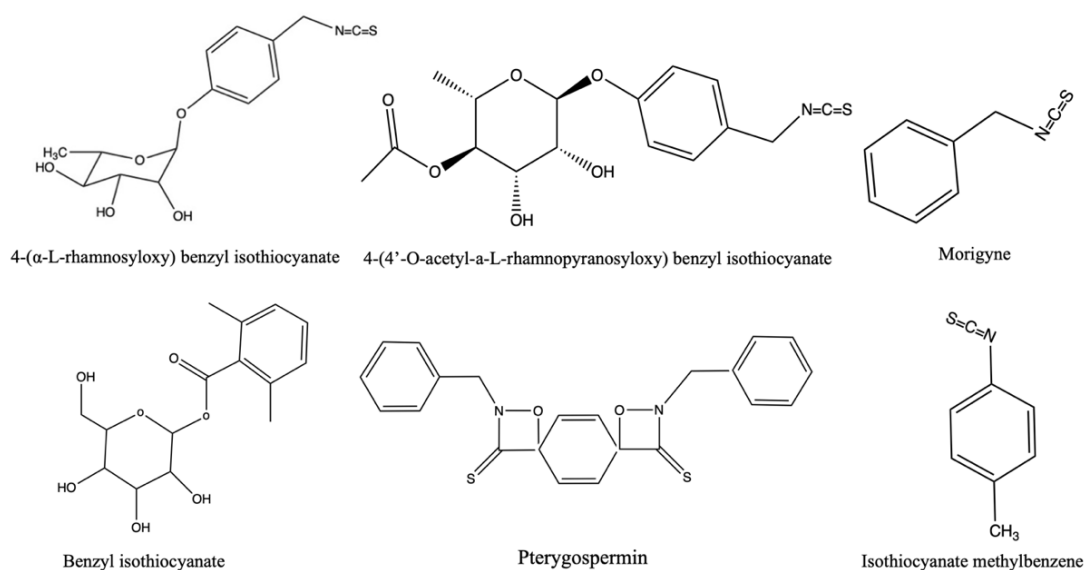


Figure 2.2: Chemical constituents of *M.oleifera* seed Illustration adapted with permission from Idris et al. (2016)

2.3.2 Comparison of *M.oleifera* with other coagulants

Plant-based coagulants have the potential to be a sustainable and cost-effective alternative to alum for water purification. Few studies have improved the efficacy *M.oleifera* as a coagulant by combining it with other plant-based coagulants, utilising synergistic effects to enhance its coagulation performance in water treatment. For instance, *M.oleifera* seeds could give the highest removal of Pb; nevertheless, the highest removal efficiency of heavy metals such as Ni and Cd can only be achieved by combining *M.oleifera* and banana peels (Aziz et al., 2016). A study conducted by Gandiwa et al. (2020) proved that the result of water treatment from the combination of plant-based coagulants, which are *M.oleifera*, *Cactus Opuntia* together with alum as synthetic coagulant falls within the required limits for potable water as per World Health Organisation (WHO) guidelines in terms of turbidity, total alkalinity, pH and conductivity. Conversely, another study reported that *M.oleifera* alone has the highest turbidity removal rate of more than 90%, followed by okra seed, soyabean and papaya seed which meets the water treatment criteria established in National Water Quality Standard Malaysia (NWQSM) (Ng & Elshikh, 2021). Dosage significantly

influences the removal of turbidity and chemical oxygen demand (COD), with fly ash and banana peels demonstrating optimal removal at 800 mg/L, while *M.oleifera* required a higher dosage for effective removal, but its efficiency declined beyond 800 mg. (Chitra & Muruganandam, 2020).

Next, *M.oleifera* owes more advantages than alum because it is relatively cheap, locally available and biodegradable, making it safe for water purification. Table 2.2 provides explicit information regarding the efficacy of *M.oleifera* and chemical coagulants in water treatments. In addition, *M.oleifera* can effectively remove microalgae from water, demonstrating high efficiency in reducing turbidity by 93% to 95.44%, COD by up to 82.4%, and *E.coli* up to 99.1%, due to its water-soluble cationic proteins, which interact with wastewater pollutants through charge destabilisation (Hoa & Hue, 2018; Silva et al., 2021). Although *M.oleifera* showed the highest removal of heavy metals for Ni and Cd at 98% and 63%, respectively by using the optimum 5000 mg/L doses of *M.oleifera* compared to alum and ethylenediaminetetraacetic acid (EDTA), FeCl_3 demonstrated foremost in reduction of heavy metals as only 1000 mg/L dose was required to work effectively (Taiwo et al., 2019). It is suggested that *M.oleifera* seeds could be used as a pre-treatment to $\text{Al}_2(\text{SO}_4)_3$, owing to its advantages of generating better effluent final quality without requiring further pH adjustment, unlike the use of aluminium sulphate alone, and this can help to generate less waste with aluminium concentration, thus provide sustainable environment and more economical (Rocha et al., 2019; Nisar & Koul, 2021). The treatment using *M.oleifera* is more effective in terms of turbidity and higher Fe removal than alum, while on the other side, alum has higher Mn removal than *M.oleifera* (Mohammed et al., 2021). Moreover, *M.oleifera* was 100% efficient at removing THMs which are the by-products from chlorination process, that known to have mutagenic and carcinogenic properties (Okoya et al., 2020). In essence,

M.oleifera is a reliable natural coagulant that can replace chemical coagulants in water purification since they are a non-hazardous, good antibacterial agent and generate biodegradable sludge.

Table 2.2: Comparative studies between *M.oleifera* and conventional coagulant

Coagulants	Water sample	Outcomes	pH	Dosage of <i>M.oleifera</i> (g/L)	References
Combination of alum and <i>M.oleifera</i>	River and sewage water samples	▪ <i>M.oleifera</i> outperformed alum in terms of improving the physicochemical qualities of water samples by greatly reducing TDS, EC and turbidity as well as neutralise the water samples. ▪ At the end of the treatment, the pH of water samples becomes more acidic because of sulphuric acid created by alum	7	12	(Thorat et al., 2020)
	River Nile water	▪ <i>M.oleifera</i> seed exhibits better performance as a coagulant than alum in the removal of Fe, turbidity, chlorides, bacteria, and total algae count. ▪ The highest Mn removal can be achieved by alum compared to <i>M.oleifera</i> seed. ▪ However, both coagulants showed an increase in total hardness, EC, magnesium hardness and TDS. ▪ <i>M.oleifera</i> contributes the most to the rising of sulphate, magnesium, potassium, ammonia, and fluoride which contrast to the amounts in the original samples collected.	8	7	(Mohammed et al., 2021)
	Natural water sources: Santa Helena Brook (SHB) & Picacha Brook (PB)	▪ Combination of <i>M.oleifera</i> and alum demonstrated high turbidity reduction rates of more than 90% which comply with the limits of acceptable turbidity in drinking water quality ▪ Only <i>M.oleifera</i> reached the colour removal standard ▪ A higher dosage of <i>M.oleifera</i> did not cause a significant decrease in both turbidity and colour removal ▪ When the optimal dosage of alum is increased, the colour removal will greatly be reduced ▪ Alum showed greater efficiencies at low dosages than <i>M.oleifera</i>	7	1-4	(Gámez et al., 2015)

Table 2.2 continued

Combination of alum and <i>M.oleifera</i>	Distillery spent wash effluent	▪ The highest removal of turbidity, colour removal and COD reduction above 90% can be achieved upon the addition of <i>M.oleifera</i> into aluminium sulphate and ferric sulphate each under optimum conditions. ▪ Increase the <i>M.oleifera</i> dosage did not give significant colour removal. ▪ The least effective among all coagulants with the help of <i>M.oleifera</i> is calcium sulphate while aluminium sulphate demonstrated the most effective as a coagulant in treating distillery spent wash effluent.	4	40	(David et al., 2015)
	Municipal wastewater	▪ At optimal temperature between 25-30 °C, the maximum removal of turbidity was over 90% when <i>M.oleifera</i> is used as an additive with alum for treating wastewater. ▪ <i>M.oleifera</i> works efficiently, especially for higher turbid wastewater that has more than 100 NTU ▪ The maximum reduction of COD can be up to 75%. ▪ Both alum and <i>M.oleifera</i> exhibit the same pattern in which increasing the coagulant concentration could reduce COD reduction	9	300	(Kane et al., 2016)

2.3.3 Coagulation-flocculation mechanism

The coagulation-flocculation method is a water treatment technique that enhances filtration by utilising specific mechanisms to aggregate and eliminate suspended particles, plankton and bacteria from water (Al-Sahari et al., 2020). Figure 2.3 displays the coagulation-mechanism process that aid to remove the contaminants from the wastewater. There are four types of coagulation and flocculation mechanisms that depend on the coagulant properties (Amran et al., 2018). The bridging mechanism involves the coagulant generating long-chain polymers that adsorb numerous particles on their surface, resulting in larger aggregates that settle more efficiently (Sulaiman et al., 2017). This technique is very efficacious for low-turbidity water. In the bridging mechanism, *M.oleifera* seeds can have the bridging ability with chitosan molecules that consist of -NH₂ and -OH functional groups, can enhance the adsorption on the surface of turbid colloidal particles and form large particles to reduce turbidity, improve the odour and taste of drinking water (Vigneshwaran et al., 2020). Sweep coagulation, often employed in conventional methods, involves the addition of high concentrations of metal coagulants like aluminum or iron salts, which form precipitates that encapsulate and entrap colloidal particles, making it highly effective for treating water with high turbidity (Nath et al., 2020). However, the continuous use of an elevated amount of aluminium sulphate as a water coagulant has caused brain changes that meet such characteristics of Alzheimer's disease blocking the bone formation, which leads to bone fractures called *osteomalacia* and also causes a lethal effect on aquatic organisms as well because aluminium can act as a toxic agent by damaging the gill-breathing mechanism in fish (Poléo et al., 2017; Corkins, 2019; Krupińska, 2020). Apart from that, charge neutralisation is a process that balances the charges of colloidal particles between cationic coagulants and the negative charges of pollutants, which leads to the formation of flocs that

can be easily removed via filtration, commonly utilised for low to moderate turbidity levels (Ali et al., 2024). *M.oleifera* is a natural cationic polyelectrolyte that contains up to six amino acids, primarily glutamic acid, methionine, and arginine (Thorat et al., 2020). Charge neutralisation occurs at optimal pH 6-8 when *M.oleifera* seeds that have cationic antimicrobial protein will be attracted to the negatively charged humic acid of wastewater by Van Der Waals forces and form flocs to achieve the reduction in turbidity, and any suspended solids present (Nisha et al., 2017; Tie et al., 2017; Ahmad et al., 2019; Ng & Elshikh, 2021; Sativa et al., 2021). Furthermore, patch flocculation occurs when coagulants create positively charged patches on particle surfaces that attract negatively charged particles to form aggregates (Yamaguchi et al., 2021). During the patch flocculation process, the removal of heavy metals can be achieved due to the presence of carboxylic acid and hydroxyl group on the surface of *M.oleifera* seeds that form an electrostatic force of attraction with heavy metal ions (Maćczak et al., 2020; Saleh et al., 2020; Nisar & Koul, 2021). The predominant coagulation mechanism for plant-based flocculants is charge neutralisation, however, bridging is also a common process that results in the creation of complexes between particle-particle and flocculant or both mechanisms can occur simultaneously (Mansour et al., 2024). In summary, coagulation-flocculation processes are important in removing contaminants from wastewater, with various mechanisms contributing to the efficient aggregation and sedimentation of particles.

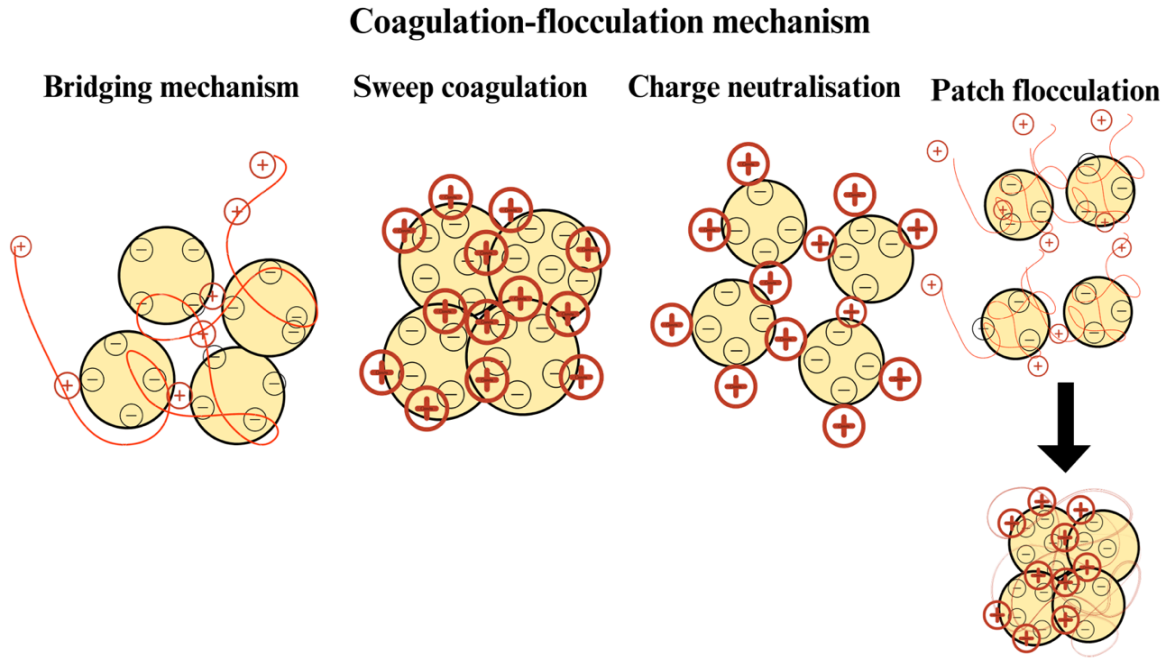


Figure 2.3: Coagulation-flocculation mechanism in water purification process. Illustration adapted with permission from Al-Sahari et al. (2020)

2.3.4 Coagulation activity

The performance of *M.oleifera* across a wide range of wastewater has shown promising results in removing various contaminants owing to its natural coagulant characteristics. Table 2.3 provides a comprehensive comparison of the reviewed studies on the effects of stirring speed, coagulant dosage, and pH on the performance of *M.oleifera* as a coagulant for turbidity removal. These factors are linked and optimising them is vital for achieving *M.oleifera*'s optimal coagulation activity in water treatment applications. The stirring speed is essential for increasing the aggregation of suspended particles and facilitating the formation of large and stable flocs, thereby strengthening the overall coagulation efficiency of *M. oleifera*. The stirring rate is primarily determined by the agglomeration of suspension to flocs, which then accumulate and settle through gravity. Fast mixing helps the coagulant to destabilize the negatively charged particles, while slow

agitation is necessary to cause the particles to aggregate into larger flocs and settle (Chitra & Muruganandam, 2020). Numerous prior studies have investigated that higher revolutions per min (rpm) (>100) was more effective than lower stirring speed, which has resulted in the most considerable reduction of turbidity, TDS, TSS and EC (Gámez et al., 2015; Novita et al., 2019; Zaid & Ghazali, 2019; Gaayda et al., 2022). According to these research, insufficient dosage and stirring speed result in incomplete charge neutralisation and decreased flocculation efficiency, whereas high dosage and speed can cause particles to restabilise, leading to weaker flocs and poor turbidity reduction. Conversely, other research implies that chemically modified *M.oleifera* with acid-treated carbonized chitosan operates effectively at lower stirring speeds (rpm) by enhancing interactions with colloidal particles through synergistic adsorption, facilitated by additional functional groups in chitosan, such as amine and hydroxyl groups, which promote stable coagulation-flocculation process (Vigneshwaran et al., 2020). Other than that, dosage optimization is very crucial because excessive coagulant dosage of *M.oleifera* could increase the level of turbidity, biological oxygen demand (BOD) and chemical oxygen demand (COD) due to the formation of unstable large flocs, which are prone to redispersion (Dehghani & Alizadeh, 2016; Camacho et al., 2017; Shan et al., 2017; Elemile et al., 2021). Since the *M.oleifera* coagulant dosage itself is organic compounds, the addition of this coagulant could contribute to an increase in the level of organic compounds in the water, which can subsequently give rise to the COD and BOD levels because the excess of organic matter may require extra oxygen for decomposition (Nonfodji et al., 2020; Nhut et al., 2021; Varsani et al., 2022). Apart from that, water's turbidity increases when the *M.oleifera* concentration surpasses the appropriate dosage since all colloids have been neutralised and settled down (Rasheed et al., 2023). Table 2.3 further explains the relationship between pH levels and the coagulation process using

M.oleifera, demonstrating that, while this coagulant can neutralise pH, an alkaline environment is more conducive to coagulation due to enhanced electrostatic interactions between the positively charged coagulant and negatively charged contaminants, which improves floc formation and removal efficiency. Additional studies reported that heavy metals and turbidity removal by using *M.oleifera* depend on the pH of wastewater samples. For instance, the proteins in *M.oleifera* seeds demonstrate their utmost capacity for adsorption within a pH range from 5 to 8.67, as evidenced by the research conducted by Al-Jadabi et al. (2023). Nonetheless, conflicting data from a study by Desta and Bote (2021) suggest that *M.oleifera* attains its optimal efficacy in a pH range of 7 to 9. Furthermore, excessive coagulant particles will occur due to their competitive binding with heavy metal ions, while an excessively low initial pH may result in the redissolution of metal ions (Hegazy et al., 2021). This phenomenon is attributed to the distinct solubility and precipitation characteristics exhibited by different heavy metals at different pH levels, thereby decreasing the efficacy of heavy metal removal from water samples. This is because the coagulant's ability to destabilise and aggregate colloidal particles is diminished when the pH is too high, resulting in less effective turbidity reduction (Hendrawati et al., 2016; Sun et al., 2019; Varsani et al., 2022). It is feasible to improve the coagulation and subsequent removal of turbidity and heavy metals from effluent by altering the pH within a specific range. Overall, stirring speed, proper dosage optimization, and suitable pH are required to minimise the undesirable effects of excessive coagulant dosage on impurities removals from the wastewater samples.

Table 2.3: Influence of dosage, stirring speed, and pH on coagulation performance

Coagulants	Coagulant dosage (g)	Stirring speed	pH	Settling time (mins)	Turbidity removal (%)	References
Acid treated carbonized chitosan with <i>M.oleifera</i>	0.2	50 rpm for 100 mins	5-8	10	84	(Vigneshwaran et al., 2020)
<i>M.oleifera</i> powder	0.05	1) 200 rpm for 4 mins 2) 40 rpm for 30 mins	7	60	98.80	(Zaid & Ghazali, 2019)
	1-4	1) 100 rpm for 1 min 2) 40 rpm for 15 mins	7	15	90	(Gámez et al., 2015)
	0.00034	135 rpm for 13.52 mins	6.93	113.15	98.5	(Gaayda et al., 2022)
	0.85	1) 400 rpm for 1 min 2) 150 rpm for 15 mins	9	60	88.15	(Novita et al., 2019)

2.3.5 Adsorptive potential of coconut husk as additional adsorbent

Agricultural wastes from agro-industrial activities have enormous potential as adsorbents in wastewater treatment. This is because the use of agricultural wastes as filter media adheres to green chemistry principles and promotes the circular economy through closed-loop recycling (Omran & Baek, 2022). Agricultural wastes have properties that render them acceptable for use because they are rich in natural substances like cellulose, lignin, tannins, proteins, and other organic elements (Wang et al., 2020; Elshabrawy et al., 2023). These components have adsorptive, ion exchange, and filtration capabilities, allowing them to effectively absorb and eliminate different pollutants from wastewater, such as heavy metals, dyes and pesticides (Dawn & Vishwakarma, 2021; Zhu et al., 2022; Jagadeesh & Sundaram, 2023). Table 2.4 represents the comparison of different agricultural wastes to remove contaminants from various water samples, including their limitations to be used for water treatment. The table summarises that coconut husk (CH) outperforms the rice husk, banana peels, sago hampas, palm kernel, and sugarcane bagasse in the removal of heavy metals, total solids, and bacteria from wastewater. This is because CH exhibits excellent adsorption capacity for heavy metals and total solids, owing to its abundance of adsorption sites and functional groups such as hydroxyl, carbonyl, and ether groups, which are derived from its high content of lignin, cellulose, and hemicellulose (Boadu et al., 2019; Odisu et al., 2019). In addition, it also contains a natural substance called activated carbon, which has excellent adsorption properties by attracting and trapping impurities such as heavy metals, pesticides and organic compounds in its porous structure (Zhang et al., 2018). CH has been proven to be the most effective for wastewater removal as it promotes an aerobic environment, which may stimulate the growth of beneficial microbes that assist in the removal of various water contaminants (James & Yadav, 2021; Mnyoro et al., 2022). CH is

resistant to biodegradation, allowing it to preserve its structural integrity during filtration (Shrestha et al., 2019; Ahmad et al., 2022). Overall, CH has the potential to be combined with *M.oleifera* due to its excellent adsorbent properties, which could enhance the efficacy of impurity removal from effluent significantly.

Table 2.4: Agricultural wastes as adsorbent for water and wastewater treatment

No.	Agricultural wastes	Water sample	Removal efficiency	Limitations	References
1	Coconut husk	Wastewater	It able to remove zinc oxide nanoparticles from wastewater.	The CH filters must be maintained and replaced on regular basis to ensure optimal performance.	(Cruz et al., 2019; Droepenu et al., 2021)
		Drinking water	The CH has a remarkable potential to reduce methyl tertiary-butyl ether (MTBE) to levels that are undetectable as well as improved the taste and odour of drinking water.		(Samdin et al., 2013)
		River water, pond water, well water, bore hole water and seawater	The CH as biofilter managed to remove total solids (TS) from all water samples better than loofah.	However, this biofilter unable to achieve optimum sorption efficiency of TS above 50% especially for water samples from river water and seawater	(Adelodun, 2021)
		Cement wastewater	Activated carbon from CH has high BET surface area and good pore diameter which allows it to adsorb heavy metals better than palm kernel husk and groundnut shell.	CH has limited metal adsorption due to low molecular weight and short chain of cationic polyelectrolyte.	(Boadu et al., 2019; Odisu et al., 2019; Saleh et al., 2020)

Table 2.4 continued

2	Activated carbon from CH and rice husk	Wastewater	The combination of activated carbon from CH and rice husk was approximately 100% effective in eliminating <i>E.coli</i> and phosphorus from wastewater. The removal rate for nitrate-nitrogen and ammonia-nitrogen were above 79%.	Activated carbon from CH provides the greatest removal rate of ammonia-nitrogen and may attain desired pH values in wastewater faster than rice husk.	(Kasmuri et al., 2022)
3	Rice husk	Stormwater runoff	Carbonized rice husk tends to remove turbidity and <i>E.coli</i> bacteria almost 90% due to high content of silica and large pores.	Unmodified rice husk exhibited low adsorption capability due to low surface area and functional groups. It has limited adsorption capacity of heavy metals especially nickel. The removal of heavy metals dependent to the initial pH value	(Shamsollahi & Partovinia, 2019; Annisa et al., 2021)
		Aquatic environment	It can eliminate polycyclic aromatic hydrocarbons from the aquatic environment.		(Rinawati et al., 2019)
4	Banana peels	Rainwater and river water	The treated water can be used as cleaning water as it able to reach the neutral pH.	The treated water cannot be used as drinking water because all samples do not comply with bacteriological requirements.	(Tjahjanti et al., 2021)

Table 2.4 continued

5	Sago hampas	Watercourses	The sago hampas are good low-cost adsorbents to eliminate harmful metal ion pollutants such as Cu (II), Pb (II), Hg (II) and Zn.	The optimum pH required for higher Pb (II) and Cu (II) efficiency removal are at lower pH while higher Hg(II) and Zn adsorption are at higher pH>7.	(Amin et al., 2019)
6	Palm kernel shell	Synthetic wastewater	The activated carbon is highly effective at eliminating hydrogen sulfide under alkaline medium as well as heavy metals from synthetic wastewater.	The removal of heavy metals is dependent to certain pH levels, agitation speed and amount of the dosage. The percentage removal of Cd ²⁺ and Zn ²⁺ ions from water lower than 85%.	(Baby et al., 2019; Alenezi & Al-Qabandi, 2023)
7	Sugarcane bagasse	Wastewater	The sugarcane bagasse demonstrated better adsorption of reactive blue dye from wastewater due to the presence of hydroxyl groups and carbonyl group under alkaline condition at 40°C temperature.	This method is only applicable to countries that produce sugarcane bagasse. By increasing both the contact period and the initial dye concentration, the removal efficiency can be diminished.	(Piffer et al., 2020; Elshabrawy et al., 2023)

2.3.6 Properties of coconut husk

CH is a prevalent agricultural by-products in tropical regions that has attracted significant interest as a cost-effective and naturally abundant adsorbent for wastewater treatment (Rahim et al., 2021). Although adsorption process using charcoal, clay mineral, zeolite and aerogels have been proven for treating pollutants, however it is more imperative to use CH as low-cost adsorbent due to its high absorptivity, porous structure, abundant availability, and minimise environmental wastes (Kennedy et al., 2018; James & Yadav, 2021). The adsorptive capacity of CH is attributed to its lignocellulosic composition, which consists predominantly of cellulose, hemicellulose, and lignin as shown in Figure 2.4 (Verma et al., 2021). These components are rich in functional groups, such as hydroxyl (-OH), carboxyl (-COOH), and methoxyl (-OCH₃), that play a key role in interacting with organic and inorganic pollutants (Hanafiah et al., 2020). Possible mechanism interactions such as charge neutralisation, electrostatic attraction, ion-exchange and complexation processes might involve between the pollutants and CH (Hussain et al., 2024). The combination of its functional groups and porous structure makes CH a versatile material for addressing various water contamination issues. Conversely, raw CH exhibits lower adsorption capacity than chemically treated adsorbents but its efficacy can be improved through treatments such as acid and alkali activation and heat treatment to increase porosity and create additional active sites to enhance pollutant removal capacity (Wang et al., 2023). Nonetheless, these methods are unfavourable due to its high chemical costs, potential environmental hazards from waste disposal and possible loss of functional groups that are essential for pollutant binding (Ahmed et al., 2021). The coagulation efficiency of CH could be improved through additional modifications with other plant-based coagulants, as it primarily relies on the physisorption process via Van Der Waals forces to remove contaminants from wastewater,

which can result in the easy desorption of pollutants (Kasmuri et al., 2018). Hence, the combined effect of *M.oleifera* and CH could provide a synergistic impact, augmenting the efficacy of both substances in wastewater treatment. This hybrid method enables each material to enhance the effectiveness of the other, consequently increasing their combined capacity to adsorb and coagulate contaminants more efficiently than when utilised separately.

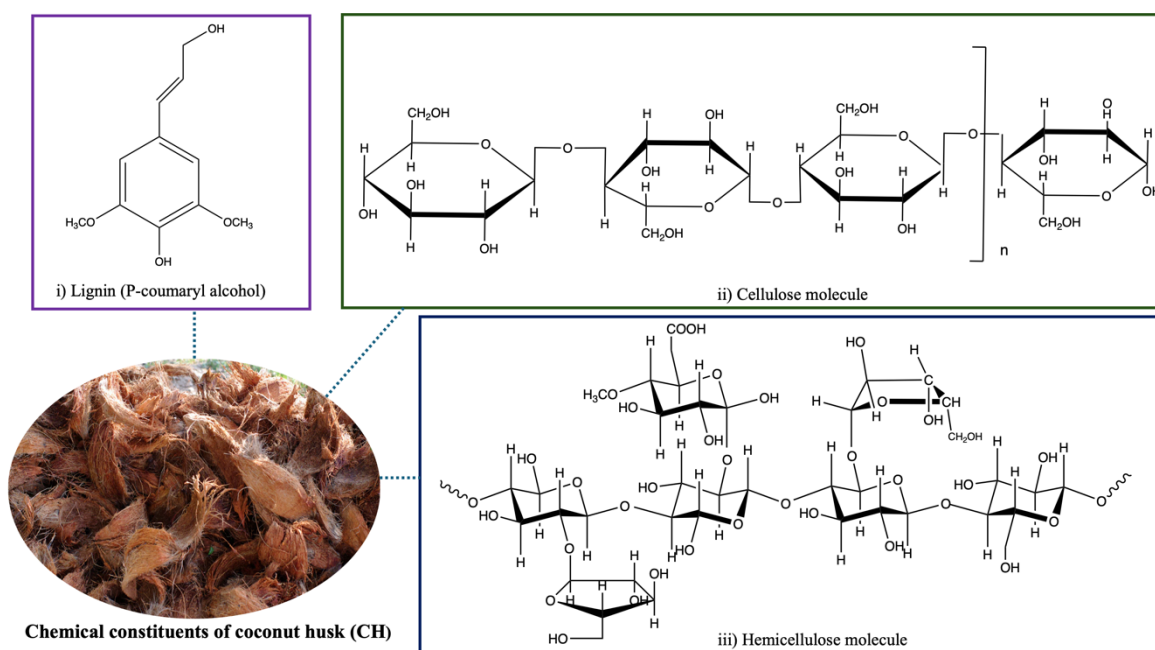


Figure 2.4: Main chemical constituents of coconut husk (CH). Illustration adapted with permission from Carre et al. (2019)

2.4 Characterization of biosorbents

Biosorption is a viable alternative that use economical natural or modified biosorbents derived from biological sources, such as plant biomass, fungi, algae, or agricultural by-products to remove harmful contaminants from wastewater (Karnwal, 2024). The efficacy of a biosorbent in pollutant elimination is dependent upon its surface characteristics, chemical composition, and the accessibility of sorption sites, significantly affected by its high polysaccharide content (cellulose, hemicellulose, lignin, pectin, and

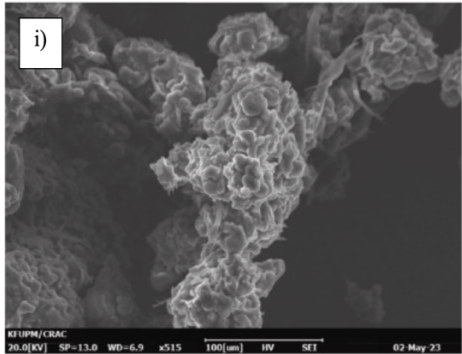
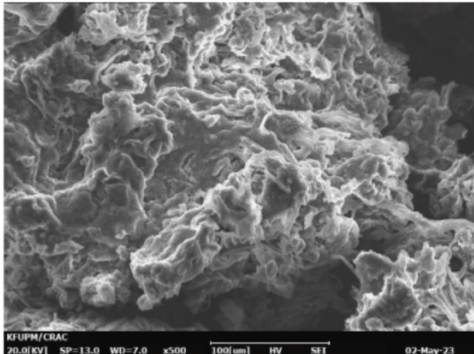
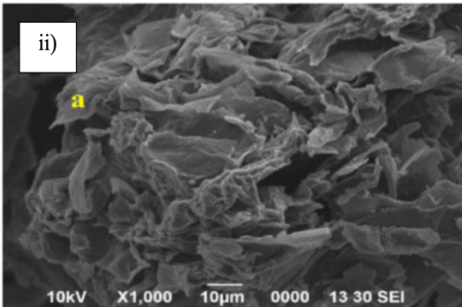
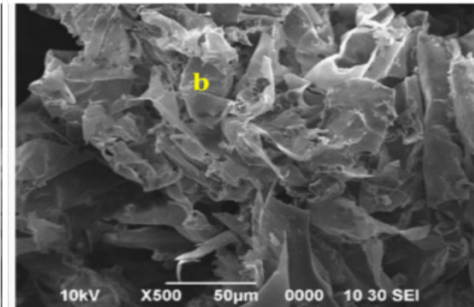
chitin), which consists of numerous functional groups (carboxyl, hydroxyl, and carbonyl groups) that are capable of adsorbing or binding contaminants from aqueous solutions via various mechanisms, including adsorption, electrostatic interaction, complexation, coordination, chelation, and ion exchange (Madeła & Skuza, 2021). Various characterization techniques are used to assess the effectiveness of biosorbents in adsorption applications, including Fourier Transform Infrared Spectroscopy (FTIR), Scanning Electron Microscopy (SEM), Brunauer-Emmett-Teller (BET), and zeta potential analysis. These techniques are used to understand the surface morphology, porosity, chemical composition, and surface charge of *M.oleifera* and CH, which are critical factors in influencing their adsorption capacity. Meanwhile, the adsorption process is evaluated based on the Langmuir and Freundlich model as well as pseudo-first-order kinetic model and pseudo-second-order kinetic model.

2.4.1 Scanning electron microscopy (SEM) analysis

The surface morphology of natural materials such as *M.oleifera* and CH have been a subject of interest in various studies. Most studies reported that the surface morphology of *M.oleifera* in natural state is heterogenous surface, rough and relatively porous which facilitate the sorption process for the removal of impurities from wastewater (Shah et al., 2024; Taoualit et al., 2024). Meanwhile, Saleh et al. (2020) reported that *M.oleifera* appears as spherical and has villous structure that help to in the adsorption and coagulation process. A study reported by Okoya et al. (2020) revealed there is alteration on the surface morphology of *M.oleifera* after the adsorption of trichloromethane and tribomethane. A similar conclusion was also made by Alghamdi et al. (2024), who reported that an alteration occurred on the surface texture of *M.oleifera* from a porous structure to a fibrillar shape that confirmed the adsorption of heavy metal on the adsorbent as shown in Table 2.5 (i).

Other than that, activated CH has a rough surface and high surface area which provide more sites for the adsorption process to take place (Mustapha et al., 2021). Table 2.5 (ii) indicates the rough surface of CH before adsorption, suggesting a significant likelihood of active sites for adsorbate binding, while after adsorption, the incorporation of ZnO-NPs onto RCH modifies the surface morphology of the adsorbent (James & Yadav, 2021). According to Anuar et al. (2018), the structure of CH is rod-like and has sharp edges. However, the study emphasized that the morphological characteristics of CH underwent modifications, with the particles becoming smaller and more irregular after being treated with acid and taking on an amorphous state after being treated with alkali. Halfhide et al. (2019) also supported the statement that morphological structure of CH underwent physical alterations after treating with acid chemicals such as HNO_3 , HCl and H_2SO_4 . Comprehensively, the morphology features of the biosorbents examined by using SEM could provide details regarding physical changes when treating the adsorbents with chemicals. Overall, a coagulant's ability to trap impurities is directly influenced by its morphological characteristics, such as its surface area, pore size, and structure, as a larger surface area and an optimised pore structure promote adsorption and aggregation, which improves the coagulation process' ability to remove contaminants from water.

Table 2.5: Surface morphology alteration for respective biosorbents from previous studies

Before adsorption	After adsorption	References
		Alghamdi et al., 2024
		James & Yadav, 2021

2.4.2 BET analysis

The surface properties which involved pore size distributions and micropore volume, are often evaluated using N₂-adsorption experiments using Micrometrics instrument to obtain the BET surface area (S_{BET}) (Mnisi & Ndibewu, 2017). The *M.oleifera* shown as a promising adsorbent for the removal of heavy metal because it has higher surface area 277.4 m² g⁻¹ and total pore volume 0.21 cm³ g⁻¹ than sugarcane bagasse (Ilaboya & Otuaro, 2019). Previous studies have altered the properties of *M.oleifera* by utilizing acid chemicals and metals to enhance the adsorbent capacity by providing a higher surface area and total pore volume. For instance, the *M.oleifera* is treated with 4% of nitric acid and methyl alcohol to increase the surface area and volume of pores for the removal of pharmaceutical wastes from

the aquatic environment (Araujo et al., 2018). Bispo et al. (2021) conducted a study that the use of phosphoric acid to modify *M.oleifera*, which had a surface area of $1356.29 \text{ m}^2 \text{ g}^{-1}$, resulted in the highest oil and grease removal capacity at 93%, compared to sodium hydroxide and zinc chloride. Meanwhile, Santos et al. (2021) using iron nanoparticles (AC-Fe) and 0.1 M hydrochloric acid to enhance the performance of *M.oleifera* with $578.4 \text{ m}^2 \text{ g}^{-1}$ of lead surface area to remove oils and grease at 94%. Nevertheless, the release of metals incorporated in the *M.oleifera* into water bodies may lead to water contamination. While prior research have demonstrated that treating the *M.oleifera* with acid chemicals and metals increases the surface area, the use of alkali and metal acids for activating the adsorbent is not sustainable. Therefore, the utilisation of agricultural wastes has the potential to be integrated with *M.oleifera* in order to increase the elimination of pollutants from wastewater.

Numerous researchers have investigated the repurposing of agricultural waste, specifically CH, to make activated carbon, which serves as an optimal adsorbent for removing various contaminants from wastewater due to its high porosity and reactivity (Soffian et al., 2022). Yuliusman et al. (2020) reported that the CH has the potential to become the activated carbon due to its high cellulose and lignin contents, cheap and can be found abundantly. Moreover, CH has high porosity and surface area that contribute to its enhanced adsorption capabilities. For instance, Mariana et al. (2019) demonstrated that the CH has high specific surface area which was $3.0438 \text{ m}^2 \text{ g}^{-1}$ with pore volume of $0.00751 \text{ cm}^3 \text{ g}^{-1}$. Another study conducted by Lo et al. (2023) reported that the surface area and pore volume of activated carbon from CH were $7.782 \text{ m}^2 \text{ g}^{-1}$ and $0.0237 \text{ cm}^3/\text{g}$ which is lower than magnetic activated CH, $95.157 \text{ m}^2 \text{ g}^{-1}$ and $0.1807 \text{ cm}^3 \text{ g}^{-1}$. Although the size of raw CH was $2.60 \text{ m}^2 \text{ g}^{-1}$, which was smaller than mentioned in previous studies, however, Droepenu

et al. (2021) concluded that the presence of multiple functional groups help to enhance the adsorption capacity. The acid activation using H_3PO_4 toward CH could increase the BET surface area and pore volume but the use of NaOH as activating agent could reduce both parameters as reported by Baharum et al. (2020). Conversely, Adorna et al. (2020) stated that the surface area and pore volume increase when treating the CH with KOH. Therefore, Ajien et al. (2023) summarised that the chemical activation does not always improve the surface area and porosity structure of the adsorbents.

2.4.3 FTIR analysis

The FTIR spectroscopy is commonly employed to identify functional groups in the biosorbents which responsible to form chemical bonds during the coagulation-flocculation process, hence indicating the potential applications of the plant in many applications. Several studies have utilised FTIR spectroscopy to investigate the functional groups present in *M.oleifera*, including hydroxyl, carboxyl, amino, phosphate and sulfonate groups to bind with pollutants such as dyes and metal ions (Escobar et al., 2021; Benettayeb et al., 2022). In addition, the use of FTIR spectroscopy has unlock the potential of *M.oleifera* for medicinal usage due to its bioactive compounds such as flavonoid, carotenoid, phenolic compound, alkaloids and amino acids (Taoualit et al., 2024). Previous studies reported that the broad band at $\sim 3400\text{ cm}^{-1}$ represent the hydroxyl group which can be found in proteins, carbohydrates, cellulose, lignin and fatty acids (Baptista et al., 2017; Araujo et al., 2018). Vibrations that observed in this region can also indicate that *M.oleifera* surface has the presence of N-H group (Viotti et al., 2019). The C-H stretching could be found at the region of $\sim 2929\text{ cm}^{-1}$ could be attributed to the CH_2 group in fatty acids found in methylcellulose (Kapse & Samadder, 2021). Other than that, Sya'banah et al. (2020) highlighted a peak at

~1746 cm^{-1} , associated with carbonyl region. Kebede et al. (2019) also identified a key peak at ~1631 cm^{-1} , corresponding to C=O bond of carboxylic acid in protein structure and fatty acid. The aromatic compound could be found at ~1500 cm^{-1} (Çelekli et al., 2019). The presence of C-O bond of carboxyl group at region 1100-1300 cm^{-1} was responsible for the adsorption sites for the removal of impurities from wastewater (Ravikumar & Udayakumar, 2020). Garcia-Fayos et al. (2016) and Villarreal et al. (2021) pinpointed that the appearance of C-O and C-N interactions in the region of 1050 cm^{-1} . In addition, the FTIR spectra of *M.oleifera*, illustrated in figure 2.5 (a) demonstrate the changes (i) before and after the adsorption of (ii) heavy metal Cr (VI) and (iii) Nile Blue B (NBB) dye, due to the presence of diverse compounds, including carboxyl, hydroxyl, and amines in the proteins of *M.oleifera*, which offer adsorption sites for pollutant capture, as reported by Shirani et al. (2018). The figure illustrates a decrease in intensity and a slight shift for NBB dye from a peak at 3433 cm^{-1} to 3415.3 cm^{-1} , highlighting the significant influence of hydroxyl and amine group domains in the adsorption of the anionic dye (NBB) on the surface of *M.oleifera*. In contrast, the asymmetric stretching frequencies shift from 2628.52 and 2922.50 cm^{-1} to 2856.53 and 2925.58 cm^{-1} , respectively, indicating electrostatic attraction between the anionic metallic species and the positively charged amino acid groups present in *M.oleifera*.

Next, CH has the ability to remove various pollutants such as heavy metal, colour, chemical oxygen demand (COD) and colour from wastewater due the presence of hydroxyl and carbonyl groups from lignin and cellulose (James & Yadav, 2021). The presence of a peak at ~3724 cm^{-1} in the hydroxyl stretching region and ~1600 cm^{-1} and in the C=O stretching region, as observed by Sidik et al. (2022), highlighting important one of the functional groups that responsible for the removal of turbidity, COD and colour from

polluted river water, primarily through electrostatic attraction. Bello et al. (2019) mentioned that C-H stretching alkane or alkyl groups can be found at $\sim 2926\text{ cm}^{-1}$ and $\sim 1377\text{ cm}^{-1}$. Other than that, Asadu et al. (2021) reported the presence of peak for silica compound, Si-O-Si at $1125\text{-}1296\text{ cm}^{-1}$. Mariana et al. (2019) asserted that the presence of silica compound could increase the adsorbent reactivity. Halfhide et al. (2019) outlined that the stretching region at $\sim 1726\text{ cm}^{-1}$ corresponded to C=O stretching of ester which can be found in hemicellulose, N-H stretching in primary amine at peak $\sim 1662\text{ cm}^{-1}$ and C-H stretching belonged to lignin at peak $700\text{-}900\text{ cm}^{-1}$. Next, the FTIR spectra of CH, as depicted in figure 2.5 (b), demonstrate oxidation in various chemical groups of the lignocellulosic coconut husk (i-iii), resulting in the formation of carboxyl and ether groups, as well as alterations in the polyphenolic structures after adsorbing heavy metal, Cr (VI) shown by the blue line, as documented by Verma et al. (2021). The alterations indicate that the adsorptive elimination of Cr (VI), a redox reaction has occurred, resulting in the oxidation of some functional groups on the organic adsorbent. Following the adsorption of heavy metals, the peak at (i) 3342.15 cm^{-1} sharpened, indicating reactions involving hydrogen bonding and a reduction in hydroxyl groups due to condensation and crosslinking processes. A study by Hanafiah et al. (2020) showed similar findings, indicating that the carbonyl group and hydroxyl group shifter to a lower wavelength, which implies that the CH has higher adsorption of toxic metals such as chromium and nickel. Concurrently, peaks shifted to ii) 1607.01 cm^{-1} and 1511.68 cm^{-1} , respectively; intensity diminished, indicating loss of the aromatic structure, potentially due to oxidation. The extensive peak transformed into distinct peaks at iii) 1218.3 cm^{-1} and 1229.2 cm^{-1} , signifying ether production resulting from condensation processes. Ultimately, the functional groups found in each adsorbent interact with different types of pollutants such as heavy metals, dyes, turbidity and other colloidal particles present in the

wastewater, facilitating a coagulation and flocculation process that aids in the purification of the wastewater.

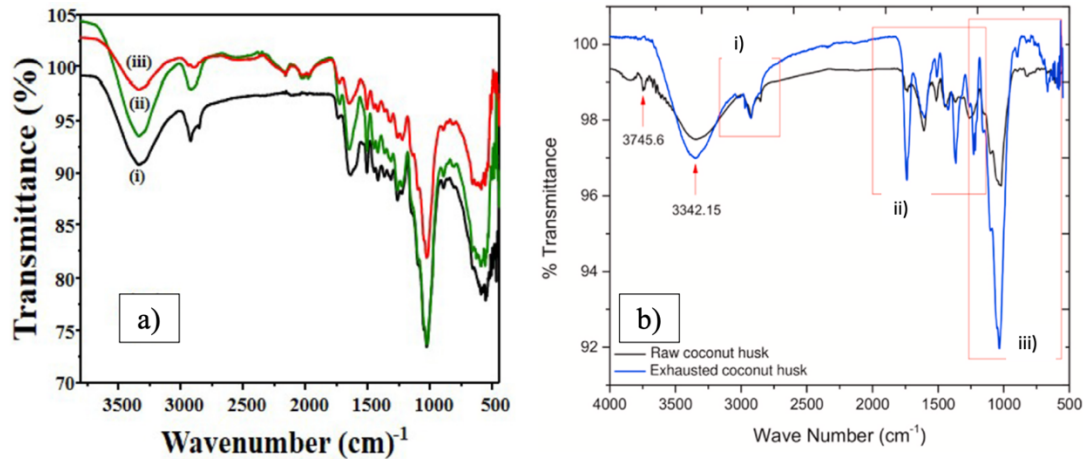


Figure 2.5: FTIR analysis of respective biosorbents (a) *M.oleifera* (b) coconut husk. Illustration adapted with permission from Shirani et al. (2018) and Verma et al. (2021).

2.4.4 Zeta potential analysis

Zeta potential measurement is crucial for evaluating the stability of colloidal suspensions, particularly in the context of wastewater treatment and the coagulation-flocculation process and the isoelectric point (IEP) is a vital indicator of the colloidal system's stability (Karunakaran, 2024). The zeta potential of *M.oleifera* and CH have been a topic of interest in various studies. According to Yamaguchi et al. (2021), the zeta potential of *M.oleifera* typically exhibits positive values at low pH and negative values at high pH. Baptista et al. (2017) found that the zeta potential for *M.oleifera* is positively charged as a result of the presence of cationic proteins. Ribeiro et al. (2019) demonstrated that the addition of *M.oleifera* into kaolin suspension resulted in a greater positive zeta potential. This indicates that *M.oleifera* can eliminate turbidity as shown by its capability to neutralise the negative zeta potential of colloid present in the kaolin suspension which achieved through the coagulation-flocculation process, where adsorption and neutralization of charges

occur. Apart from that, Jung et al. (2018) discovered that prolonging the extraction period of *M.oleifera* led to a decrease in its positive charge, hence impeding its ability to effectively eliminate turbidity.

Other than that, Yadav et al. (2021) stated that the adsorption of heavy is more favourable at higher pH levels when the CH is negatively charged, as shown by the negative zeta potential. The study concluded that the CH had a positive zeta potential at a pH lower than 4, which could impede the adsorption of heavy metals. Similar conclusions were drawn by Aktar et al. (2023), emphasising that the rate of heavy metal removal was enhanced with elevated pH values due to the occurrence of electrostatic attraction. Overall, the zeta potential which conducted at different pH levels is crucial to understand the surface characteristics and underlying mechanisms of both *M.oleifera* and CH.

2.4.5 Isotherm and kinetic models

Isotherm and kinetic models are essential in adsorption studies to understand adsorbate-adsorbent interactions and adsorption mechanisms. The adsorption efficacy and mechanism of biocoagulants, such as *M.oleifera* and CH, in the removal of impurities from wastewater are often evaluated via isotherm models, such as Langmuir and Freundlich isotherm models, and kinetic models, such as pseudo-first-order and pseudo-second-order kinetic model (Praveena & Shamsudin, 2020; Imran et al., 2022).

The Langmuir and Freundlich's isotherms are commonly utilised to comprehend the mechanism and occurrence of adsorption process for the removal of contaminants from the water. Çelekli et al. (2019) investigated the adsorption of reactive red 120 dye on *M.oleifera* and determined that the Freundlich isotherm provided the most accurate representation of

the adsorption process. Meanwhile, Ofgea et al. (2022) has modified *M.oleifera* using H_3PO_4 to maximise the removal of methylene blue dye from the water bodies, with the Langmuir model isotherm fitting the experimental data better than Freundlich isotherm. This suggests that the process involved the uniform adsorption of the adsorbate onto the surface of adsorbent, consistent with the Langmuir model. Additionally, Cusioli et al. (2021) studied the adsorption capacity of atrazine by using modified *M.oleifera* using nitric acid and found that the Langmuir model provides the most accurate description of the process occurring in a monolayer. Apart from that, previous studies has reached common agreement that the Langmuir model provides the most accurate representation on the adsorption capability of heavy metals utilising *M.oleifera* from contaminated water (Hegazy et al., 2021; Khader et al., 2022; Nwagbara et al., 2022). Some studies reported that Freundlich model described the adsorption of heavy metals using *M.oleifera* better than the Langmuir model (Ogarekpe et al., 2019; Swelam et al., 2019). This implies the adsorbate creates a single-layered film on the surface of the of the adsorbent (Bagheri et al., 2020). A study conducted by Rosmawanie et al. (2018) suggested that Langmuir model was the most suitable to represent the removal of turbidity from wastewater. On the other hand, Adebayo et al. (2022) investigated that the removal of Congo red dye by incorporating the iron salts and clay into CH followed the Langmuir model. A study by Bello et al. (2019) on the removal of rhodamine-B dye using acid activated CH also agreed with Langmuir model. In addition, although Verma et al. (2021) Furthermore, Verma et al. (2021) asserted that the Langmuir model is the most appropriate model for representing the experimental data on the uptake of chromium from wastewater using CH. However, the regression value falls below 0.9, indicating that there may be other mechanisms of removal involved besides simple adsorption.

Other than that, kinetic models such as pseudo-first order and pseudo-second-order kinetic models are usually used to investigate the mechanism of adsorption of heavy metals, dye and turbidity from the aqueous solutions by using *M.oleifera* and CH. Jabar et al. (2020) investigated the removal of congo-red dye removal using *M.oleifera* and found that the adsorption kinetics followed a pseudo-second order with high correlation coefficients. Kowanga et al. (2016) explored that adsorption of heavy metals such as Cu (II) and Pb (II) using *M.oleifera* and figured out that the experimental data fitted the pseudo-second-order kinetics better than the pseudo-first order kinetic. Similarly, Gautam et al. (2020) observed that the pseudo-second order kinetic model provided better correlation coefficients for *M.oleifera*. The findings revealed that the highest removal of lead from textile wastewater can be achieved by using *M.oleifera* compared to other natural adsorbents such as *Prosopis juliflora* and peanut shell. Therefore, the mentioned studies suggest that the adsorption of pollutants onto *M.oleifera* is most likely controlled by the chemisorption mechanisms, Chemisorption is often characterised by the formation of strong chemical bonds, such as ionic or covalent bonds, between the adsorbent surface and the adsorbate (Oribayo et al., 2021). However, Varsani et al. (2022) revealed that the experimental data for the removal of turbidity using *M.oleifera* treated with HCl agreed with pseudo-first order kinetic model, indicative of physisorption, which is governed by weak interaction such as Van Der Waals and dipole interactions between the colloidal particles with adsorbent. A similar result was obtained by Rasheed et al. (2023) which suggests that the physisorption process occurred in the removal of turbidity from the synthetic wastewater. In addition, the removal of dyes from wastewater effluent using activated CH as adsorbent fits the pseudo-second order kinetic model with high regression value (Imran et al., 2022). According to Johari et al. (2016), the highest removal of mercury from wastewater could be done by treating the CH with NaOH

compared to H_2O_2 and NaOCl which all the adsorbents agreed with pseudo-second order kinetic model. Hence, this highlights the effectiveness of *M.oleifera* and CH as a sustainable and eco-friendly adsorbent for various pollutants in wastewater. In summary, the literature review demonstrates an increasing interest in using *M.oleifera* and CH as biosorbents to eliminate different contaminants from water solutions. The studies have explored various modification techniques, adsorption capacities, and kinetic models to enhance the efficiency of the process.

2.5 Water quality parameters

2.5.1 Turbidity

Turbidity is influenced by the light absorption and dispersion of suspended particles which usually quantified by using turbidity meter and expressed in Nephelometric Turbidity Units (NTU) (Oladoja et al., 2017). The turbidity removal rate could be substantially influenced by pH values since a highly alkaline environment can decrease the turbidity removal rate due to protein instability in *M.oleifera* seed (Ng & Elshikh, 2021). The right amount of coagulant dosage plays a vital role in determining the performance of turbidity removal because the excess dosage of coagulant will not interact with oppositely charged colloidal particles, but leading to higher turbidity in the water (Nhut et al., 2021).

Nisar and Koul (2021) listed that the required coagulant dosage range (mg/L) of *M.oleifera* seeds powder to be used in turbid water are as follows in the concentration of *M.oleifera* seeds being 10 -50 mg/L for less than 50 NTU water, 30 - 100 mg/L for 50 - 150 NTU water while 50 - 200 mg/L for more than 150 NTU water. Following a study conducted by Vunain et al. (2019), *M.oleifera* seeds are capable of reaching the allowed turbidity of drinking water as listed by WHO standards when high concentration doses of *M.oleifera* seeds, which are 5000 mg/L and 150000 mg/L explicitly used to treat high turbidity water with more than 150 NTU. *M.oleifera* is more suitable for high turbidity water as the chemical bridging and adsorption process can occur when there are more charged sites (Gaikwad & Munavalli, 2019; Rocha et al., 2019).

The application of *M.oleifera* seed for water treatment exhibited the highest removal efficiency of turbidity over 90% (Bouchareb et al., 2020; Andrade et al., 2021; Araújo et al.,

2021). Previous studies also reported that the use of CH could remove turbidity (Liza et al., 2022; Sidik et al., 2022). Taylor et al. (2023) stated that the utilisation of untreated CH effectively reduced turbidity in greywater by up to 63%. Contrarily, the utilisation of both CH and limestone results in high removal of turbidity at 83% from greywater (Nurulhuda & Imran, 2023). In short, both *M.oleifera* and CH have the capacity to eliminate turbidity effectively from various water sources.

2.5.2 Electrical conductivity

Conductivity is precisely correlated to the ion concentration in water, and the conductivity values must not exceed 400 $\mu\text{S}/\text{cm}$ according to WHO standards (Meride & Ayenew, 2016). The addition of *M.oleifera* seed to a water sample can reduce conductivity significantly (Adeniran et al., 2017; Olanrewaju et al., 2018; Gandiwa et al., 2020; Al-Jadabi et al., 2021; Kithaka et al., 2022). This is because the EC removal was caused by floc formation as a result of the coagulation-flocculation process, which then separated from water, as opposed to alum, which increases water conductivity due to the dissociation of inorganic matter and the reaction of water with both acids and bases metals (Hendrawati et al., 2016). Contrarily, the addition of *M.oleifera* to water did not considerably lower its conductivity but increased it due to more anionic polyelectrolytes in the seeds (Shan et al., 2017).

On the other hand, Bigat & Awang (2021) studied that the EC increased after adding CH to remove the methylene blue solution. The study found that CH had a greater capacity to adsorb dyes compared to bamboo and palm shell, while having higher conductivity values than palm shell but lower than bamboo shell. Another study reported that the treating the coconut using H_3PO_4 has high EC (Nyamful et al., 2021). Consequently, the combination

of *M.oleifera* and CH might be able to remove EC from the effluent as a result of the enhanced surface area and the presence of more functional groups (Shah et al., 2024).

2.5.3 Total dissolved solids (TDS) and total suspended solids (TSS)

The total amounts of charged ions, such as salts, minerals, and metals, are referred to as TSS and TDS, in which excessive solid particles in water can result in corrosion and scaling in water system applications, as well as adversely affect the aquatic environment by reducing sunlight penetration into the water (Adjovu et al., 2023). The TDS and TSS levels, which are measured in milligrams per litre (mg/L) of water, have a direct correlation with the effectiveness of wastewater treatment systems and the permissible limit for both parameters should be less than 25 mg/L to allow the treated wastewater to be used as water supply in accordance with National Water Quality Standard Malaysia (NWQSM) (Hussain & Abdullah, 2018).

Many methods have been employed to modify the properties of *M.oleifera* to remove TDS and TSS from the wastewater with more efficient. For example, maximum removal of TDS and TSS from sewage wastewater up to 76% can be achieved when *M.oleifera* treated using NaOH compared to HCl according to Varsani et al. (2022). This is because the *M.oleifera* contains a positively charged protein such as lysine, histidine and arginine that bound to negatively charged particles in sewage wastewater, causing them to form clumps and settle down as sediment, thus reduces the levels of dissolved solids and suspended solids. Ntibrey et al. (2020) highlight that the use of *M.oleifera* when coupled with sand filtration able to remove TDS from bath wastewater at 72.7% after 2 hours. In addition, Lulesa et al. (2022) reported that the removal of TDS and TSS increase when the dosage of *M.oleifera*

increase. However, it is noteworthy to use suitable coagulant dosage to prevent oversaturation of contaminants in the water. This statement is strengthened by Kaushal & Goyal (2019). Likewise, an insufficient amount of coagulant dose is also unfavourable as it prolongs the deposition process and results in smaller floc size (Merdana et al., 2021).

Instead of employing concentrated acids like sulphuric acid and hydrochloric acid, incorporating activated CH into *M.oleifera* offers a more environmentally responsible approach and promotes circular economy, in line with the principles of green chemistry. The utilisation of CH has been found to surpass the effectiveness of palm kernel shell, sugarcane bagasse, and banana trunk in terms of removing TSS from wastewater, achieving a remarkable removal rate of up to 93.84% (Nasaruddin & Mohamed, 2021). Pradeep et al. (2016) investigated that the effective reduction of TDS using CH could be achieved within 1 hour. Lastly, Sundari et al. (2023) reported that the use of CH could remove TDS, TSS, BOD and COD from textile wastewater to a desired water quality. Overall, both *M.oleifera* and CH have the capability to reduce the TDS and TSS content from water samples but certain adjustments are necessary to effectively enhance their removal efficiency.

CHAPTER 3

METHODOLOGY

3.1 Introduction

This chapter covers the materials, procedures, and equipment used in this study. Further elucidation is provided on the formulation of various dosages employing *M.oleifera* and coconut husk (CH), as well as the synthetic wastewater. In addition, this chapter also presents an overview of the coagulation-flocculation process utilising specific coagulants to eliminate contaminants from the synthetic wastewater. The main procedures in this work can be categorised into three stages: (1) Determining the impact of various dosages of *M.oleifera* and CH on the physicochemical properties of biosorbents (2) Evaluation of the effectiveness of removing total dissolved solids (TDS), total suspended solids (TSS), electrical conductivity (EC), pH, and turbidity from wastewater at various pH and temperature levels utilising individual coagulants and combined *M.oleifera* and coconut husk (MOCH) and (3) Explaining the coagulants' adsorption mechanism using the Freundlich, Langmuir, and pseudo-first and pseudo-second order kinetic models. Illustration of the research flowchart for this study is provided in Figure 3.1.

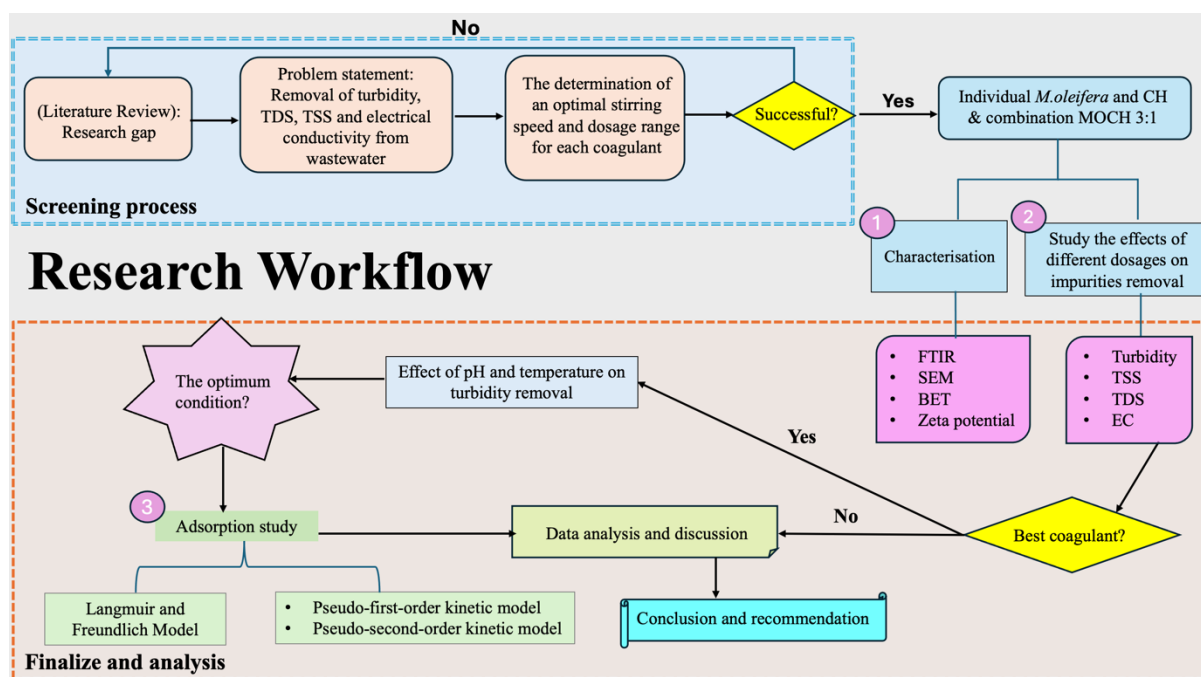


Figure 3.1: Summary of research workflow

3.2 Materials and instrumentations

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 (Fereja et al., 2020). 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 (Zaid & Ghazali, 2019). The initial turbidity of the raw water was assessed, and the average value obtained from these measurements would serve as the starting point. Before immersing the clean probe into the synthetic wastewater samples, the

multifunction PCD 650 waterproof portable meter sensor must be carefully chosen and calibrated to accurately measure EC and TDS (Shan et al., 2017). The *M. oleifera* and CH were commercially procured from Babymommom.com.my, a supplier company based in Kuala Lumpur, Malaysia.

3.3 Preparation of coagulants

A 300-mesh sieve (50 μ m) ASTM was used to sieve the seed powder (Choudhary & Neogi, 2017; Zaid & Ghazali, 2019). The preparation of coagulants was conducted following the method proposed by Mohammed et al. (2021). A 0.02 g of seed powder was mixed with 100 mL of 1M NaCl salt solution to extract the active component from the seed. The method of determining optimum stirring speed was trial and error with stirring speed of 100 rpm, 200 rpm 300 rpm, 400 rpm and 500 rpm based on the suggested in the literature review (Gómez et al., 2015; Novita et al., 2019; Zaid & Ghazali, 2019). The solution was stirred for 15 minutes using a magnetic stirrer (Remi, 1 MLH India) at optimum speed which was 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 (Al-Jadabi et al., 2021).

Next, the CH was prepared using a method suggested by Sujiono et al. (2022). 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 NaOH activator was used to activate the sample chemically by a ratio of 1:2 (1 g of CH activated carbon: 2 g powder of NaOH), which was dissolved with 10 ml of aquabides. The sample was dried in an oven to obtain a powder form, and it was then combined with the optimum coagulant dosage of *M.oleifera* to treat turbidity,

TDS, TSS, and EC. The EC was recorded in microsiemens per centimetre ($\mu\text{S}/\text{cm}$) using a conductivity meter, while the pH of the solution was determined using a pH meter (EUTECH, Singapore). The instruments must be calibrated using a standard buffer solution before taking the initial measurement to obtain accurate results. Finally, 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 most effective combination for application in this project was determined by conducting a systematic evaluation of the performance of these ratios in the coagulation-flocculation process. The sample designation for the coagulant dosages is shown in Table 3.1.

Table 3.1: Coagulant dosages used for coagulation-flocculation process

Coagulants	Dosages				
<i>M.oleifera</i> (g)	0.02	0.04	0.06	0.08	0.10
Coconut husk (CH) (g)	0.02	0.04	0.06	0.08	0.10
Combined <i>M.oleifera</i> and coconut husk (MOCH)	1:3	1:4	3:1	4:1	4:5

3.4 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 as shown by figure 3.2. 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. (2020). 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 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 were adjusted according to different kaolinite ranges from 10-60 g. The filtrate produced was utilized as a stock solution in the coagulation experiment. The turbidity of synthetic wastewater was measured before and after treatment with the coagulants.

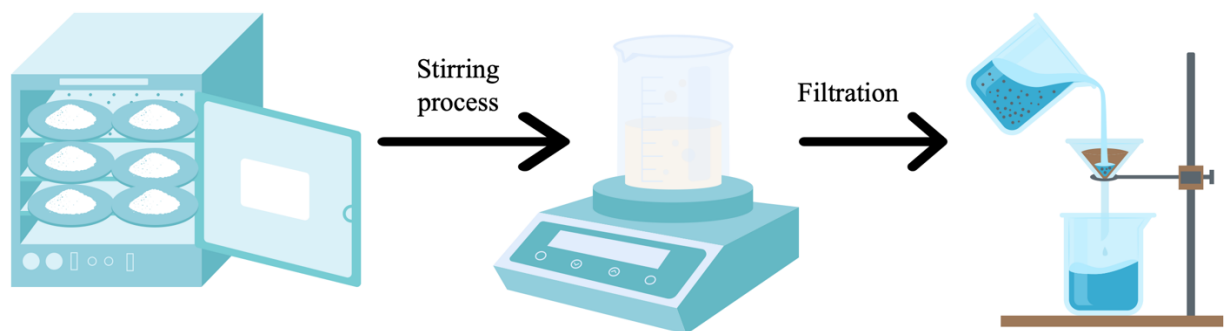


Figure 3.2: Preparation of synthetic wastewater

3.5 Turbidity measurement

A turbidity meter is frequently employed to evaluate the effectiveness of 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. (2024). The turbidity meter was first calibrated using standard calibration solutions 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 Equation 3.1:

$$\text{Turbidity removal (\%)} = \frac{\text{Turbidity initial} - \text{Turbidity final}}{\text{Turbidity initial}} \times 100\% \quad \text{Equation 3.1}$$

3.6 Surface morphology and chemical characterization method

The Scanning Electron Microscope (SEM-JEOL JSM-6390LV/LGS, Tokyo, Japan) 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 and image was obtained. Apart from that, FTIR analysis was conducted based on previous research by Okoya et al. (2020). The FTIR KBr (Thermo Scientific Nicolet iS10, Waltham, MA, USA) technique was used to analyze the materials. The IR-grade KBr was dried first at 110°C by using 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

3.7 Surface area and pore analyser method

The adsorbents were analysed using a surface area and pore size analyzer (Thermo Scientific, USA), followed by the Tokay & Akpınar (2021) 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 analysed 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 according to Equation 3.2 using the software provided by the manufacturer.

$$\frac{P}{n^a(P_0 - P)} = \frac{1}{n_m^a} + \left(\frac{C-1}{n_m^a C} \right) \frac{P}{P_0} \quad \text{Equation 3.2}$$

Where, the variable " n^a " represents the quantity of gas adsorbed at the relative pressure P/P_0 . " 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 Equation 3.3.

$$D_p = \frac{4V_T}{S} \quad \text{Equation 3.3}$$

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

3.8 Zeta potential method

Zeta potential measurements were conducted using a Laser Particle Size Analyser (LPSA-MASTERSIZE 3000, Malvern Panalytical, UK) to determine the surface charge of the composites with size of particles ranging from 1 nm to 10 μm by measuring the intensity of light scattered as a laser beam passes through a distributed sample of particles. The following method was in accordance with a study by Bezerra et al. (2020). 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.

3.9 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 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. This method was derived from Fajar (2020) with modifications to the stirring speed aimed to improve coagulation efficiency for this study's conditions. The solution was stirred up at 100 rpm for 1 minute, followed by flocculation at 40 rpm for 10 minutes. 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 were 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 ratio.

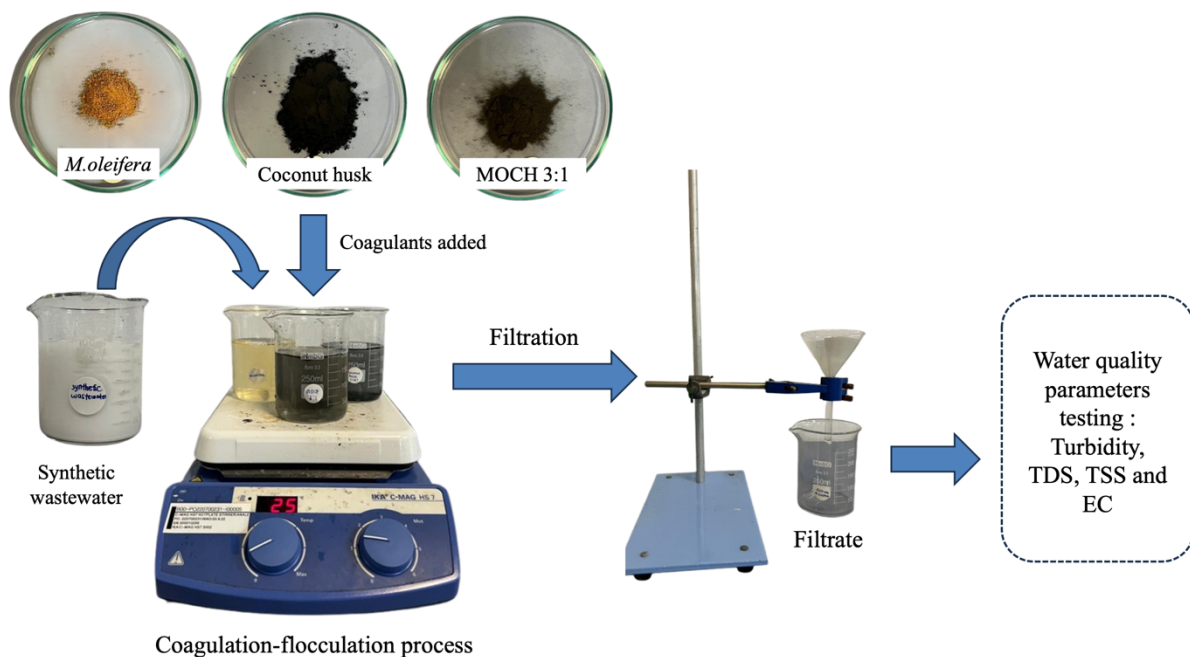


Figure 3.3: Illustration scheme for coagulation-flocculation process

3.10 Total suspended solids method

The total suspended solids (TSS) measurement was according to the standard method as described in American Public Health Association (APHA) (2012). 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 weight was recorded in milligrams per litre (mg/L) as the TSS in the water sample. The TSS was calculated before and after treating the synthetic wastewater were calculated based on the Equation 3.4.

$$\text{Total suspended solids} = \frac{W_F - W_I \times 1000}{\text{Volume of water}} \quad \text{Equation 3.4}$$

W_F = Final weight (mg/L)

W_I = Initial weight (mg/L)

3.11 Adsorption isotherm of MOCH 3:1

The adsorption process for turbidity reduction using MOCH 3:1 was investigated using an isotherm study on a sample of 0.2 L of synthetic wastewater. The initial concentration for turbidity varied between 50 NTU and 600 NTU. The experiment was carried out under optimised conditions at pH 6 and 25 °C. The solution underwent rapid mixing at 100 rpm for 1 minute, followed by slow mixing at 40 rpm for 10 minutes. An investigation was conducted on the equilibrium conditions for adsorption in synthetic wastewater samples using Langmuir and Freundlich isotherms to determine the interaction between colloidal particles and MOCH 3:1. The equations included in the isotherm model analysis are the following linearized equations (Rajalingam et al., 2021). The C_e/q_e was calculated by using Equation 3.5

Langmuir equations:

$$\frac{C_e}{Q_e} = \frac{1}{Q_{\max}} KL + \frac{C_e}{Q_{\max}} \quad \text{Equation 3.5}$$

Where q_e represents the equilibrium sorption capacity in milligrams per gram (mg/g), C_e indicates the equilibrium concentration in milligrams per litre (mg/L). $Q_{\max}$ is the maximum concentration of pollutants that can be adsorbed by a complete monolayer on the surface (mg/g) determined by the constant k , whose value reflects the affinity of the binding sites (L/mg). The value of $Q_{\max}$ was determined by using Equation 3.6.

$$Q_{\max} = V \times \frac{C_i - C_f}{M} \quad \text{Equation 3.6}$$

The variables in Equation 4.2 are as follows: V represents the volume of the wastewater sample in litres, C_i represents the initial concentration of the pollutant in milligrams per litre, C_f denotes the remaining concentration of the pollutant in milligrams per litre, and M indicates the amount of coagulant on a dry basis in grams.

Freundlich equation is shown in Equation 3.7.

$$\text{Log } q_e = \frac{1}{n} \log C_e + \log K_f \quad \text{Equation 3.7}$$

The variables in Equation 4.3 are as follows: K_f corresponds to the Freundlich constants, which indicate the adsorption capacity in milligrams per gram (mg/g), q_e symbolises pollutant uptake per unit weight of biosorption in mg/g, C_e denotes the equilibrium concentration in milligrams per litre (mg/L), and n signifies the empirical constant indicating the adsorption intensity in litres per milligram (L/mg). The slope corresponds to the reciprocal of n, while the intercept corresponds to K_f .

3.12 Adsorption kinetics of MOCH 3:1

The pseudo-first order kinetic model and pseudo-second order kinetic models were utilised to determine the adsorption process of colloidal particles from synthetic wastewater using MOCH 3:1 as an adsorbent (Rasheed et al., 2023). The pseudo-first-order equation is represented in Equation 3.8 (Hegazy et al., 2021).

$$\text{Log } (q_e - q_t) = \log q_e - \frac{K_1}{2.303} t \quad \text{Equation 3.8}$$

The variables in Equation 4.4 were q_t and q_e (mg/g), representing the quantity of kaolin particles adsorbed per unit weight of the adsorbent at time t and equilibrium, respectively. k_1 (min^{-1}) is the pseudo-first-order rate constant of the sorption process, and t (min) is the mixing time. This kinetics model assumes that the adsorption rate is directly proportional to the number of accessible free sites, with each ion exclusively occupying one site (Gomes et al., 2022).

Next, the pseudo-second-order kinetics model assumes that the sorption rate is governed by a chemical sorption mechanism that involves the sharing or transfer of electrons between the adsorbent and adsorbate (Viotti et al., 2019). It can be formulated as shown in Equation 3.9:

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e} \quad \text{Equation 3.9}$$

Thereby, variables q_t and q_e (mg/g) as shown in Equation 3.9, represent the quantity of kaolin particles adsorbed per unit weight of the adsorbent at time t and equilibrium, respectively. Additionally, k_2 (g/mg.min) is the pseudo-second-order rate constant of the sorption process, and t (min) refers to the mixing time.

CHAPTER 4

RESULTS AND DISCUSSION

4.1 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. 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.

4.1.1 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 for after coagulation-flocculation process. The adsorption process occurred, as evidenced by the discernible shift to the left 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 (Adeniyi et al., 2020; Raji et al., 2020).

The wide band at 3480 cm^{-1} corresponds to the hydroxyl group of *M.oleifera* from protein protein and there may be a contribution in this region pertaining to the N-H stretch of

amides (Escobar *et al.*, 2021). Following the coagulation-flocculation process, the band narrows due to hydrogen bond formation with Al-OH from kaolin (Dewi *et al.*, 2018). 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 Van der Waals forces. The peak at 1650 cm^{-1} indicates C=O vibration in the amide, which can form a covalent bond with Al^{3+} , acting as a Lewis acid that accepts electrons from the C=O. Additionally, the peak at 1400 cm^{-1} , corresponding to primary amine (NH_2), is capable of forming hydrogen bonds with Al-OH, resulting in a reduction of peak intensity following the coagulation-flocculation process while another potential interaction that may have occurred is the development of a coordination complex, as Al^{3+} can accept an electron pair from the nitrogen group of a primary amine (Bispo *et al.*, 2021). The peak at 1030 cm^{-1} in this region 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, leading to an increase in peak intensity and shiftment peak to the left, as shown in label (ii) in Figure 4.1, following turbidity removal (Faisle *et al.*, 2023). Subsequently, two peaks were detected between 500 cm^{-1} and 1000 cm^{-1} following 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 (Çelekli *et al.*, 2019).

The FTIR study of *M.oleifera* before coagulation, as depicted in Figure 4.1, revealed that these functional groups, which include the -COO, -OH, and - NH_2 play an essential role in increasing flocculation through neutralisation and bridging mechanisms (Nisha *et al.*, 2017). The peaks attributed at 1641 cm^{-1} and 1532 cm^{-1} in *M.oleifera* are due to the C=O vibration in the amide I and II, respectively (Baptista *et al.*, 2017; Swelam *et al.*, 2019). The presence of

a solid peak in the protein structure of *M.oleifera* for C-O corresponds to the presence of bands of 1800 cm^{-1} and 1600 cm^{-1} , while for C=O band in aliphatic ester was assigned in peak at 1746 cm^{-1} (Vigneshwaran et al., 2020). The peak at 2930 cm^{-1} was attributed to the stretching vibration of C-H bonds in polysaccharides, whereas the peak at 1030 cm^{-1} is associated with the C-O groups from the pyranose ring in polysaccharides of *M.oleifera* (Zhang et al., 2022). After the coagulation-flocculation process, the peaks were marginally altered to a higher wavenumber, and no peak was observed at 2930 cm^{-1} , indicating that the successful interaction between *M.oleifera* and the colloidal particle occurred (Bezerra et al., 2020).

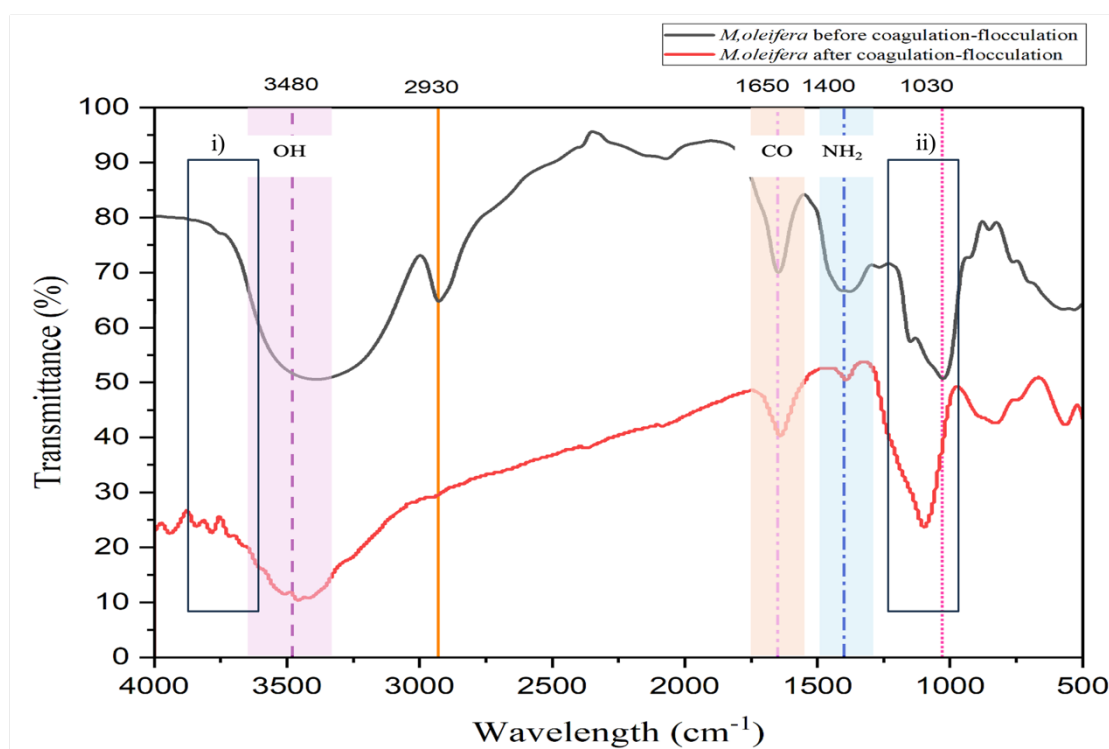


Figure 4.1: FTIR result of treating synthetic wastewater using *M.oleifera*

Other than that, Figure 4.2 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 (Johari et al., 2016). The absorption band at 3460 cm^{-1} corresponds to the hydroxyl group, OH, from the polysaccharide component of CH, 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 (Sidik et al., 2022). The minor peak at 2360 cm^{-1} indicates the presence of $\text{C}\equiv\text{C}$ in lignocellulosic material, as it exhibits minimal absorption of infrared light and the peak intensity increases after the coagulation-flocculation process, due to the generated dipole-dipole interaction between $\text{C}\equiv\text{C}$ and Al-OH from kaolin (Taiwo et al., 2019). Additionally, the $\text{C}=\text{O}$ 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 (Bello et al., 2019). The C-H stretching of the alkyl group in hemicellulose was detected around 1400 cm^{-1} and this alkyl group can form a transient dipole when interacting with Al-OH from kaolin via Van der Waals forces (Hanafiah et al., 2020). The Si-O group exhibited asymmetric bending at 1100 cm^{-1} , which can interact with Al-OH, resulting in hydrogen bonding (Mariana et al., 2019). The peaks 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 (Kaur et al., 2024). The decrease in peak intensity of C-H vibration from CH upon interaction with kaolin may be attributed to hydrogen bonding or electrostatic interactions.

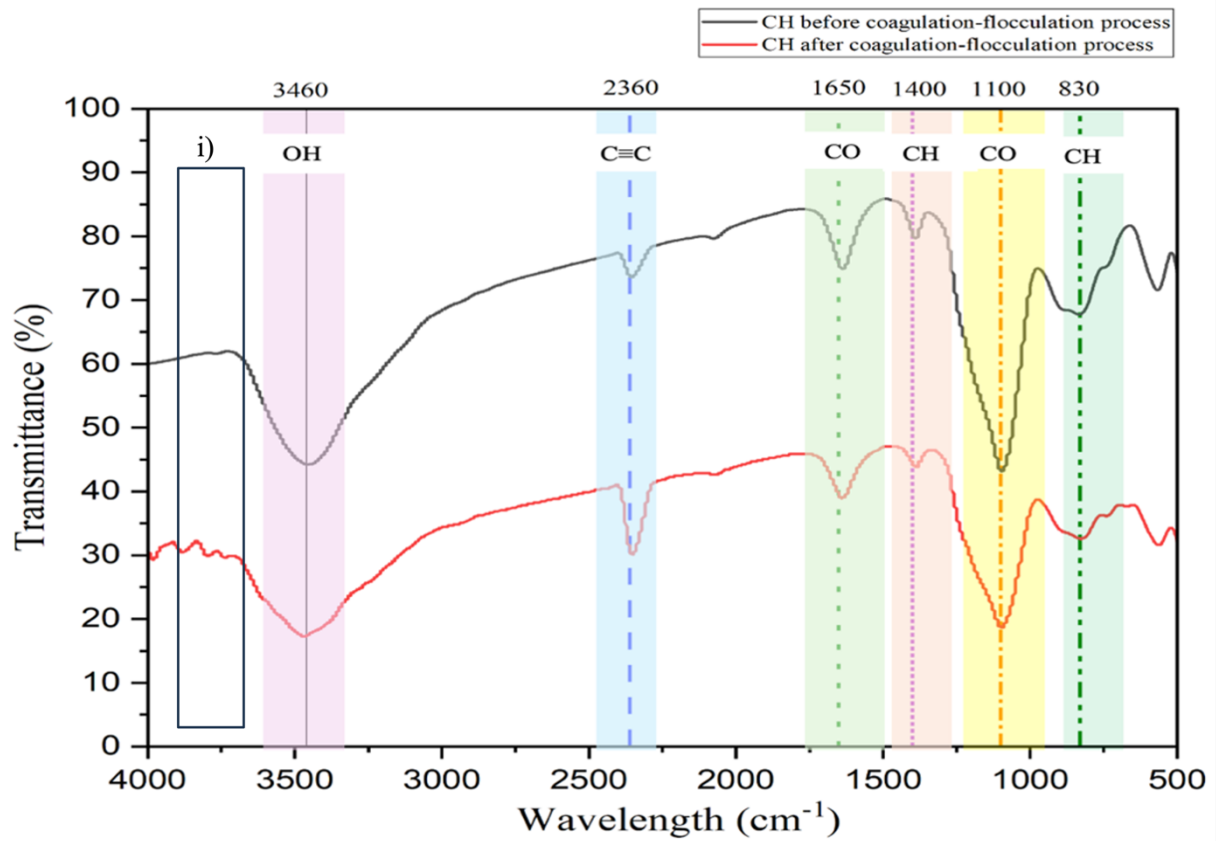


Figure 4.2: FTIR result of treating synthetic wastewater using CH

Nevertheless, the resultant composite is combined MOCH 3:1 as depicted in Figure 4.3. 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 (Sujiono et al., 2022). 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 (Escobar et al., 2021). 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, a non-polar siloxane group, via hydrophobic contact (Nonfodji et al., 2020). 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 (Paramita et al., 2024). The

interaction of both coagulants affected the electron distribution and weakened the bond frequency, hence reducing the intensity peak relative to each coagulant individually, as previously mentioned (Ahmed et al., 2022). 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 (Aal et al., 2022). The combination of *M.oleifera* and CH has facilitated an interaction between the NH₂ group of *M.oleifera* and the C-H group of hemicellulose in CH through weak hydrogen bonding (Benettayeb et al., 2022). Subsequently, during the coagulation-flocculation process, the Al-OH and Si-OH from kaolin interact with both functional groups present in the *M.oleifera* and CH combination, which caused an increase in peak intensity (Adeniyi et al., 2020). 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 (Yang et al., 2024). 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. Additionally, the two peaks found at 600 cm⁻¹ and 843 cm⁻¹ correspond to the contact between the C-O bond of the pyranose ring in *M.oleifera* and the Si-O bond in CH, facilitated by dipole-dipole interactions (Azlin & Syufiana, 2022). The decreased intensity in this observed peak resulted from the C-O group that can physically adsorb onto the surface of silica from CH. Upon the completion of the coagulation-flocculation process, the interaction between kaolin and MOCH 3:1 become more ordered which amplifies the peak because of the synergistic effect from the dipole-dipole interaction of both coagulants. In addition, the new peak observed at 1100 cm⁻¹ might indicate the interaction between *M.oleifera* and CH by the carbonyl group before the coagulation-flocculation process (Mohammed & Hawar, 2022). The peaks at 843 cm⁻¹ and 600 cm⁻¹ were

attributed to the stretching of hemicellulose or cellulose and stretching of lignin, respectively, from the CH (Siha et al., 2018). The hydroxyl, carbonyl, and amine groups present in the substance are responsible for its ability to coagulate and purify water via the adsorption process, confirming their role as excellent contaminants absorbents (Afolabi et al., 2021). This observation proved the potential for an interaction or connection between the components, which raises the prospect of harnessing the combined properties of both substances in composite applications.

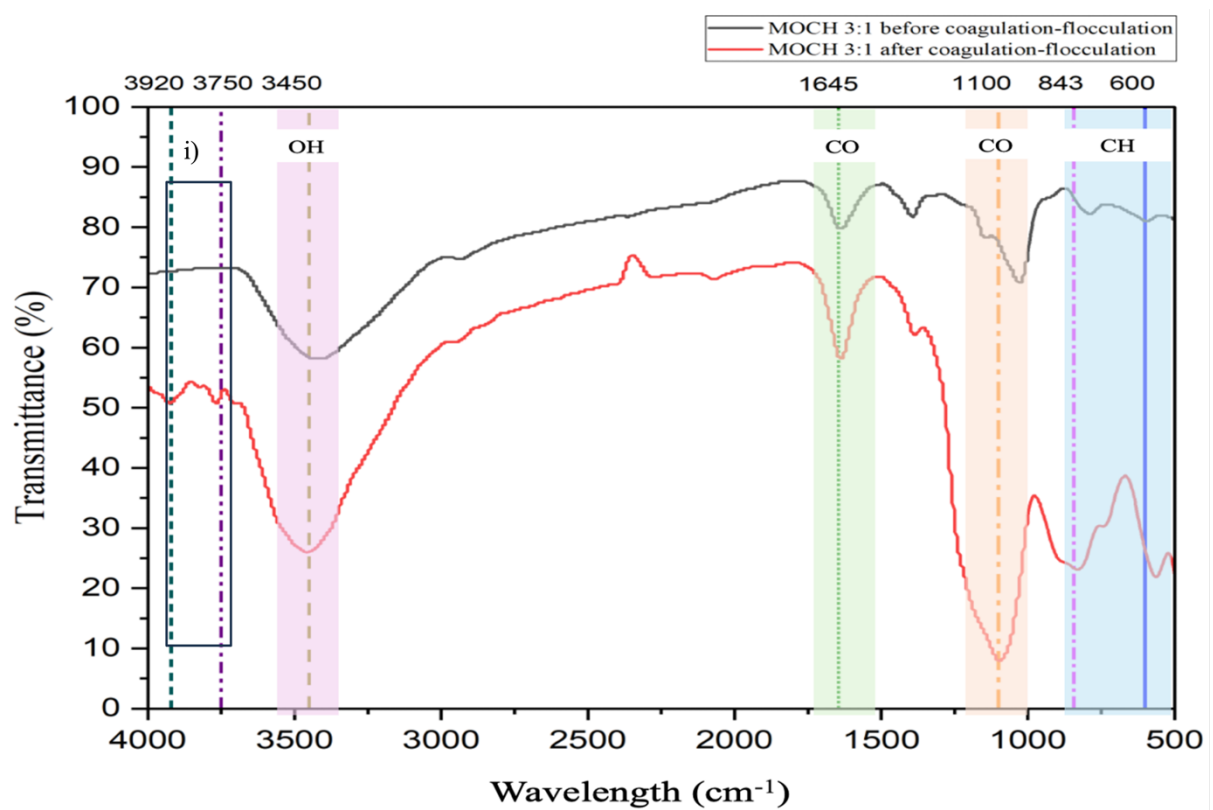


Figure 4.3: FTIR result of treating the synthetic wastewater using MOCH 3:1

4.1.2 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 analysed using scanning electron microscopy (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 4.4. The results of this study are concurrent with the conclusions made by Nonfodji et al. (2020) and Alghamdi et al. (2024). Numerous studies also stated that *M.oleifera* has a heterogeneous structure with a rough surface, irregularly formed blocks, many pores of varying sizes, and appears villous, indicating that the villus plays a vital role in the adsorption process (Swelam et al., 2019; Gautam et al., 2020; Gaayda et al., 2022; Gomes et al., 2022). The closed pores in *M.oleifera*'s structure were due to a significant amount of cellulose and lignin in its structure (Cusioli et al., 2021). Nevertheless, other studies have shown contradictory findings in which the natural *M.oleifera* has a smooth surface with a high number of pores (Çelekli et al., 2019; Escobar et al., 2021).

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. Previous studies found that CH is more effective than other plant biomass in water purification due to its dense surfaces with heterogenous pores, uneven, rough, irregular shapes, and particle sizes up to 100 μm which means that it has a large surface area and porosity (Taer et al., 2018; Mariana et al., 2019; Ahmad et al., 2022). 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. Furthermore, 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 resulted in notable physical changes to their structure after the coagulation-flocculation process. The

whole surface of the biosorbents was wholly coated with primarily the kaolinite that was initially added to the wastewater, as shown in Figure 4.4. Furthermore the pores on the biosorbent surface become more irregular, densely packed and intricate, making them difficult to discern. The observed outcomes might be attributed to the interaction of kaolinite and the biosorbents particles. This finding is concurrent with the study by Vigneshwaran et al. (2020). The SEM results demonstrated that the surface texture for all adsorbents exhibited softer, irregular, and porous following the flocculation procedure, as well as seemed to possess a substantial specific surface area that serves as a bridge between the coagulants and kaolin particles, thus improving the removal of turbidity. These findings elucidate the impact of coagulation and flocculation on the surface morphology of the coagulants, in which the accumulation of contaminants from the wastewater on the coagulants' surface leads to increased softness (Rosmawanie et al., 2018). 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 (Shah et al., 2024).

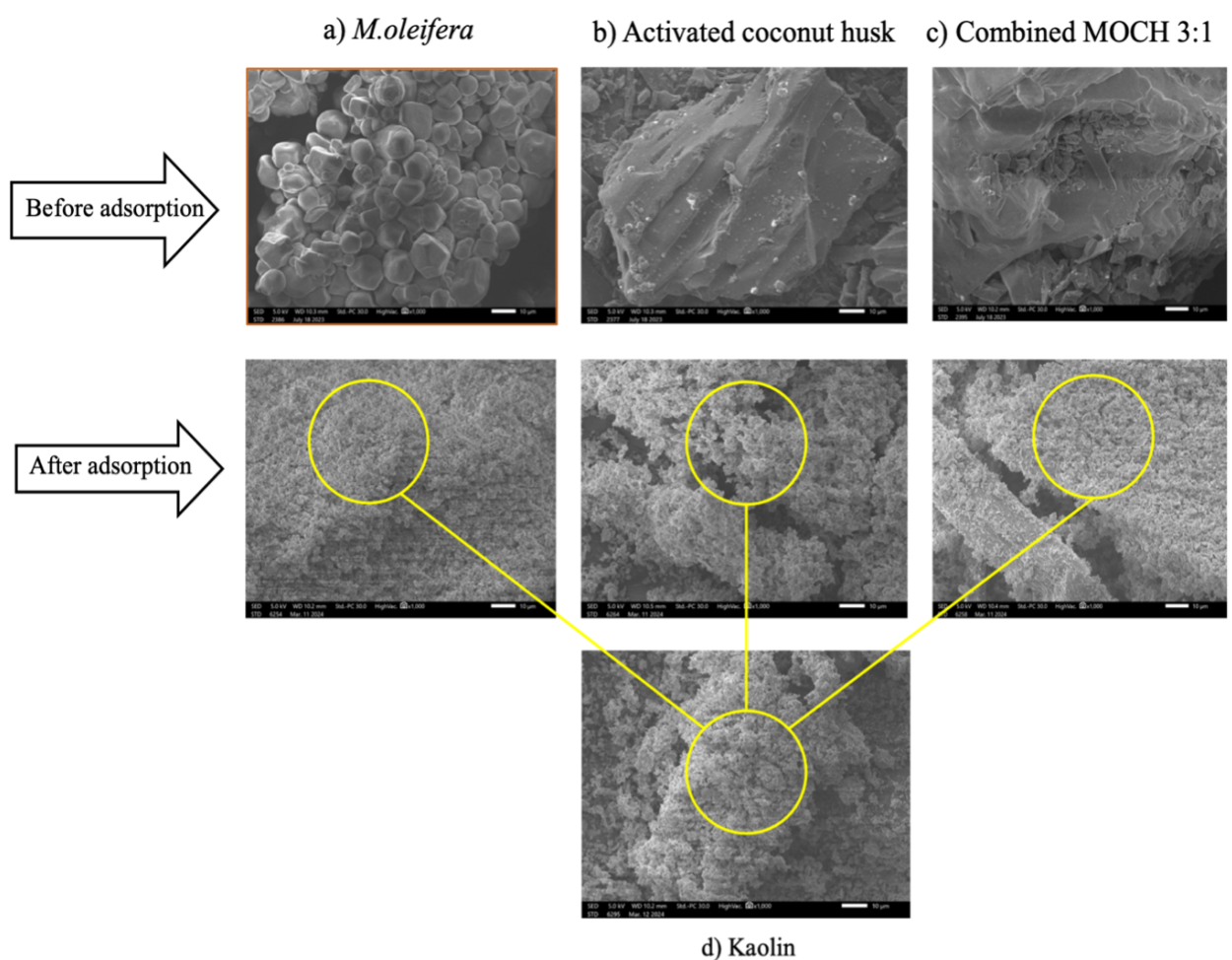


Figure 4.4: Surface morphology of respective coagulants at x1000 magnifications

4.1.3 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 4.5 show the BET results for individual coagulants. The morphological features of *M.oleifera* exhibit the smallest pore volume and surface area, with $0.001313 \text{ cm}^3 \text{ g}^{-1}$ and $0.1273 \text{ m}^2 \text{ g}^{-1}$, respectively. The coconut husk (CH) exhibited the greatest pore volume and surface area due to its porous

structure, at $0.343164 \text{ cm}^3 \text{ g}^{-1}$ and $552.1280 \text{ m}^2 \text{ g}^{-1}$. Fig. 4.5 (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 (Hendrasarie & Maria, 2021). 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 (Dragan et al., 2021). Subsequently, a type IV isotherm is initially established in the monolayer at low pressure, followed by the formation of a multilayer, as illustrated in Fig. 4.5 (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 (Ismail et al., 2020). 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.

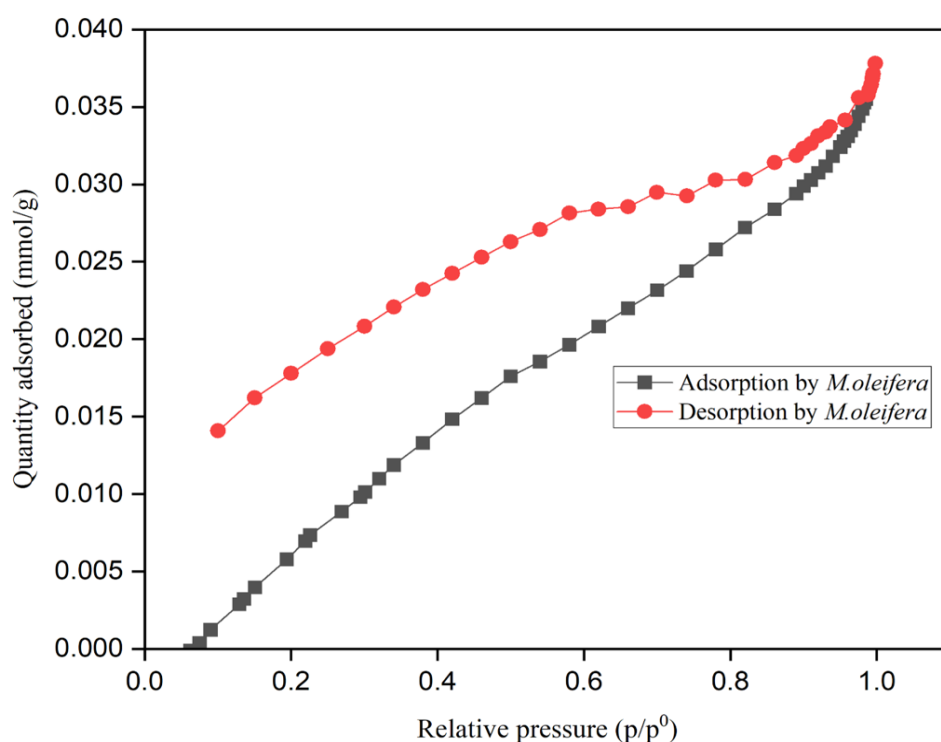
Next, Figure 4.6 illustrates that the MOCH 3:1 adheres to a Type IV isotherm with a moderate open loop hysteresis, indicating its mesoporous nature and a balance between physisorption and chemisorption. Consequently, the narrow necks of the mesopores in the MOCH 3:1 induce medium open loop hysteresis, hindering the escape of certain adsorbates and thereby obstructing the desorption process (Buttersack, 2019). 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. On the other hand, the MOCH 3:1 mixture exhibited greater pore volume and surface area compared to *M.oleifera*, with values of $0.010866 \text{ cm}^3 \text{ g}^{-1}$ and $17.1006 \text{ m}^2 \text{ g}^{-1}$, 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 (Samad et al., 2019; Benettayeb et al., 2022).

This discrepancy can be attributed to activated carbon CH being a porous carbonaceous material with a much larger surface area than *M.oleifera* (Soffian et al., 2022). Previous studies have shown that unprocessed *M.oleifera* have a specific surface area ranging from 1.2 to $1.52 \text{ m}^2 \text{ g}^{-1}$ and a total pore volume ranging from 0.002 to $0.021 \text{ cm}^3 \text{ g}^{-1}$ (Bezerra et al., 2020; Cusioli et al., 2021; Lopes et al., 2022). 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 (Araujo et al., 2018). Apart from that, the BET textural properties of MOCH have a high specific surface area of $17.1006 \text{ m}^2 \text{ g}^{-1}$, along with a total pore volume of $0.010866 \text{ cm}^3 \text{ g}^{-1}$. 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 (Bispo et al., 2021). *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*

treated with acid chemicals could produce higher BET values than the unprocessed seed. *M.oleifera* which has been treated with highly concentrated nitric acid (HNO_3), showed that its specific surface area was $5.77 \text{ m}^2 \text{ g}^{-1}$, and its total pore volume was $0.0409 \text{ cm}^3 \text{ g}^{-1}$, which is higher than *M.oleifera* seed that had been exposed to H_2SO_4 with a specific surface area of $1.2 \text{ m}^2 \text{ g}^{-1}$ and a total pore capacity of $0.002 \text{ cm}^3 \text{ g}^{-1}$ (Tavengwa et al., 2016; Cusioli et al., 2021). Therefore, the surface of MOCH 3:1 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.

a)



b)

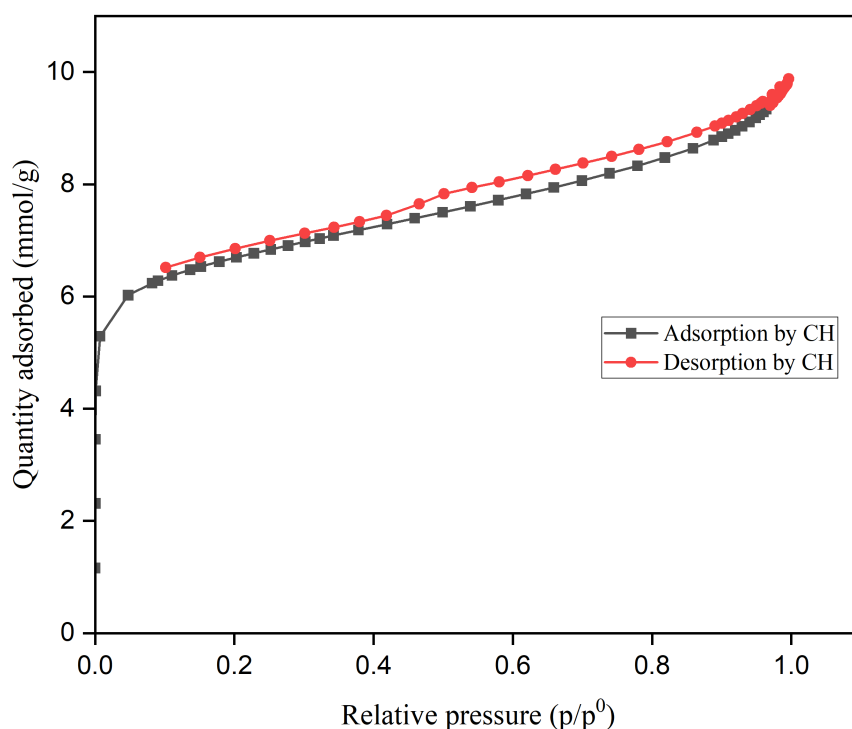


Figure 4.5: BET analysis of individual coagulants (a) *M.oleifera* (b) CH

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) (Abdullah *et al.*, 2017). 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 (Mane *et al.*, 2024). Previous studies reported similar observations (Hendrasarie & Maria, 2021; Sriramoju *et al.*, 2021).

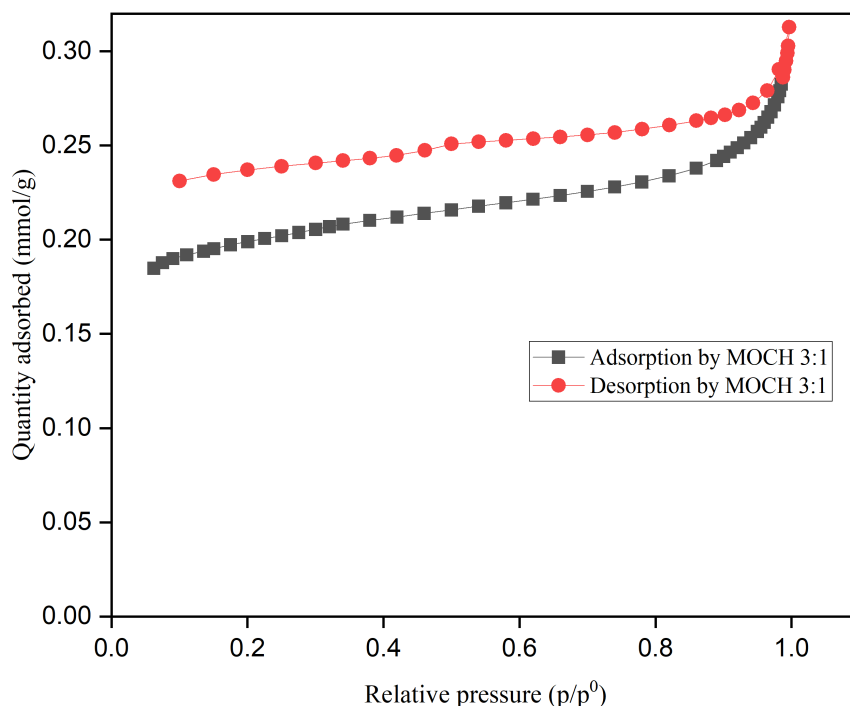


Figure 4.6: BET analysis of MOCH 3:1

4.1.4 Zeta potential analysis

The zeta potential is correlated with pH variations to understand better the mechanism underlying coagulation-flocculation capabilities of coagulants, as depicted in Figure 4.7. 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 is from -0.572 to -20.0.

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 (Hasan & Fatehi, 2019). 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 (Lester-Card et al., 2023). 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 (Vigneshwaran et al., 2020; Ng & Elshikh, 2021). This statement is consistent with researchers attributing that the zeta potential remained negative when using *M.oleifera* (Kansal & Kumari, 2014). Hence, the occurrence of coagulation through charge neutralisation was improbable (Skaf et al., 2021). The primary mechanism for coagulation is either adsorption or interparticle bridging of destabilised particles following the Langmuir adsorption model. Furthermore, it was mentioned that the two mechanisms could be occurring concurrently.

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 (Koul et al., 2022; Zainol et al., 2022). 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. This study is in contrast with the findings by Gaayda et al. (2022), which mentioned that the presence of cationic proteins in *M.oleifera* results in a positive zeta potential, while synthetic water has a negative zeta potential. As a result, the negatively charged colloids in a kaolin suspension are destabilised by the cationic polyelectrolytes in *M.oleifera*.

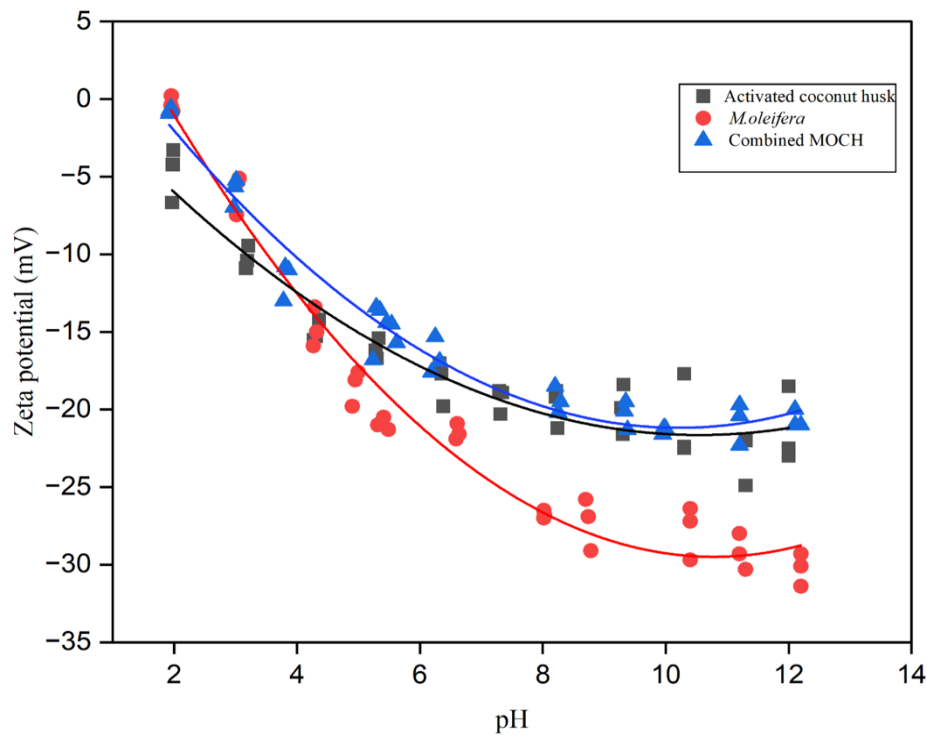


Figure 4.7: Zeta potential of respective coagulants

4.2 Turbidity removal

The initial and final turbidity of the synthetic wastewater samples were assessed before and after treatment, respectively. Table 4.1 conveys initial turbidity (300 NTU), TDS (2170 mg/L), TSS (8758.5 mg/L), and conductivity (2440 $\mu\text{S}/\text{cm}$) as a baseline for comparison with the final assessment. The difference between the original and final turbidity measurements were then used to calculate turbidity removal efficiency.

Table 4.1: Initial synthetic wastewater characterization

Water sample	Turbidity (NTU)	TDS (mg/L)	TSS (mg/L)	EC ($\mu\text{s}/\text{cm}$)
Synthetic wastewater	300	2170	8758.5	2440

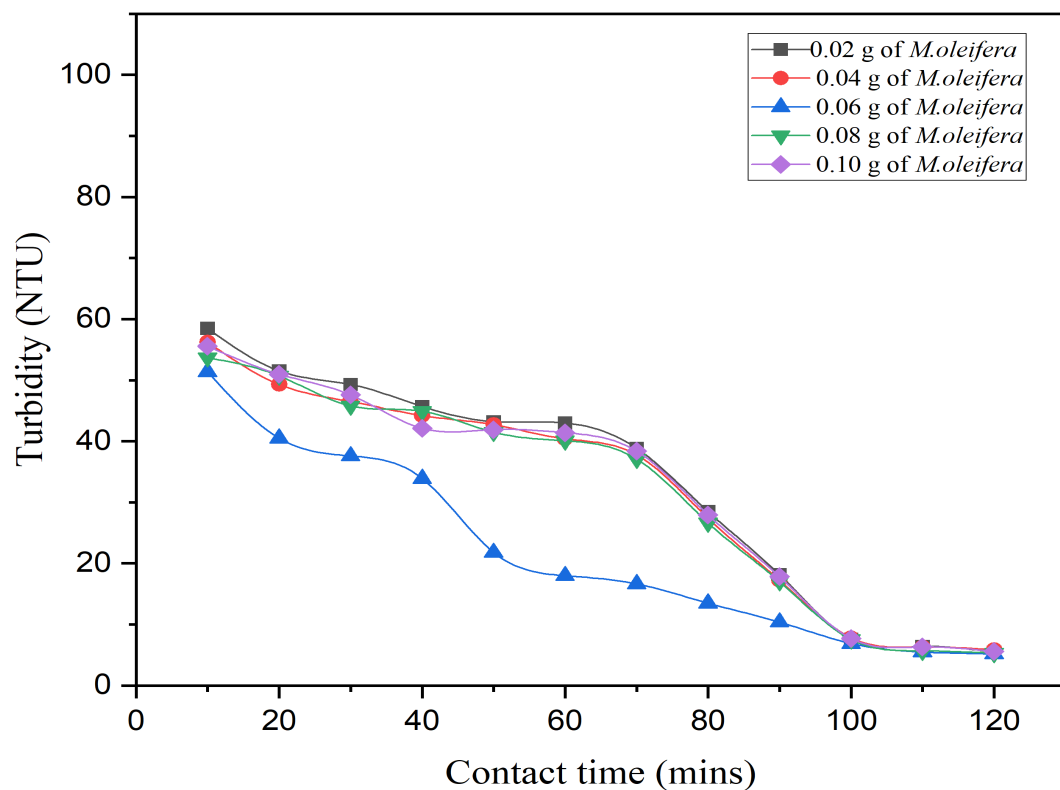
4.2.1 Turbidity removal using individual coagulants

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 mixture continued to swirl at 40 rpm for 10 minutes to clump the particles together. The stirring speed was operated slowly during the second stage to promote flocculation and prevent the irreversible disruption of flocs caused by polymer chain breakage.

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 4.8. 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 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 DOE (2024), which dictates turbidity levels below 5 NTU, underlining its effectiveness as a coagulant surpassing the control and further underlining their effectiveness as coagulants (Kamarudin et al., 2020). A study by Adelodun et al. (2019) 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 its optimal value due to their inability to interact with oppositely charged colloidal particles (Gaikwad & Munavalli, 2019). This study supports the hypothesis that peak efficiency is reached when the coagulant dosage and turbidity levels are optimal (Rasheed et al., 2023).

a)



b)

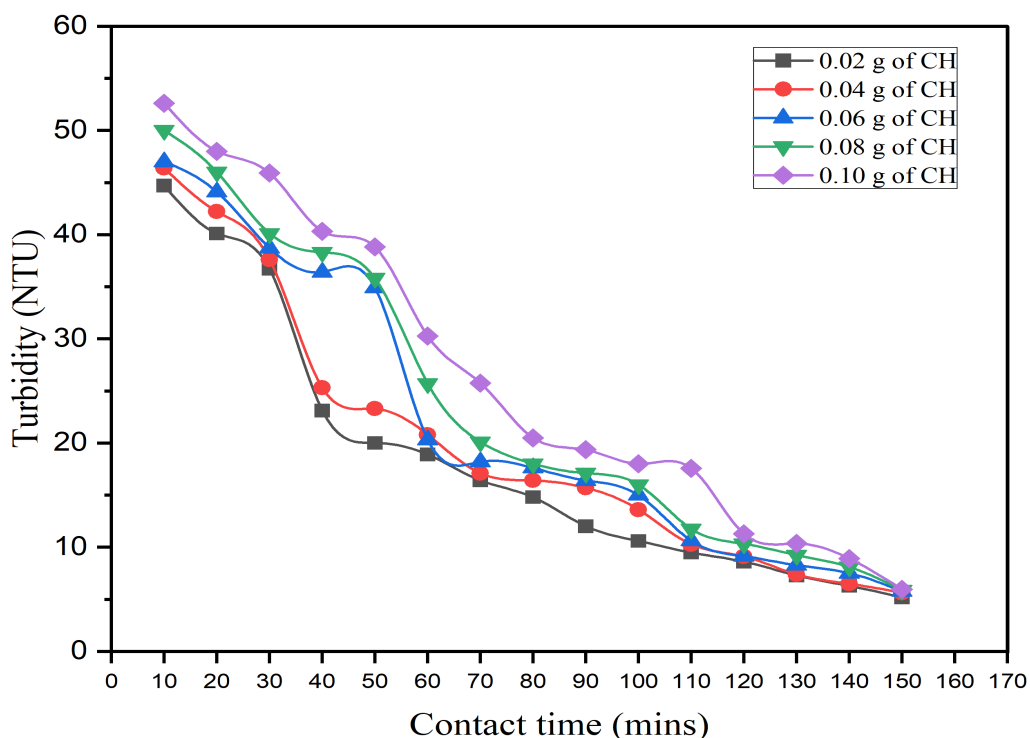


Figure 4.8: Turbidity reduction using (a) *M.oleifera* (b) CH at 300 NTU of synthetic wastewater

4.2.2 Turbidity reduction using different combined MOCH ratio

The optimal coagulant dosages from *M.oleifera* and CH are divided into the corresponding combined MOCH ratio in order to assess the coagulant combination's efficacy. Five distinct ratios were formed by combining the three optimal dosages of specific coagulants. This aims to determine that the combination of MOCH ratio achieves the highest level of turbidity removal at 300 NTU from the synthetic wastewater. Table 4.2 outlines various combinations of the MOCH ratios. Establishing the optimal coagulant mixture ratio is crucial for maximizing chemical dosage, thereby avoiding both overdosing and underdosing. The proper balance is essential for improving coagulation efficacy and guaranteeing the synergistic interaction among coagulants. Moreover, the strategic combination facilitates the

compensatory function of one coagulant, enabling a less efficient coagulant to be enhanced by another, thereby preserving or enhancing the overall efficacy of the coagulation process.

Table 4.2: Combination of different MOCH ratio

MOCH ratio	<i>M.oleifera</i> (g)	CH (g)	Total
MOCH 1:3	0.02	0.06	0.08
MOCH 3:1	0.06	0.02	0.08
MOCH 1:4	0.02	0.08	0.10
MOCH 4:1	0.08	0.02	0.10
MOCH 4:5	0.08	0.10	0.18

Figure 4.9 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. All the different combined MOCH treatments resulted in an immediate reduction in turbidity after 10 minutes of treating water at 300 NTU. The MOCH 3:1 facilitates more rapid and stable floc formation than individual coagulants due to its synergistic impact. The *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 (Dandesa et al., 2023). Conversely, MOCH 1:3 demonstrate lower turbidity removal than MOCH 3:1. This can be associated with a higher dosage of CH in MOCH 1:3 that hinders the capability of the adsorbent to remove turbidity (Sheba et al., 2019). Meanwhile, MOCH 4:5 showed the lowest efficacy in eliminating turbidity. This may arise from an excessive dosage of coagulant, which reduces its capacity to eliminate turbidity. This statement was supported by Getahun et al. (2024), 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, an excessive quantity of *M.oleifera* may result in oversaturation, causing the active sites to become completely occupied and subsequently diminishing the total coagulation effectiveness. Conversely, an overabundance of CH may lead to excessive bridging, causing the production of unstable flocs (Oyekeye et al., 2022). This instability results from excess CH particles disrupting the ideal bridging mechanism, thereby impeding proper 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 eliminate the contaminants from the synthetic wastewater (Adelodun, 2021). 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, achieving the required standard while outperforming the control solution. Thus, MOCH 3:1 provides the optimal combination for eliminating contaminants from the synthetic wastewater.

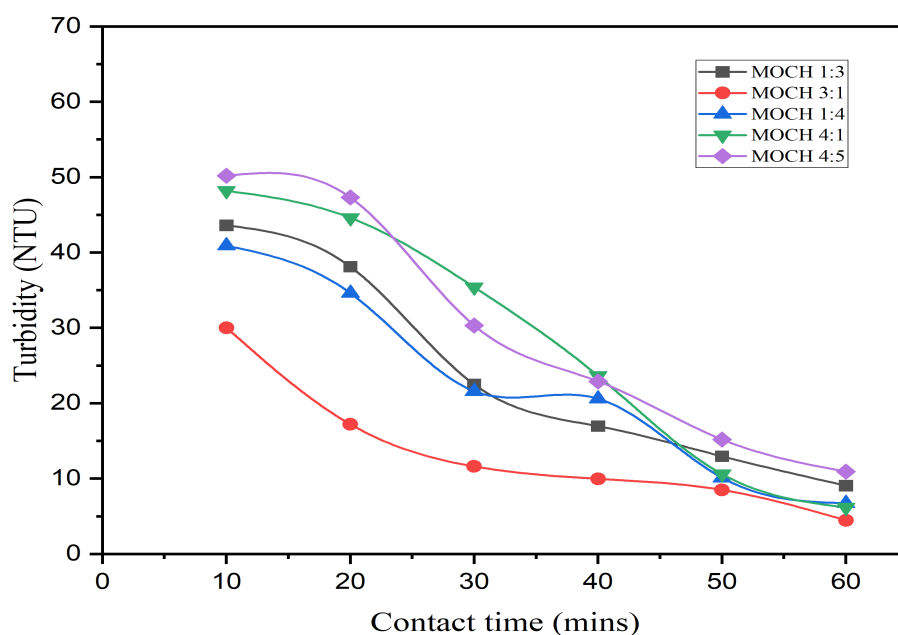
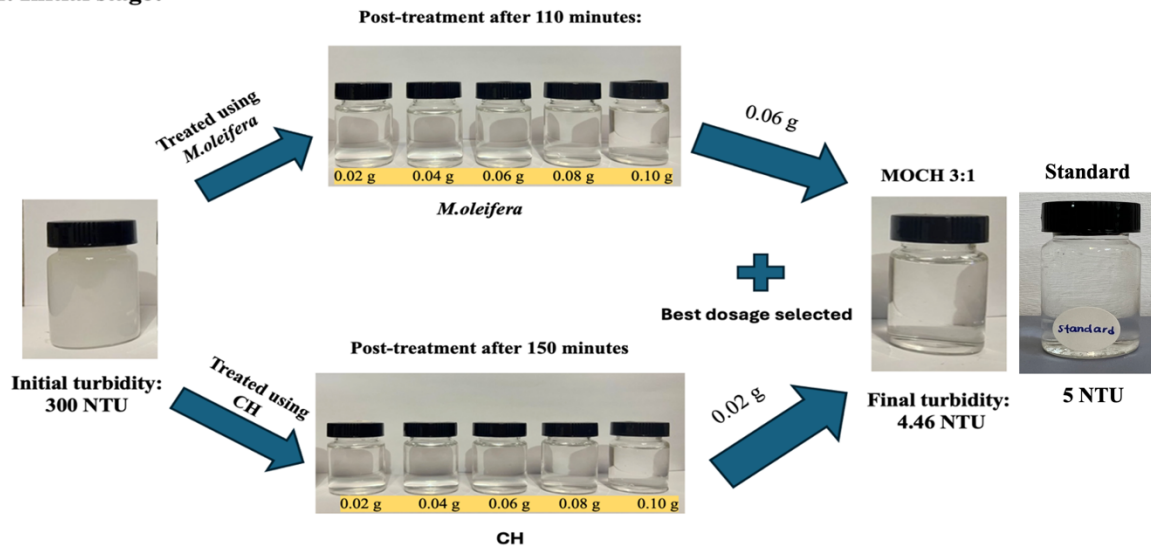


Figure 4.9: Turbidity reduction using different combined MOCH ratios at 300 NTU of synthetic wastewater

4.2.3 Turbidity removal using combined MOCH 3:1

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). Further details of this graph can be found in Appendix H1. Figure 4.10 illustrates the summary of respective coagulants in treating wastewater at different turbidity levels.

1. Initial stage:



2. Final stage:

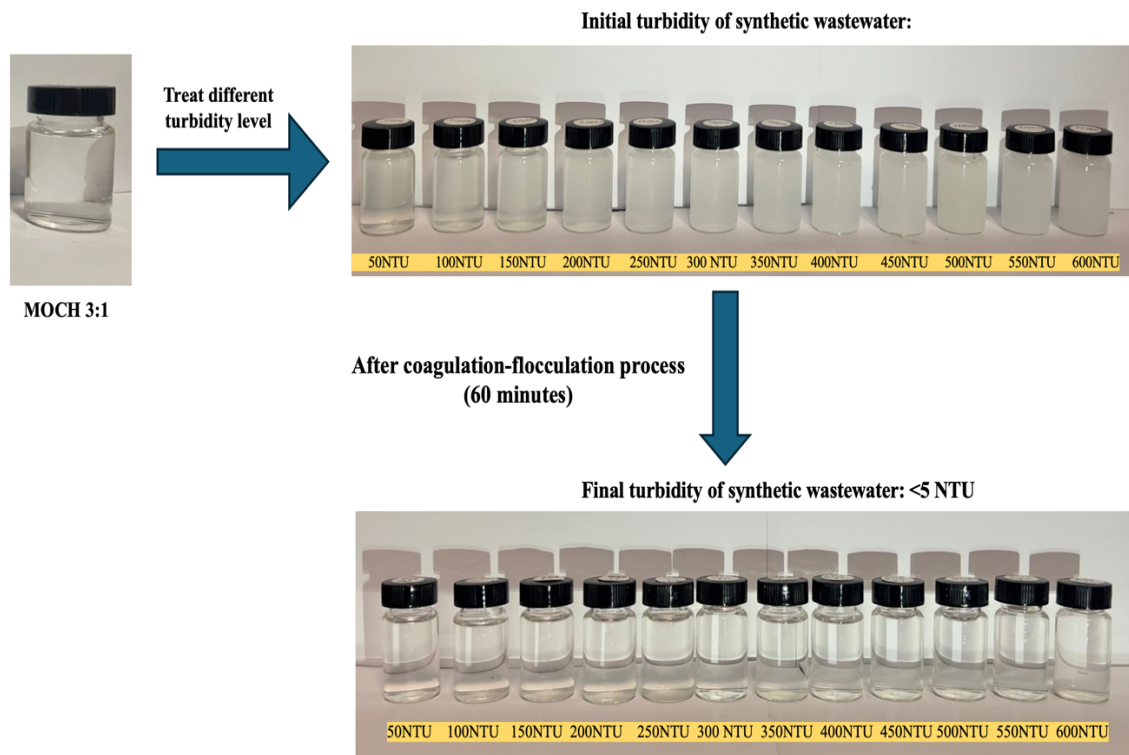


Figure 4.10: Process flowchart for using coagulants to treat synthetic effluent

The combined MOCH 3:1 can reduce low turbid wastewater rapidly after 10 minutes from 50 NTU to 3.72 NTU. Next, the combined 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 4.11. The combined 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 effective removal of low turbidity (50 NTU) owing to its capacity to efficiently neutralize charges and facilitate flocculation at comparatively low concentrations of suspended particles. At low turbidity levels, the active sites of the coagulants are sufficiently accessible, promoting efficient aggregation and elimination of particles. Other than that, the turbidity was still reduced at medium turbidity (300 NTU), however the reduction was less pronounced than that reported at 50 NTU with the application of MOCH 3:1. This may be because to the restricted coagulation ability of both coagulants at elevated turbidity levels, where the increased concentration of suspended particles could surpass the flocculation process (Shahzadi et al., 2024). Meanwhile, both coagulants exhibit lower efficacy at high turbidity levels (600 NTU), 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. Other than that, a comparable study conducted by Dandesa et al. (2023) revealed a contrasting outcome, as the use of *M.oleifera* alone was ineffective in removing turbidity from low to high levels (600 NTU), thereby failing to meet the NWSQM criteria. Moreover, Vidaurre et al. (2023) 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*. According to the study by Rasheed et al. (2023),

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. 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 (Chitra & Muruganandam, 2020; Shah et al., 2024). 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. Figure 4.12 represents the comparison between MOCH 3:1 and the best coagulant dosages from *M.oleifera* and CH. When compared to the control and standard results, MOCH 3:1 demonstrated highest turbidity removal, indicating its effectiveness in enhancing water purification. This shown that MOCH 3:1 significantly enhanced the reduction of turbidity from the synthetic wastewater compared to the use of respective coagulants.

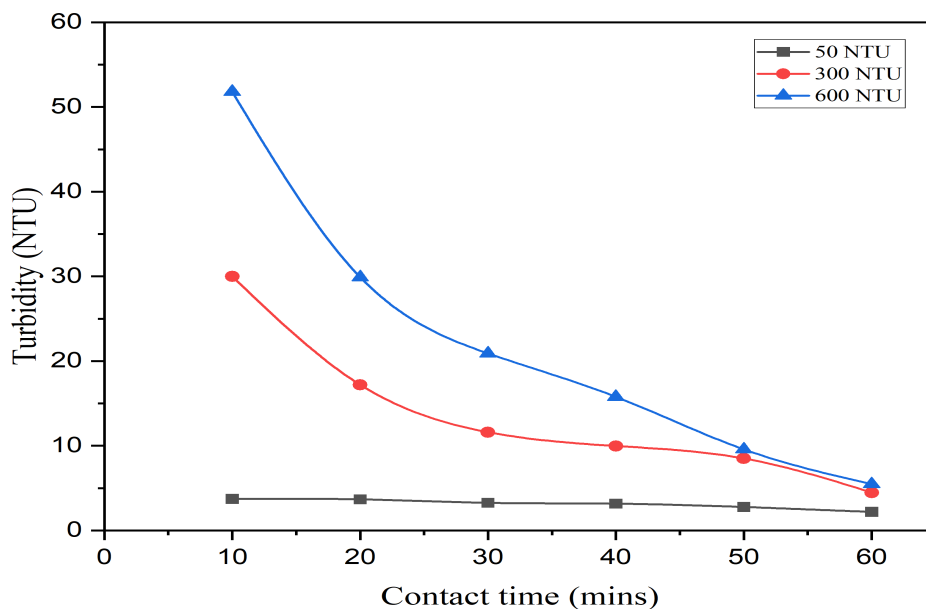


Figure 4.11: Turbidity reduction using MOCH 3:1 at different turbidity levels after 1 hour

Next, the findings of the FTIR analysis, illustrated Figure 4.1-4.3, in conjunction with the SEM surface morphology assessment depicted in Figure 4.4, as previously described, provide important information about the adsorption capacities of the coagulants and uses in water treatment due to a variety of functional groups on their surface. Furthermore, the heterogeneous surface structure offers many opportunities for particle bridging, improving flocculation and speeding up impurity sedimentation by gravitational force (Shah et al., 2024). This statement is also supported by Benettayeb et al. (2022), 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. A recent study conducted by Kitheka et al. (2022) demonstrated that the utilization of *M.oleifera* can effectively clarify water by causing suspended particulates to clump together, resulting in enhanced water clarity quickly. 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 (Gaikwad & Munavalli, 2019; Rocha et al., 2019; Taiwo et al., 2019). Meanwhile, several studies reported that a high dosage of coagulant, above 5 g, is necessary to treat high turbidity over 150 NTU effectively (Vunain et al., 2019; Nisar & Koul, 2021).

However, the dosage mentioned is significantly higher than the concentrations used in this experiment. Thus, specific studies must decrease the dosage rate to mitigate this problem. This is because a large quantity of *M.oleifera* protein may attract bacteria to the water due to its high organic content, providing a rich food source for microorganisms that promote bacterial growth and proliferation, which can eventually deplete oxygen levels,

leading to eutrophication, and some studies suggest this reduces the dosing rate (Shah et al., 2024). This method was advantageous in this study because it only required a small dosage of the biocoagulants to attain the desired turbidity level. Thus, the combined MOCH 3:1 could effectively decrease the turbidity to below 5 NTU at all types of turbidity levels within one hour when using the combined MOCH 3:1, which is faster than the study mentioned. Therefore, it was found that the combined MOCH 3:1 is better in removing turbidity with 99% compared to *M.oleifera* with another plant-based coagulant (Kasmuri et al., 2023).

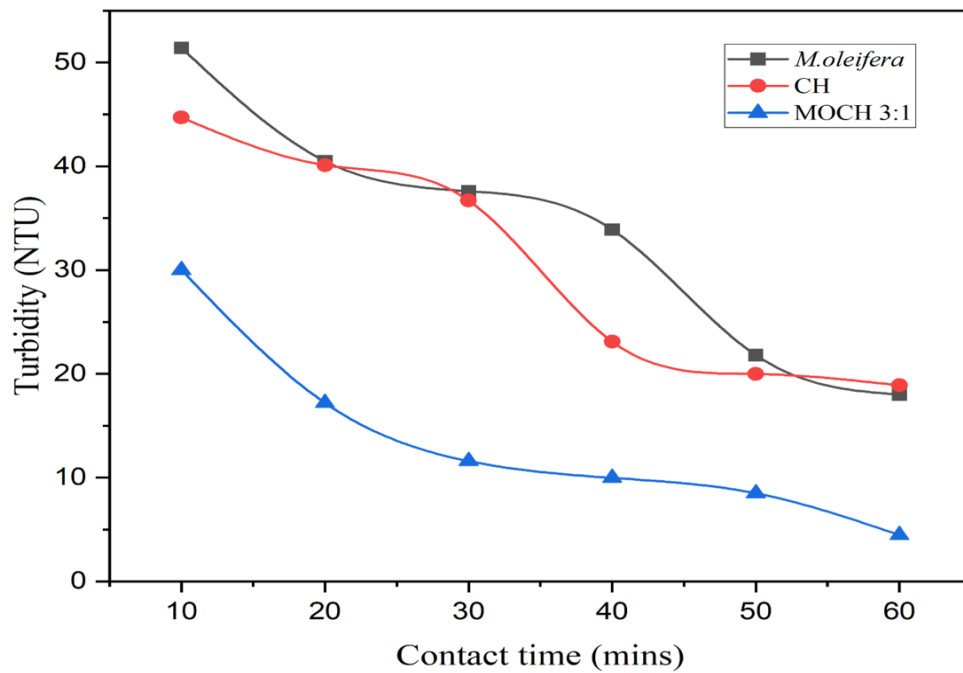


Figure 4.12: Turbidity removal using different coagulants at 300 NTU of synthetic wastewater

4.2.4 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 4.13. At 25 °C, the maximum turbidity removal was attained with MOCH 3:1 at 50 NTU, attributable to the very low initial particle concentration. This facilitates enhanced flocculation, as the coagulant efficiently neutralizes and aggregates the scattered particles. The reduced particle density enhances interaction between the coagulant and the suspended particles, resulting in effective turbidity elimination. The lowest turbidity removal using MOCH 3:1 was recorded at 600 NTU. The phenomenon can be attributed to particle overcrowding, resulting in a complex suspension that hinders the coagulant's ability to completely neutralize all particles. The increased particle density at high turbidity level likely leads to inadequate floc formation due to competition among particles for coagulant binding sites, thereby diminishing overall removal efficiency.

Overall, the optimum turbidity removal can be observed at room temperature 25 °C, where the removal rate is in accordance with the water quality standard and performs better than the control group. This result aligns with findings demonstrating that high turbidity removal can be accomplished at room temperature (Kane et al., 2016; Hadadi et al., 2022). Some studies show that higher temperatures encourage particles' instability, increasing the likelihood of colloidal particles settling and aggregating (Kurniawan et al., 2020; Pandey & Khan, 2022). In addition, 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 (Gopalakrishnan et al., 2016). On the contrary, another study by Maćczak et al. (2020) 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 (2023), which indicated that the optimal turbidity removal by 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.

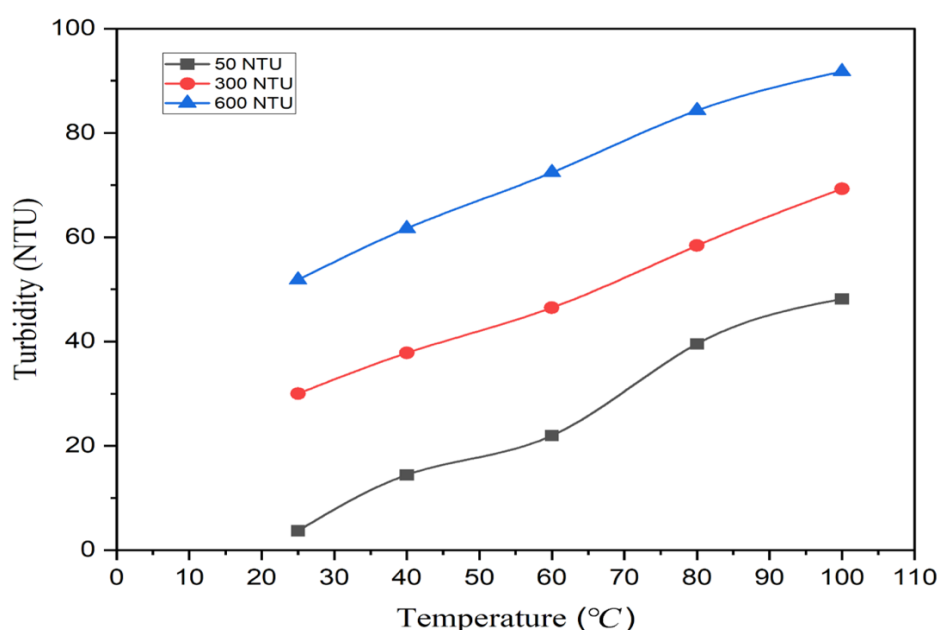


Figure 4.13: Effect of temperature on different turbidity levels removal using MOCH 3:1 after 1 hour

4.2.5 Effect of pH on turbidity removal

The selection of 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 (Adeniyi & Ighalo, 2019). A pattern of increase was observed as the pH levels were raised for 50 NTU, 300 NTU, and 600 NTU levels by using MOCH 3:1, as illustrated in Figure 4.14. The highest level of turbidity reduction was

recorded at a turbidity level of 600 NTU with a pH of 2, after 1 hour with a turbidity of 0.53 NTU. Conversely, the lowest level of turbidity removal was found at pH 10 with 7.3 NTU at turbidity level of 600 NTU. At a pH of 2, the lowest turbidity removal was recorded at a turbidity level of 50 NTU with a value of 1.21 NTU, but MOCH 3:1 was able to achieve the most significant reduction in turbidity at a pH of 10, with a value of 2.51 NTU within the same turbidity level.

The interaction between the biocoagulants and the kaolinite surface can be attributed to multiple causes, leading to improved adsorption.. At pH 2, the greatest turbidity removal efficacy was shown by MOCH 3:1 at 600 NTU. This can be ascribed to the augmented interactions between the adsorbent and kaolin particles, enabled by a higher frequency of particle-particle collisions. 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 (Faisle et al., 2023). This mechanism facilitates the enhanced adsorption of positively charged kaolin ions to the biosorbents through the electrostatic force of attraction. However, this competition diminishes as the pH increases due to the neutralization or increased negative charge of these active sites on the kaolinite surface. As a result, there is competition between the coagulants and the kaolin ions for available adsorption sites. Plus, fast collision dynamics facilitated the creation of bigger flocs, which efficiently settled, leading to a substantial decrease in turbidity. In contrast, the least turbidity removal was shown by MOCH 3:1 at 50 NTU under identical conditions. This is because the reduced particle concentration restricted collision frequency, resulting in inadequate floc formation and unsatisfactory turbidity reduction. At pH 10, the sample with 50 NTU

exhibited the greatest turbidity reduction. This enhancement is 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. The 600 NTU sample exhibited reduced removal effectiveness at pH 10. The negatively charged kaolin particles encountered heightened electrostatic repulsion with the biosorbent in alkaline conditions, impeding effective flocculation. The increased viscosity at high turbidity levels presumably diminished collision frequency, leading to weaker and smaller flocs that were less efficient in decreasing turbidity. The observed zeta potential data indicate an increase in negative charges for all biosorbents when pH rises, consistent with the findings regarding the impact of pH on turbidity. Under alkaline conditions, the increased negative surface charge amplifies electrostatic repulsion among particles, preventing their aggregation. As a result, the decreased coagulation efficiency leads to lower turbidity reduction, as the particles remain uniformly scattered in the solution.

Lulesa et al. (2022) 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 (Rahmadyanti et al., 2020). Conversely, several studies reported that the observed drop in water pH aligns with an increase in the concentration of *M.oleifera* dosage (Semanka et al., 2022; Dandesa et al., 2023). A study by Varsani et al. (2022) 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 DOE (2024) for drinking water (Kamarudin et al., 2020).

Thus, the study demonstrated that combined MOCH 3:1 had improved coagulation-flocculation by destabilising positively charged particles without requiring further pH adjustment to achieve better turbidity removal. 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.

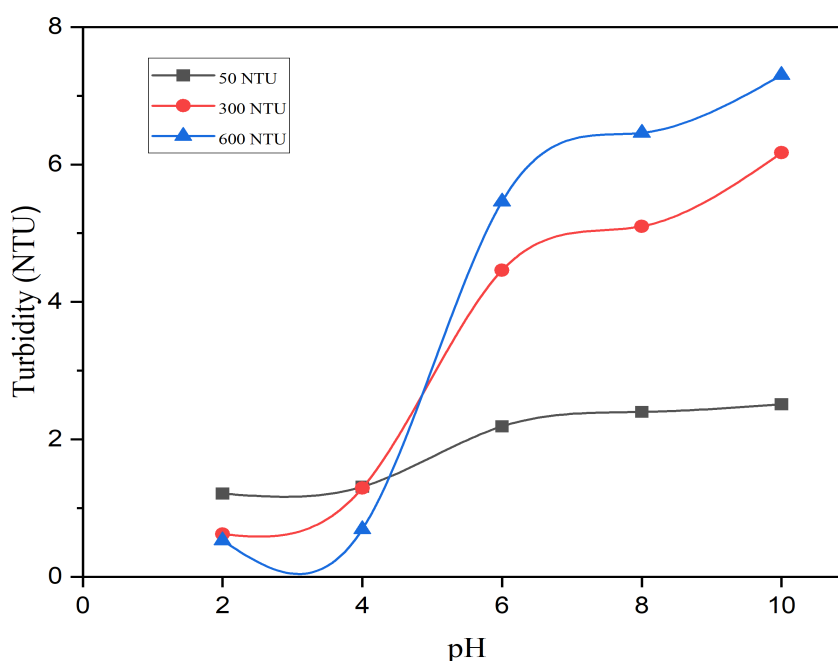


Figure 4.14: Effect of pH on different turbidity levels removal using MOCH 3:1

4.3 Total dissolved solids and total suspended solids removal

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 (Varsani et al., 2022). 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's flavour is also a crucial factor to consider. Meanwhile, TSS are the concentration of suspended particles that dissolve in water samples, which are usually quantified in milligrams per liter (mg/L) (Vlotman et al., 2024). 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 (López-Ramírez et al., 2022). The effects of TDS and TSS were often studied both before and after the treatment processes.

4.3.1 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. 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 4.15. 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. Following treatment with both coagulants, there was a drop in the TDS level at 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 TDS 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 (World Health Organisation, 2022).

Nonetheless, the increase in coagulant dosage also led to a rise in TDS levels. This observation aligns with the study by Shan et al. (2017), which documented a rise in TDS in water when the concentration of *M.oleifera* increased. Kitheka et al. (2022) found a notable rise in TDS in water as the concentration of *M.oleifera* increased. This statement is also supported by Vidaurre et al. (2023). 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 (James & Yadav, 2021). Thus, the findings are consistent with those obtained in this research.

Next, Figure 4.15 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 (Adjovu et al., 2023). It was observed that the highest reduction in TDS was 147 mg/L achieved with 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 (Bezerra et al., 2020). 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 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 (Pradeep et al., 2016). This implies that the MOCH 3:1 can effectively form larger and denser flocs thus reduce the TDS content at 300 NTU effectively compared to individual coagulant.

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. CH exhibits powerful adsorption properties, particularly in activated carbon form. It efficiently absorbs both organic and inorganic substances, aiding in the elimination of TDS (Packialakshmi et al., 2021). Thus, combining *M.oleifera* and CH can synergistically increase the removal efficiency for a wider variety of pollutants. Efficiency relies on the ideal dosage of each substance and the compatibility of their mechanisms. Incorrect dosages or unfavourable water chemistry can diminish the combined impact. The combined MOCH 3:1 showed promising coagulation activity for dissolved solids removal from water samples.

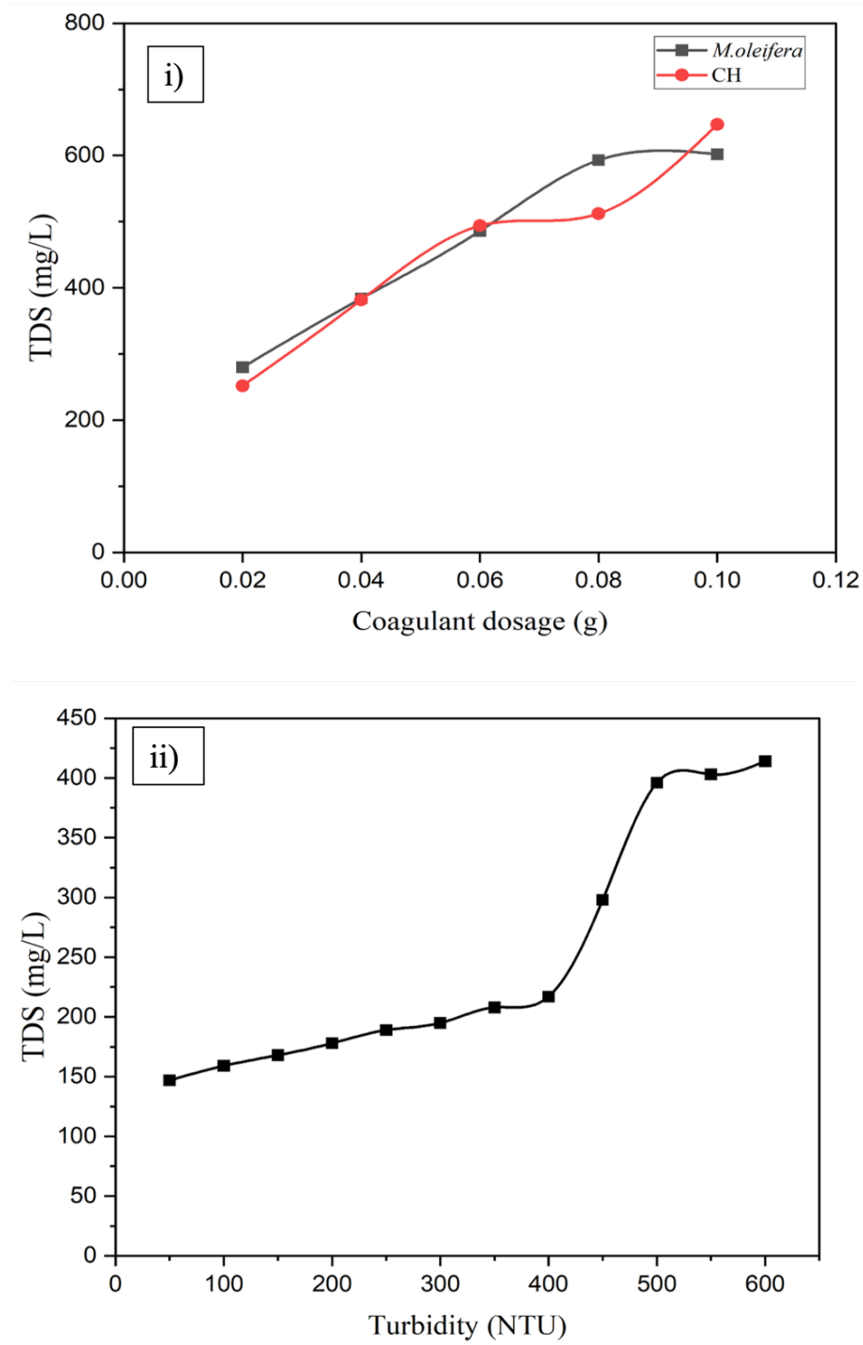


Figure 4.15: TDS removal using i) individual coagulant at 300 NTU and ii) MOCH 3:1 at different turbidity levels after 1 hour

4.3.2 Total suspended solids

The initial 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 4.16. After 1 hour, the highest TSS content recorded

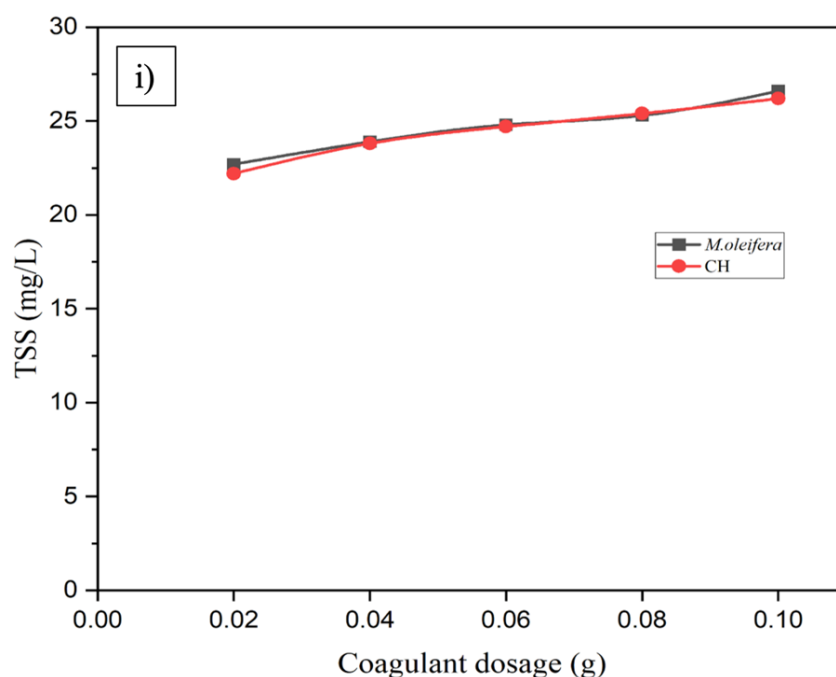
was 26 mg/L, exceeding the acceptable limit of 25 mg/L, when when employing 0.10 g of *M.oleifera* and CH, respectively. Meanwhile, the lowest TSS content in synthetic wastewater was found to be 22.7 mg/L when using 0.02 g dosage from each biocoagulant. All dosages of both biocoagulants, except 0.10 g, led to a decrease in the TSS level according to the DOE (2024) 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 total suspended solids (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 (Shahzadi et al., 2024). 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 coagulant (Olanrewaju et al., 2018). Consequently, the destabilized particles could re-stabilize due to

an inadequate number of particles available to construct additional interparticle bridges. Gaayda et al. (2022) 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 (López-Ramírez et al., 2022). Conversely, other studies also reported that a decrease in TSS was observed as the content of *M.oleifera* also increased (Olanrewaju et al., 2018; Al-Jadabi et al., 2021; Semanka et al., 2022).

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 4.16. The graph illustrates the consistent rise in TSS content as the turbidity level increases. This is because the high amount of TSS could increase the turbidity of water samples (Adjovu et al., 2023). 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 listed in Appendix C3, 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 total suspended solids (TSS) at low turbidity due to its ability to immediately interact with accessible particles, resulting in an improved flocculation process. The synergistic benefits and complementing 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 (Nasaruddin & Mohamed, 2021; Lawal et al., 2023). 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. Previous studies revealed that using individual respective of CH and *M.oleifera* only results in a low percentage of TDS and TSS removal, which is lower than 83% (Rambe et al., 2018; Oyekeye et al., 2022). Conversely, it was shown that the combined MOCH 3:1 successfully removed TDS and TSS with more than 90% 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.



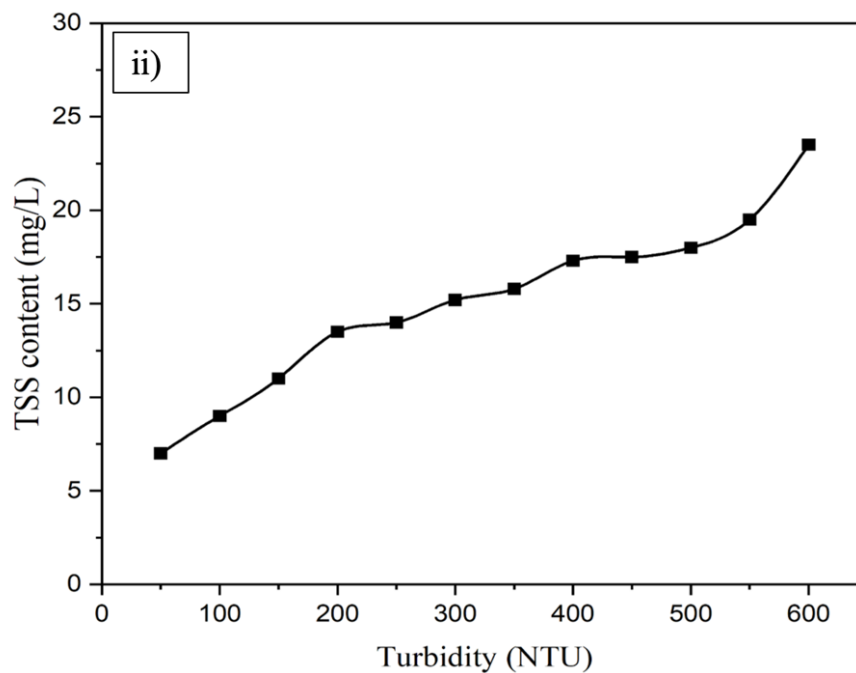


Figure 4.16: TSS removal using i) individual coagulant at 300 NTU and ii) MOCH 3:1 at different turbidity levels after 1 hour

4.4 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 (Adjovu et al., 2023). Based on DOE (2024), the value of EC below 1000 $\mu\text{S}/\text{cm}$ is acceptable, but elevated conductivity in drinking water may be undesirable for consumers because it estimates high level of dissolved ions such as heavy metals, inorganic ions and other contaminants that present in the water (Ng & Elshikh, 2021). 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.

An increasing pattern was observed with an increase in the dosage for *M.oleifera*. However, the CH showed a decrease pattern followed by a slightly increased EC value when

high dosage was used, as shown in Figure 4.17. The *M.oleifera* exhibited a notable reduction in conductivity with a coagulant dosage of 0.02 g. The water samples treated with different dosages of *M.oleifera* showed some variation from 499 $\mu\text{S}/\text{cm}$ to 519 $\mu\text{S}/\text{cm}$. Nonetheless, there was an increase in conductivity as the coagulant dosage increased. This may be linked to the release of ions such as K^+ , Ca^{2+} , Mg^{2+} , and COOH^- from *M.oleifera*. The drop in EC was noted following the application of 0.06 g, indicating that a greater number of ions are adsorbed onto the surplus *M.oleifera* particles in the solution. Next, 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. 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.

However, the EC values rose were observed when the dosage of CH increased from 0.08 g (453 $\mu\text{S}/\text{cm}$) to 0.10 g (471 $\mu\text{S}/\text{cm}$). This might have occurred due to the organic component from the CH dissolving and interacting with the existing ions, thereby increasing the ionic strength.

The high water conductivity in this study can be attributed to the elevated TDS content from adding coagulants to the wastewater (Vidaurre et al., 2023). The conductivity increased as a higher quantity of *M.oleifera* was used due to higher levels of cationic polyelectrolyte found in *M.oleifera* seeds, which resulted in more tremendous dissolved solids and thus increased conductivity (Olanrewaju et al., 2018). There is a positive correlation between the concentration of *M.oleifera* seed and the conductivity of wastewater

(Shan et al., 2017). According to Kitheka et al. (2022), 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 between conductivity and concentration in water samples treated with *M.oleifera* (Gandiwa et al., 2020). 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 (Bouchareb et al., 2021; Elsergany, 2023). Conversely, the current findings contradict the study conducted by Thorat et al. (2020), 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. According to Varsani et al. (2022), 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.

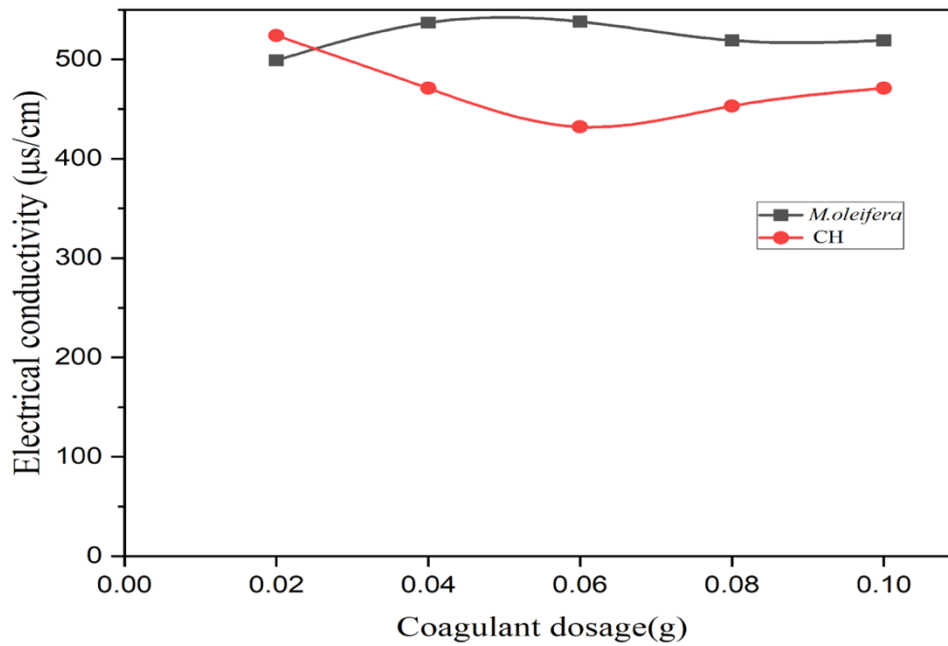


Figure 4.17: EC reduction using individual coagulant after 1 hour

The combined MOCH 3:1 was tested on the removal of EC from low to high turbidity levels. The combined MOCH 3:1 exhibited the lowest conductivity value at 300 NTU compared to the other treatments, 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. (2024), who stated that the reduction of EC was due to the coagulation-flocculation method using the proper dosage of biosorbents. 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 $\mu\text{S}/\text{cm}$), as shown in Figure 4.18. 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 an significant increase in electrical conductivity from 350 to 450 NTU due to high dissociation capacity in water from *M.oleifera* and CH (Gandiwa et al., 2020). 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 DOE (2024) standards.

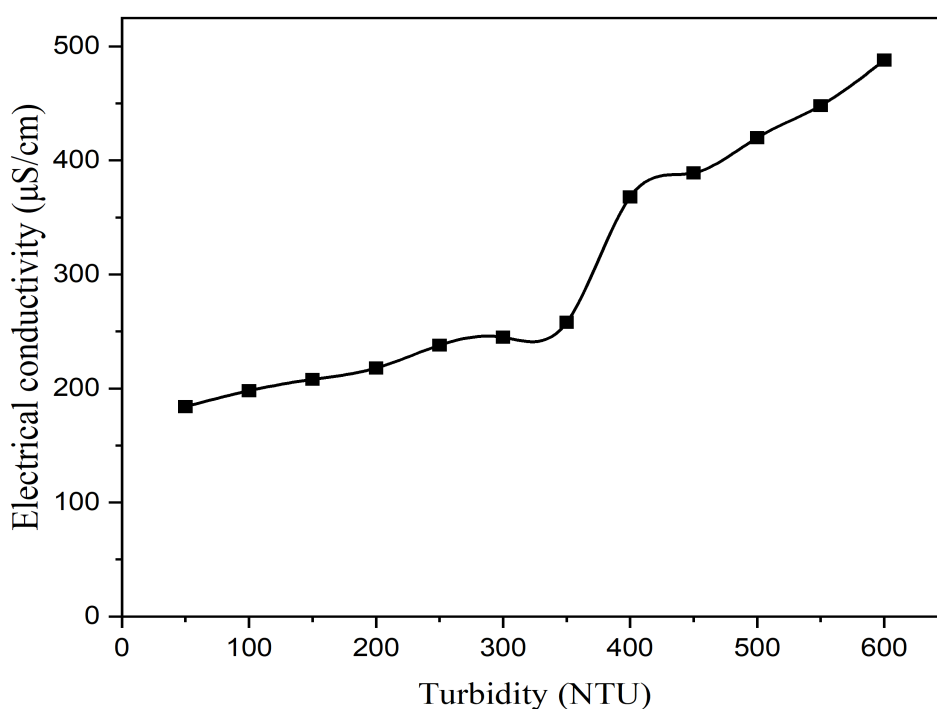


Figure 4.18: EC removal using MOCH 3:1 at different turbidity levels after 1 hour

Overall, MOCH 3:1 enhanced the elimination of turbidity, TSS, TDS, and EC within one hour, compared to the individual coagulants, from the synthetic wastewater with a initial turbidity value of 300 NTU and pH 6 at $25\ ^\circ\text{C}$, as shown in Table 4.3. 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.

Table 4.3: Summary of treating synthetic water with respective biosorbents

Type of coagulants	Total weight (g)	Turbidity (NTU)	TSS (mg/L)	TDS (mg/L)	EC (μ S/cm)	Settling time (mins)
Standard	-	5	<10	500	>400	-
Control	-	300	8758.5	2170	2440	-
<i>M.oleifera</i>	0.06	5.48	24.8	486	538	110
CH	0.02	5.18	22.2	494	524	140
MOCH 3:1	0.06 : 0.02	4.46	15.2	195	245	60

4.5 Adsorption studies on bio coagulants

Adsorption is widely used to remove different impurities from wastewater. The present work employs both kinetic and isotherm models to examine the adsorption behaviour, which is presented in the following sections. Therefore, this study will provide the efficiency and mechanisms implicated in the adsorption process using the adsorbents.

4.5.1 Langmuir and Freundlich model

An isotherm study was conducted to analyse the adsorption process of turbidity using combined MOCH 3:1. The investigation was performed on a sample of 200 mL of synthetic turbid wastewater. The initial concentration varied between 50 to 300 NTU for turbidity. The experiment was carried out under optimal conditions, as specified in Table 4.4, using combined *M.oleifera* and CH introduced into the synthetic turbid wastewater at a pH of 6.

The samples were vigorously agitated at a speed of 100 rpm for 1 minute and after that, gently mixed at a speed of 40 rpm for 10 minutes. The adsorption equilibrium was examined by employing Langmuir and Freundlich isotherms to determine the correlation between the contaminants adsorbed by combined MOCH 3:1 in the wastewater samples. The data points were subjected to fitting procedures using both the Freundlich and Langmuir models to evaluate the adsorption process.

The Langmuir model's relevance to sorption data was demonstrated by plotting C_e/q_e against C_e , resulting in straight lines, as shown in Figure 4.19. The value of the Freundlich isotherm constant is presented in Table 4.4. The analysis of the adsorption coefficient using the Langmuir isotherm revealed that the coefficient for combined MOCH 3:1 ($R^2 = 0.99997$) was higher than the coefficients for the Freundlich isotherms ($R^2 = 0.99179$). The Langmuir isotherm constant is listed in Table 4.4. The values of $Q_{\max}$ and K_L were determined by analysing the slope and intercept of the plots of C_e/q_e vs C_e , which demonstrate the suitability of the Langmuir isotherm for sorption data. The maximum adsorption capacity ($Q_{\max}$) of combined MOCH for removing turbidity from wastewater samples was 1226.5 mg/L. The coefficient of determination (R^2) for the adsorption of turbidity by combined MOCH 3:1 exceeded 0.90. The variable C_0 (mg/L) represents the initial concentration of the turbidity, while K_L (L/mg) is the Langmuir constant associated with the energy of adsorption. The R_L number reveals the shape of the isotherms, which can be classified as either unfavourable ($R_L > 1$), linear ($R_L = 1$), favourable ($0 < R_L < 1$), or irreversible ($R_L = 0$) (Gomes et al., 2022). The computed R_L value for combined MOCH 3:1 is 0.03167 with an R^2 of 0.99997, indicating that the interaction process between kaolin and MOCH 3:1 is favourable.

The relationship between the natural logarithm of q_e and the natural logarithm of C_e resulted in a linear relationship of the Freundlich model, as shown in Figure 4.19. The findings are presented in Table 4.4. The favourable adsorption of this model can be characterized by the condition that if the value of n is greater than one, the adsorption is considered favourable and is a physical process (Alghamdi et al., 2024). The current investigation found that the value of $(1/n = -0.0257)$ is less than 1, suggesting that the adsorption process is unfavourable. The values of n and K_f (g/L) were estimated from the slope and intercept plots $\text{Log } Q_e$ versus $\text{Log } C_e$. The K_f values for the adsorption capacity of combined MOCH 3:1 was 0.274878 for turbidity. Nonetheless, this study revealed that the K_L value is greater than the K_f value, implying that Langmuir adsorption isotherm is the best model for removing turbidity from synthetic wastewater. In addition, the X^2 value of the Langmuir model is 7.129×10^{-8} , is smaller than the X^2 value of the Freundlich model, 4.359, demonstrating an ideal alignment of the experimental data with this model. Plus, the Langmuir model exhibited a strong correlation with a higher regression value of 0.99997 on turbidity removal compared to the other, which suggests that the adsorption process follows a monolayer adsorption mechanism on a surface with a limited number of identical sites (Raji et al., 2022).

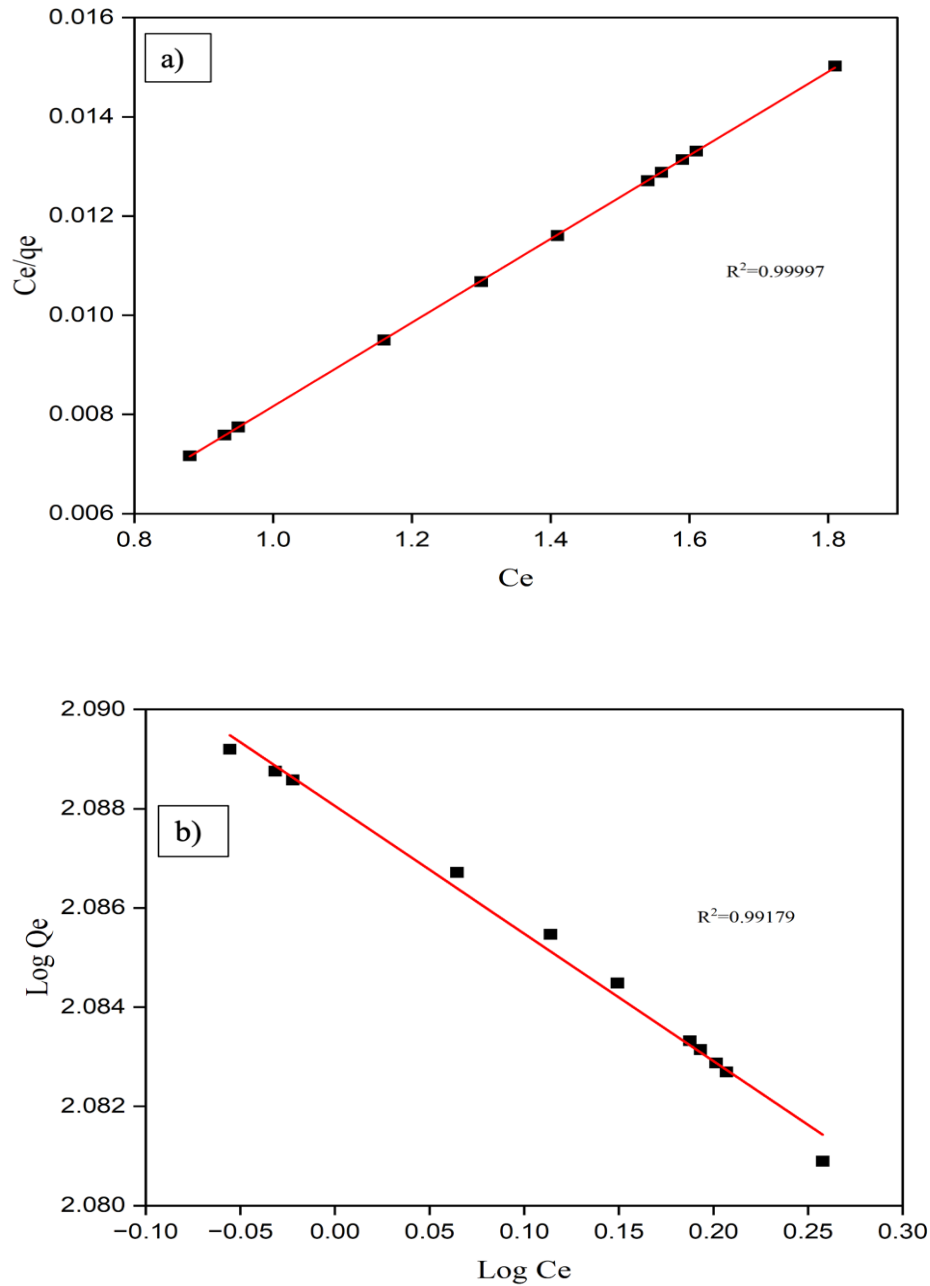


Figure 4.19: The (a) Langmuir (b) Freundlich isotherm models for turbidity adsorption using MOCH 3:1

Most studies concurred that the adsorption data conformed to the Langmuir model and is considered the most suitable isothermal adsorption model for describing the adsorption process employing *M. oleifera* with $0.91 < R^2 < 0.984$ compared to the Freundlich model for removing turbidity from wastewater samples (Rosmawanie et al., 2018; Praveena &

Shamsudin, 2020). The findings in this study suggest that the utilisation of a combination of *M.oleifera* and CH demonstrated a superior capacity for removing turbidity compared to individual coagulants because the coefficient of the R^2 value was higher than in the previous study, as stated previously. In contrast, Rajalingam et al. (2021) reported that equilibrium data was best presented by the Freundlich model with R^2 values of 0.9226, which implies that the adsorption occurs on a heterogeneous surface. Therefore, the results of this study indicate that the utilisation of MOCH 3:1 with an adsorption capacity of 1226.5 mg/g at 298 K exhibits significant potential as a sustainable and economically viable adsorbent for diverse water treatment purposes, particularly in the elimination of pollutants and undesirable substances from wastewater.

Table 4.4: Langmuir and Freundlich isotherm parameters for combined MOCH 3:1

Model isotherms	Parameters	Values
Langmuir	$Q_{\max}$	1226.5
	K_L	0.38705
	R_L	0.03167
	R^2	0.99997
	X^2	7.129×10^{-8}
Freundlich	K_f	0.274878
	$1/n$	-0.0257
	R^2	0.99179
	X^2	4.359

4.5.2 Pseudo-first-order and pseudo-second-order kinetic model

Kinetic models play a crucial role in assessing the fundamental characteristics of an adsorbent, such as the time needed for the removal of specific impurities, the efficiency of the adsorbent, and the identification of the mechanisms involved in an adsorption system (Raji et al., 2023). Typically, in practical applications, kinetic studies are conducted using batch experiments that involve adjusting the adsorbate concentration, adsorbent mass, agitation time, temperature, and the kind of adsorbent and adsorbate. In this study, the MOCH 3:1 was utilised to remove the turbidity of synthetic wastewater with 50 NTU at room temperature. Afterwards, the data is analysed and utilized in the linear regression to ascertain the kinetics model that offers the most accurate alignment. Table 4.5 displays the computed values of Q_e , rate constant (K), and correlation coefficient (R^2). For most pseudo-second-order kinetics models, the predicted Q_e values closely approximate the experimental q_e values. However, to determine the accuracy of the adsorption process, two criteria need to be assessed. The first criterion is based on the regression coefficient (R^2), and the second on the calculated Q_e values (Ahmed et al., 2023).

Table 4.5: Kinetic isotherms parameters

Parameters	Pseudo-first-order kinetic	Pseudo-second-order kinetic
Intercept	1.88312	0.00968
Slope	-0.01884	0.0082
Q_e (mg/g)	6.57	121.95
K	0.01884	0.847107
R^2	0.58648	0.99958
X^2	3.5461	9.37024×10^{-5}

The primary models employed to assess the kinetics model profile are the pseudo-first-order and pseudo-second-order kinetic models. Additionally, the correlation coefficients are near 1, showing that the pseudo-second model effectively describes the kinetics of the kaolin ion adsorption process. The biosorption of kaolin ion might occur through complexation, coordination, and chelation, which do not involve mass transfer in the solution, and chemisorption involving valence forces through the sharing or exchange of electrons with the adsorbate (Raji et al., 2022). The regression value for the pseudo-first-order kinetic model is 0.56848, indicating that the model cannot accurately capture the kinetics of the kaolin particle adsorption processes. Figure 4.20 illustrates the adsorption of turbidity using MOCH 3:1 according to the prescribed kinetic models. The adsorption kinetics adhere to a pseudo-second-order kinetic model with a regression value of 0.99958, suggesting that the adsorption process is governed by chemisorption and the formation of a monolayer on the adsorbent surface (Masekela et al., 2022). Therefore, the adsorption kinetics followed the pseudo-second-order model and strongly correlated with the experimental data ($X^2 = 0.0000937$). The Q_e value obtained from the pseudo-second order kinetic model (121.95 mg/g) is significantly higher than the value of pseudo-first order kinetic model (6.57 mg/g). This suggests that the pseudo-second-order model offers a more suitable representation of the adsorption capacity. Next, a greater K value of 0.847107 for the pseudo-second-order kinetic model indicates that this model more accurately characterizes the adsorption process. Similar findings also agreed that the pseudo-second-order kinetic model fitted the data well (James & Yadav, 2021; Shouzheng et al., 2021). Based on the articles, it was found that only Varsani et al. (2022) and Rasheed et al. (2023) exclusively utilized the pseudo-first-order kinetic model to analyse the collected data and

show the best fit for the experimental data but the correlation values obtained were low compared to this study.

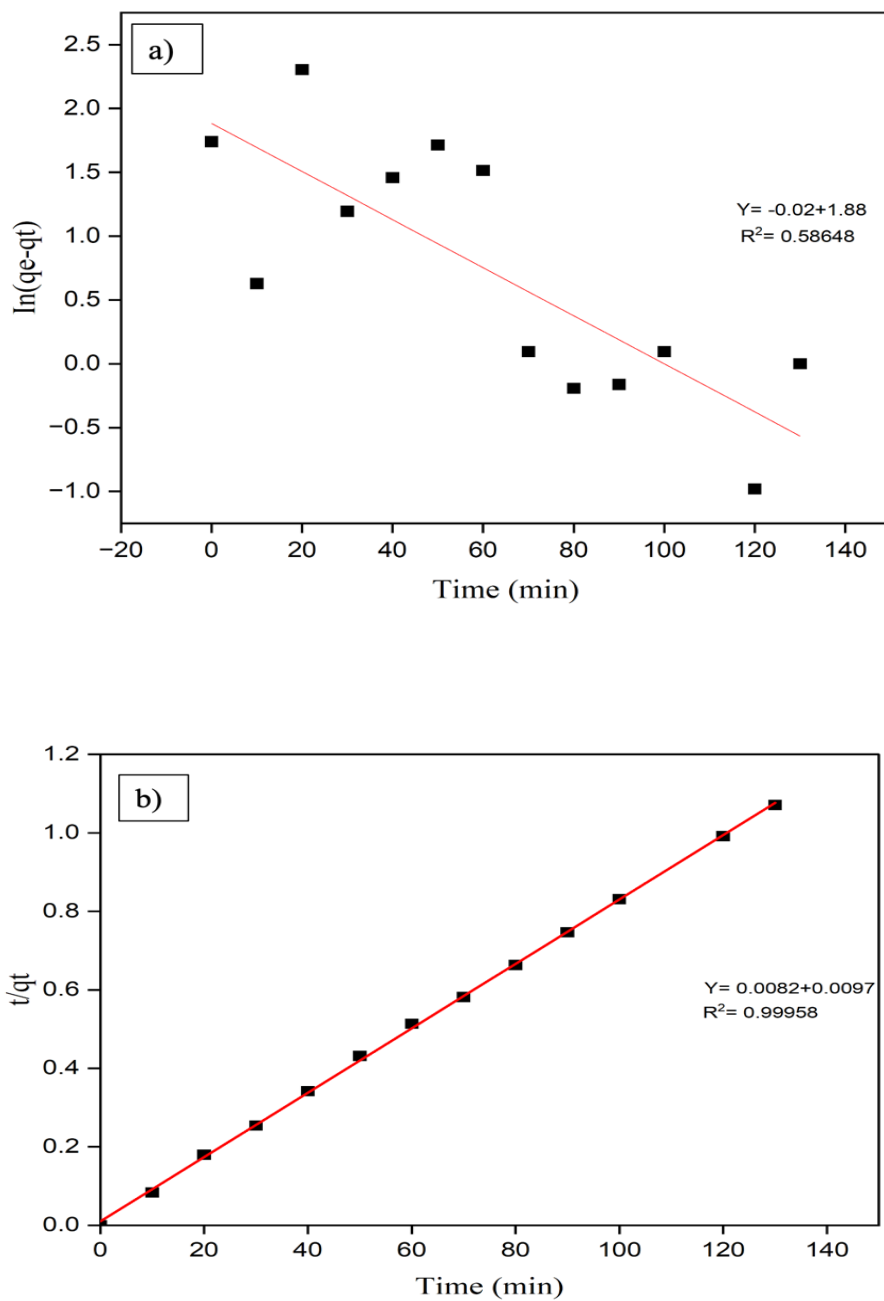


Figure 4.20: Kinetic graphs of (a) Pseudo-first-order and (b) Pseudo-second-order for turbidity adsorption by MOCH 3:1

4.6 Comparative analysis of several combination coagulants with *M.oleifera*

The use of *M. oleifera* as a natural coagulant has resulted in growing interest among researchers in its applications as a sustainable water treatment method. Nevertheless, the usage of individual *M.oleifera* is insufficient to efficiently eliminate a turbid TDS, TSS, and EC from the wastewater. Modifications have been implemented using this approach, using either alum or other plant materials as highlighted in Table 4.6. Multiple studies have documented that the use of alum and *M.oleifera* has resulted in a removal efficiency of less than 86% for turbidity and TSS within one hour at a pH of 6 (Delelegn et al., 2018; Elemile et al., 2021). The adsorption process utilising alum and *M.oleifera* for the removal of colloidal particles from synthetic wastewater was found to be consistent with the pseudo-first-order kinetic model, indicating that the process is controlled by the physisorption process (Rasheed et al., 2023). A distinct study conducted by Gandiwa et al. (2020) demonstrated that the use of *M.oleifera* in conjunction with cactus opuntia and alum may effectively remove 91.5% of turbidity and 308.2 $\mu\text{S}/\text{cm}$ of EC from wastewater at a pH level of 6. Nevertheless, the usage of alum in combination with *M.oleifera* remains undesirable due to the potential contribution of alum to sludge toxicity (Kurniawan et al., 2020).

Hence, several researchers have identified alternate options by substituting the use of alum with other plant materials, which could enhance the efficiency of *M.oleifera* as biosorbents for water purification applications. A finding by Kasmuri et al. (2023) discovered that the application of *M.oleifera* in conjunction with maize seed leads to a significant 92% decrease in turbidity and a complete elimination of TSS at 100% from the wastewater effluent. In addition, Kenea et al. (2023) demonstrated that the combination of aloe vera with *M.oleifera* exhibited turbidity and TDS removal rates of 88% and 92%,

respectively, at a pH of 8. The combination of *M.oleifera* and *Cicer arietinum* achieved optimal turbidity and TSS removal of 86.24% and 87.7% at pH 8, respectively, along with the adsorptive capacity of this adsorbent effectively characterises the experimental data obtained from the adsorption experiments using the Langmuir model (Rosmawanie et al., 2018). However, the most recent results of this study showed the greatest performance in removing turbidity, TDS, TSS, and EC from synthetic wastewater compared to earlier studies. The experimental data from this study agreed with Langmuir and pseudo-second-order kinetic model, which describes the monolayer adsorption and chemisorption process. Overall, MOCH 3:1 have the potential to serve as an excellent adsorbent for efficient wastewater treatment.

Table 4.6: Combination of *M.oleifera* with other materials to remediate wastewater

Types of combination	Turbidity removal (%)	TDS removal (%)	TSS removal (%)	EC removal ($\mu\text{S}/\text{cm}$)	Settling time (min)	Adsorption kinetic model	pH	References
Combination of alum and <i>M.oleifera</i>	89.06	-	86.42	-	60	Pseudo-first-order kinetic model	6.2	(Delelegn et al., 2018; Elemile et al., 2021; Rasheed et al., 2023)
Combination of <i>M.oleifera</i> and maize seeds	92.00	-	100	-	-	-	-	(Kasmuri et al., 2023)
Combination of aloe vera plants with <i>M.oleifera</i>	88.5	92.1	-	-	-	-	8.4	(Kenea et al., 2023)
Combination of <i>M.oleifera</i> with <i>cactus opuntia</i> -alum blend	91.50	-	-	308.2	-	-	6.5	(Gandiwa et al., 2020b)
Combination of <i>M.oleifera</i> and <i>Cicer arietinum</i>	86.24	-	87.7	-	60	Langmuir isotherms	8	(Rosmawanie et al., 2018)
Combination of <i>M.oleifera</i> and CH (MOCH 3:1)	99.46	92	99.88	245	60	Langmuir isotherms and pseudo-second-order kinetic model	6	Recent study

4.6.1 Adsorption mechanism

The adsorption process was characterised using a combination of analytical techniques that include FTIR spectroscopy, SEM analysis, and zeta potential measurements. These methodologies, in conjunction with kinetic modelling using the pseudo-second-order and fitting equilibrium data to the Langmuir isotherm, offer significant evidence supporting the dominance of an electrostatic contact mechanism in the kaolin-adsorbents surface behaviour. Figure 4.21 illustrates the possible mechanisms that encompasses all biosorbents designed to remove kaolin particles from the synthetic wastewater.

The presence of hydroxyl (OH), carbonyl (C=O), and carboxyl groups (COOH) in all biosorbents allows them to come into contact with kaolin particles in synthetic wastewater and aggregate the particles, resulting in a flocculation process followed by sedimentation (Darshan et al., 2020). The finding is further substantiated by the SEM findings, which demonstrate a smooth surface after the coagulation-flocculation process, which indicates the presence of kaolin particles adsorbed on the surface of adsorbents (Rosmawanie et al., 2018). Other than that, all biosorbents exhibit a negative charge on their surfaces across all pH levels, as seen by the zeta potential results, suggesting that electrostatic attraction occurs upon contact with surfaces or particles carrying opposing charges (Militao et al., 2022). This study revealed that strong repulsion between biosorbents and colloidal particles occur at pH 10, whereas less repulsion is observed at pH 2, indicating that electrostatic attraction is likely to occur. Although the Langmuir isotherm shows monolayer adsorption, compatible with electrostatic interaction between the surface of biosorbents and kaolin from the synthetic wastewater, the pseudo-second-order kinetics suggest a chemisorption mechanism involving electron sharing or transfer (Raji et al., 2023). This is supported by FTIR findings, as the shifts in peak positions and changes in intensity suggest that interactions between the coagulants and particles have occurred, resulting in hydrogen bonding and complexation (Mariana et al., 2019; Benettayeb et al., 2022).

In summary, the findings from these methods demonstrate that the adsorption process is predominantly governed by hydrogen bonding, complexation, and electrostatic attraction between the charged particles and the surface of biosorbents. Additionally, chemical interactions play a secondary role, as indicated by the pseudo-second-order kinetics

and FTIR results. This integrated methodology provides a thorough comprehension of the adsorption mechanism and establishes a strong basis for future studies aimed at optimising the use of MOCH 3:1 as an adsorbent.

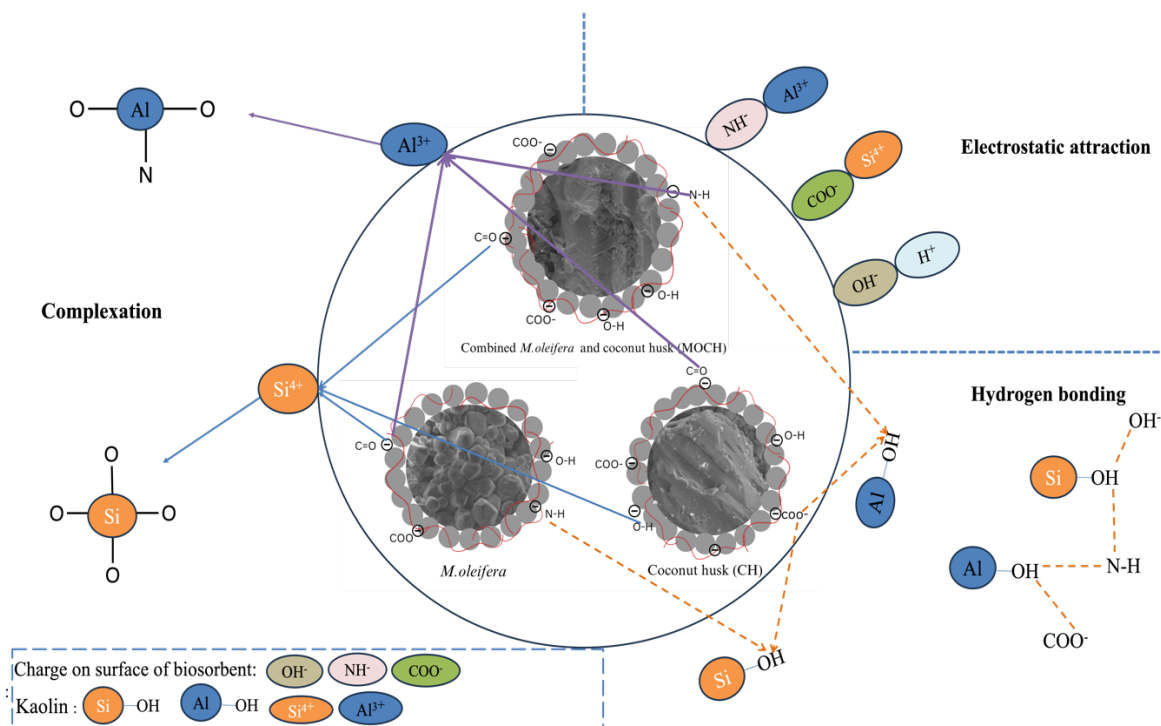


Figure 4.21: Possible mechanisms of interaction between biosorbents and kaolin

CHAPTER 5

CONCLUSION AND RECOMMENDATIONS

5.1 Conclusion

In conclusion, the effect of dosage on the surface characteristics of MOCH 3:1 and individual *M.oleifera* with CH 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 electrostatic interaction. 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. 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 $\mu\text{S}/\text{cm}$ was shown by MOCH 3:1. Therefore, it was demonstrated that MOCH 3:1 exhibited the highest removal of turbidity, TDS, TSS, and EC from synthetic wastewater. The adsorption of kaolin particles by MOCH 3:1 followed the Langmuir isotherm, and pseudo-second-order kinetic model.

Therefore, the results of this research establish a basis for improving the efficiency of *M.oleifera* by incorporating CH to eliminate turbidity, TDS, TSS, and EC from wastewater.

5.2 Recommendations

The results of this study suggest numerous sectors for further investigation and improvement to improve the comprehension and practicality of the adsorption process.

1. Analysis of actual wastewater samples

Although this study employed synthetic wastewater, it is crucial to evaluate the performance of the MOCH 3:1 in actual wastewater. It is imperative to expand this study to assess the efficacy of the adsorbent in eliminating heavy metals, leachates, textile dyes, fertiliser, and pharmaceutical wastes from real wastewater. Validating the adsorbent's efficacy would confirm its effectiveness and offer valuable insights into its suitability for treating contaminated water sources, which are frequently tainted with highly toxic heavy metals and nutrients.

2. Study on the reusability of MOCH 3:1 as adsorbent

This project became complicated by the need for an additional filtration process to remove remaining contaminants from the treated solution. While this phase was necessary to ensure product quality, it could contaminate treated water if the filtration process was inefficient. This complexity contributed variability and uncertainty to the treatment procedure, reducing findings reliability. Hence, future research should prioritise the investigation of the regeneration and reusability of MOCH 3:1 as an adsorbent to remove contaminants without necessitating a subsequent filtration step. It is necessary to determine

the appropriate number of regeneration cycles before performance deteriorates to comprehend its long-term sustainability.

3. Assessing the scalability of the adsorption process

The application of MOCH 3:1 could be used in high volumes of wastewater and pilot-scale tests to evaluate its practicality, economic viability, and long-term stability. This could advance the use of MOCH 3:1 as an adsorbent in large-scale water treatment systems.

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APPENDICES

APPENDIX A: Turbidity removal

APPENDIX A1: Turbidity removal using *M.oleifera*

Coagulant dosage for <i>M.oleifera</i> (g)	Contact time (mins)											
	10	20	30	40	50	60	70	80	90	100	110	120
0.02	58.5	51.5	49.3	45.7	43.2	43	38.84	28.48	18.19	7.57	6.4	5.45
0.04	56.2	49.3	46.5	44.2	42.7	40.4	37.79	27.39	17.24	7.82	6.3	5.88
0.06	51.4	40.5	37.6	33.9	21.8	18	16.65	13.49	10.4	6.89	5.48	5.18
0.08	53.7	50.7	45.8	44.9	41.5	40.1	37.08	26.64	17.04	7.52	5.68	5.3
0.10	55.6	51	47.6	42.1	41.9	41.4	38.37	27.94	17.84	7.68	6.31	5.56

APPENDIX A2: Turbidity removal using CH

Coagulant dosage for CH (g)	Contact time (mins)														
	10	20	30	40	50	60	70	80	90	100	110	120	130	140	150
0.02	44.7	40.1	36.7	23.1	20	18.9	16.4	14.8	12	10.6	9.48	8.6	7.29	6.28	5.18
0.04	46.4	42.2	37.6	25.3	23.3	20.8	17.1	16.4	15.7	13.6	10.27	9.1	7.38	6.48	5.62
0.06	47	44.1	38.7	36.4	34.9	20.3	18.2	17.6	16.4	15	10.61	9.14	8.26	7.51	5.76
0.08	50	46	40.1	38.3	35.8	25.7	20.1	18	17.1	16	11.75	10.3	9.23	8.09	5.86
0.10	52.6	48	45.9	40.3	38.8	30.3	25.8	20.5	19.4	18	17.6	11.3	10.4	8.91	5.94

APPENDIX A3: Turbidity removal using different MOCH

MOCH ratio	Contact time (mins)					
	10	20	30	40	50	60
1:3	43.6	38.1	22.49	16.96	12.96	9.06
3:1	30	17.2	11.6	9.97	8.5	4.46
1:4	40.9	34.6	21.6	20.6	10.1	6.74
4:1	48.2	44.6	35.4	23.64	10.6	6.18
4:5	50.2	47.3	30.3	22.9	15.2	10.92

APPENDIX A4: Turbidity removal using MOCH 3:1

Time/ Turbidity	50 NTU	100 NTU	150 NTU	200 NTU	250 NTU	300 NTU	350 NTU	400 NTU	450 NTU	500 NTU	550 NTU	600 NTU
10	3.72	22	43	45	57.9	30	47.4	49.7	67.1	64	64	51.8
20	3.66	20.3	34.8	31.2	45.1	17.2	40	41.4	52.2	51.5	50.9	29.9
30	3.26	20.2	26.8	29.4	29.4	11.6	37	31.2	29.3	35.9	36.3	20.9
40	3.16	17.7	15.9	14.8	14.5	9.97	7.32	5.88	10.1	18.8	16.8	15.8
50	2.76	17.3	8.89	7.6	7.16	8.5	6.97	5.62	7.35	8.06	9.8	9.57
60	2.19	5.69	4.74	4.55	5.58	4.46	5.85	5.19	5.6	5.65	5.5	5.46
70	1.88	3.87	4.68	4.39	5.33	4.37	5.2	5.14	4.94	4.19	4.79	5.42
80	1.88	3.84	4.58	4.31	4.26	3.37	4.49	4.95	4.36	4.12	4.02	2.84
90	1.78	3.79	4.17	4.27	4.17	3.04	3.99	4.9	4.09	3.38	3.84	2.75
100	1.77	2.83	3.36	2.38	3.08	2.47	3.97	4.86	3.74	3.32	3.09	2.7
120	1.59	2.25	2.3	2.3	3.03	2.07	2.93	2.79	2.48	3.03	3.05	2.69
130	1.44	1.92	1.41	2.22	2.56	1.61	2.3	2.33	2.29	2.97	3.03	2.68
140	0.95	1.81	1.25	1.59	1.16	1.61	0.93	0.88	0.95	1.54	1.56	1.3

APPENDIX A5: Turbidity removal using MOCH 3:1 at 50 NTU, 300 NTU & 600 NTU

MOCH 3:1 / Turbidity	50NTU	300NTU	600NTU
10	3.72	30	51.8
20	3.66	17.2	29.9
30	3.26	11.6	20.9
40	3.16	9.97	15.78
50	2.76	8.5	9.57
60	2.19	4.46	5.46
70	1.88	4.37	5.42
80	1.88	3.37	2.84
90	1.78	3.04	2.75
100	1.77	2.47	2.7
120	1.59	2.07	2.69
130	1.44	1.61	2.68
140	0.95	1.61	1.3

APPENDIX A6: Effect of temperature on turbidity removal using MOCH 3:1 at 50 NTU, 300 NTU & 600 NTU

Turbidity/Temperature °C	25	40	60	80	100
50NTU	3.72	14.43	21.97	39.57	48.16
300NTU	30	37.8	46.5	58.4	69.3
600NTU	51.8	61.7	72.4	84.3	91.8

APPENDIX A7: Effect of pH on turbidity removal using MOCH 3:1 at 50 NTU, 300 NTU & 600 NTU after 1 hour

Turbidity/pH	2	4	6	8	10
50 NTU	1.21	1.31	2.19	2.4	2.51
300 NTU	0.62	1.29	4.46	5.1	6.17
600 NTU	0.53	0.69	5.46	6.46	7.3

APPENDIX B: Total dissolved removals**APPENDIX B1: Total dissolved removals using individual coagulant**

Coagulant (g)	TDS content (mg/L)	
	<i>M.oleifera</i>	CH
0.02	280	252
0.04	384	382
0.06	486	494
0.08	593	512
0.10	602	647

APPENDIX B2: Total dissolved removals using different combination of MOCH

Different ratio	MOCH 1:3	MOCH 3:1	MOCH 4:5	MOCH 1:4	MOCH 4:1
TDS content (mg/L)	257	195	235	232	231

APPENDIX B3: Total dissolved removals using MOCH 3:1

Turbidity (NTU)	TDS content (mg/L)
50	147
100	159
150	168
200	178
250	189
300	195
350	208
400	217
450	298
500	396
550	403
600	414

Appendix C: Total suspended solids removals

APPENDIX C1: Total suspended solids reduction using individual coagulant

Coagulant (g)	TSS content (mg/L)	
	<i>M.oleifera</i>	CH
0.02	22.7	22.2
0.04	23.9	23.8
0.06	24.8	24.7
0.08	25.3	25.4
0.10	26.6	26.2

APPENDIX C2: Total suspended solids reduction using different MOCH ratio

Different ratio	MOCH 1:3	MOCH 3:1	MOCH 4:5	MOCH 1:4	MOCH 4:1
TSS content (mg/L)	23	15.2	30	26.7	24.5

APPENDIX C3: Total suspended solids reduction using MOCH 3:1

Turbidity (NTU)	TSS content (mg/L)
50	7
100	9
150	11
200	13.5
250	14
300	15.2
350	15.8
400	17.3
450	17.5
500	18
550	19.5
600	23.5

APPENDIX D: Electrical conductivity removal

APPENDIX D1: Electrical conductivity removals using individual coagulant

Coagulant (g)	Electrical conductivity ($\mu\text{S}/\text{cm}$)	
	<i>M.oleifera</i>	CH
0.02	499	524
0.04	537	471
0.06	538	432
0.08	519	453
0.10	519	471

APPENDIX D2: Electrical conductivity removals different combined MOCH

Different MOCH ratio	MOCH 1:3	MOCH 3:1	MOCH 4:5	MOCH 1:4	MOCH 4:1
Electrical conductivity ($\mu\text{S}/\text{cm}$)	451	245	470	472	471

APPENDIX D3: Electrical conductivity removals using MOCH 3:1

Turbidity (NTU)	Electrical conductivity ($\mu\text{S}/\text{cm}$)
50	184
100	198
150	208
200	218
250	238
300	245
350	258
400	368
450	389
500	420
550	448
600	488

APPENDIX E: Isotherm Model

APPENDIX E1: Langmuir Isotherm Model

Experiment no	Ci(mg/L)	Ce(mg/L)	qe(mg/g)	1/Ce	1/qe	Ce/qe
1	50	0.95	122.625	1.05263158	0.00815494	0.0077472
2	100	1.81	120.475	0.55248619	0.00830048	0.01502386
3	150	1.41	121.475	0.70921986	0.00823215	0.01160733
4	200	1.59	121.025	0.62893082	0.00826276	0.01313778
5	250	1.16	122.1	0.86206897	0.00819001	0.00950041
6	300	1.61	120.975	0.62111801	0.00826617	0.01330853
7	350	0.93	122.675	1.07526882	0.00815162	0.00758101
8	400	0.88	122.8	1.13636364	0.00814332	0.00716612
9	450	0.95	122.625	1.05263158	0.00815494	0.0077472
10	500	1.54	121.15	0.64935065	0.00825423	0.01271151
11	550	1.56	121.1	0.64102564	0.00825764	0.01288192
12	600	1.3	121.75	0.76923077	0.00821355	0.01067762

APPENDIX E2: Freundlich Isotherm Model

Experiment no	Ci(mg/L)	Ce(mg/L)	qe(mg/g)	Log Ce	Log Qe
1	50	0.95	122.625	-0.02228	2.08858
2	100	1.81	120.475	0.25768	2.0809
3	150	1.41	121.475	0.14922	2.08449
4	200	1.59	121.025	0.2014	2.08288
5	250	1.16	122.1	0.06446	2.08672
6	300	1.61	120.975	0.20683	2.0827
7	350	0.93	122.675	-0.03152	2.08876
8	400	0.88	122.8	-0.05552	2.0892
9	450	0.95	122.625	-0.02228	2.08858
10	500	1.54	121.15	0.18752	2.08332
11	550	1.56	121.1	0.19312	2.08314
12	600	1.3	121.75	0.11394	2.08547

APPENDIX F: Kinetic models

APPENDIX F1: Pseudo-first-order kinetic models and Pseudo-second-order kinetic model

Time	Ci (mg/L)	Ce (mg/L)	Qt (mg/g)	ln (qe-qt)	t/qt
0	50	3.72	115.7	1.74047	0
10	50	2.19	119.525	0.62861	0.08366
20	50	5.45	111.375	2.30508	0.17957
30	50	2.76	118.1	1.19392	0.25402
40	50	3.16	117.1	1.45862	0.34159
50	50	3.66	115.85	1.7138	0.43159
60	50	3.26	116.85	1.51513	0.51348
70	50	1.88	120.3	0.09531	0.58188
80	50	1.77	120.575	-0.19237	0.66349
90	50	1.78	120.55	-0.16252	0.74658
100	50	1.88	120.3	0.09531	0.83126

APPENDIX G: Bet analysis

APPENDIX G1: BET analysis of *M.oleifera*

MicroActive 6.00	TriStar II 3020 Version 3.02
	Serial # 1992 Unit 1 Port 2
Sample:	<i>M.oleifera</i>
Sample mass:	0.4345 g
Analysis free space:	45.7135 cm ³
Equilibration interval:	10 s
Sample density:	1.000 g/cm ³
Summary Report	
Surface Area	
Single point surface area at $p/p^\circ = 0.301262929$:	0.6903 m ² /g
BET Surface Area:	0.1273 m ² /g
Langmuir Surface Area:	-0.2209 m ² /g
t-Plot external surface area:	2.2178 m ² /g
BJH Adsorption cumulative surface area of pores between 17.000 Å and 3,000.000 Å width:	0.8800 m ² /g
BJH Desorption cumulative surface area of pores between 17.000 Å and 3,000.000 Å width:	1.1865 m ² /g
Single point adsorption total pore volume of pores less than 10,222.591 Å width at $p/p^\circ =$ 0.998129912:	0.001313 cm ³ /g
Single point desorption total pore volume of pores less than 10,222.591 Å width at $p/p^\circ =$ 0.998129912:	0.001313 cm ³ /g
t-Plot micropore volume:	-0.000759 cm ³ /g
BJH Adsorption cumulative volume of pores between 17.000 Å and 3,000.000 Å width:	0.001091 cm ³ /g
BJH Desorption cumulative volume of pores between 17.000 Å and 3,000.000 Å width:	0.001159 cm ³ /g
Pore Size	
Adsorption average pore diameter (4V/A by BET):	412.555 Å
Desorption average pore diameter (4V/A by BET):	412.555 Å
BJH Adsorption average pore width (4V/A):	49.588 Å
BJH Desorption average pore width (4V/A):	39.075 Å

APPENDIX G2: BET analysis of CH

MicroActive 6.00	TriStar II 3020 Version 3.02
	Serial # 1992 Unit 1 Port 1
Sample:	Coconut husk
Sample mass:	0.1546 g
Analysis free space:	47.5871 cm ³
Equilibration interval:	10 s
Sample density:	1.000 g/cm ³
Summary Report	
Surface Area	
Single point surface area at $p/p^\circ = 0.136419628$:	545.7288 m ² /g
BET Surface Area:	552.1280 m ² /g
Langmuir Surface Area:	703.2358 m ² /g
t-Plot Micropore Area:	377.7304 m ² /g
t-Plot external surface area:	174.3976 m ² /g
BJH Adsorption cumulative surface area of pores between 17.000 Å and 3,000.000 Å width:	128.0333 m ² /g
BJH Desorption cumulative surface area of pores between 17.000 Å and 3,000.000 Å width:	125.3558 m ² /g
Pore Volume	
Single point adsorption total pore volume of pores less than 5,159.045 Å width at $p/p^\circ = 0.996283835$:	0.343164 cm ³ /g
Single point desorption total pore volume of pores less than 5,159.045 Å width at $p/p^\circ = 0.996283835$:	0.343164 cm ³ /g
t-Plot micropore volume:	0.155818 cm ³ /g
BJH Adsorption cumulative volume of pores between 17.000 Å and 3,000.000 Å width:	0.158499 cm ³ /g
BJH Desorption cumulative volume of pores between 17.000 Å and 3,000.000 Å width:	0.154701 cm ³ /g
Pore Size	
Adsorption average pore diameter (4V/A by BET):	24.861 Å
Desorption average pore diameter (4V/A by BET):	24.861 Å
BJH Adsorption average pore width (4V/A):	49.518 Å
BJH Desorption average pore width (4V/A):	49.364 Å

APPENDIX G3: BET analysis of MOCH 3:1

MicroActive 6.00	TriStar II 3020 Version 3.02
	Serial # 1992 Unit 1 Port 3
Sample:	MOCH
Sample mass:	0.6645 g
Analysis free space:	45.8608 cm ³
Equilibration interval:	10 s
Sample density:	1.000 g/cm ³
Summary Report	
Surface Area	
Single point surface area at $p/p^\circ = 0.074871359$:	16.9464 m ² /g
BET Surface Area:	17.1006 m ² /g
Langmuir Surface Area:	20.5852 m ² /g
t-Plot Micropore Area:	13.2563 m ² /g
t-Plot external surface area:	3.8443 m ² /g
BJH Adsorption cumulative surface area of pores between 17.000 Å and 3,000.000 Å width:	
	2.8543 m ² /g
BJH Desorption cumulative surface area of pores between 17.000 Å and 3,000.000 Å width:	
	1.7493 m ² /g
Pore Volume	
Single point adsorption total pore volume of pores less than 5,642.711 Å width at $p/p^\circ =$ 0.996604027:	
	0.010866 cm ³ /g
Single point desorption total pore volume of pores less than 5,642.711 Å width at $p/p^\circ =$ 0.996604027:	
	0.010866 cm ³ /g
t-Plot micropore volume:	0.005224 cm ³ /g
BJH Adsorption cumulative volume of pores between 17.000 Å and 3,000.000 Å width:	
	0.004618 cm ³ /g
BJH Desorption cumulative volume of pores between 17.000 Å and 3,000.000 Å width:	
	0.003331 cm ³ /g
Pore Size	
Adsorption average pore diameter (4V/A by BET):	
	25.416 Å
Desorption average pore diameter (4V/A by BET):	
	25.416 Å
BJH Adsorption average pore width (4V/A):	64.711 Å

APPENDIX H1: Turbidity removal using MOCH 3:1

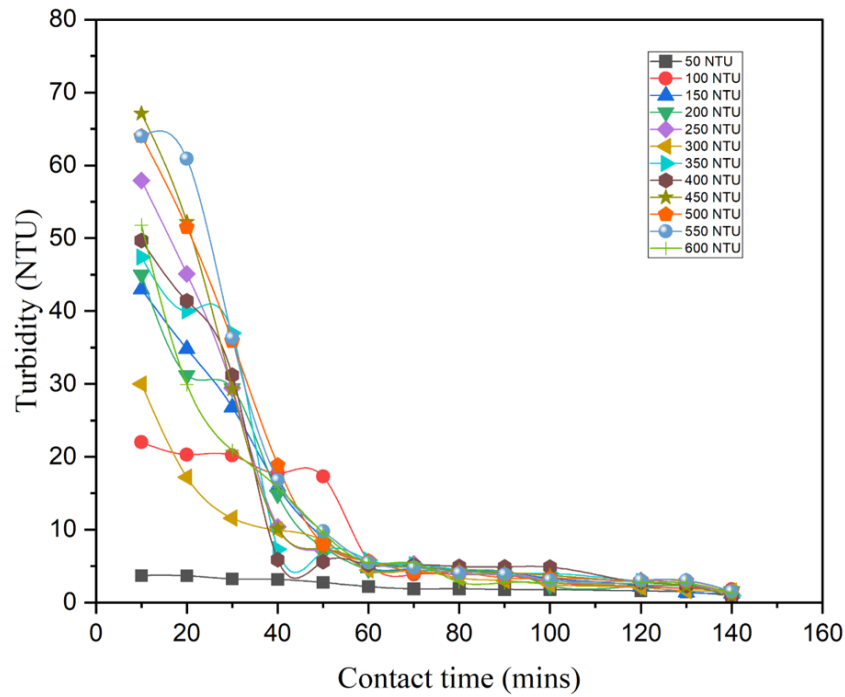


Figure 1: The turbidity reduction at different turbidity levels from 50 NTU to 600 NTU by using MOCH 3:1

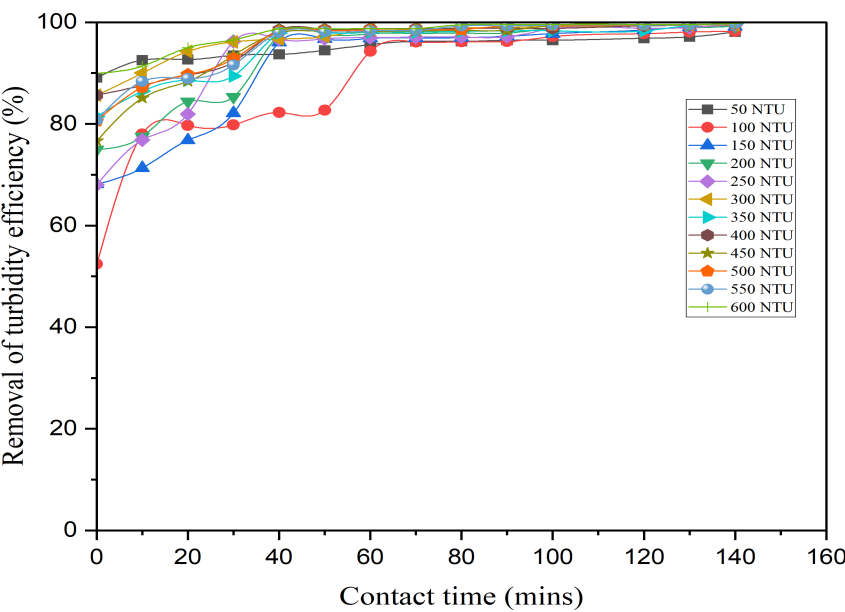


Figure 2: Turbidity removal efficiency using MOCH 3:1 at all turbidity levels

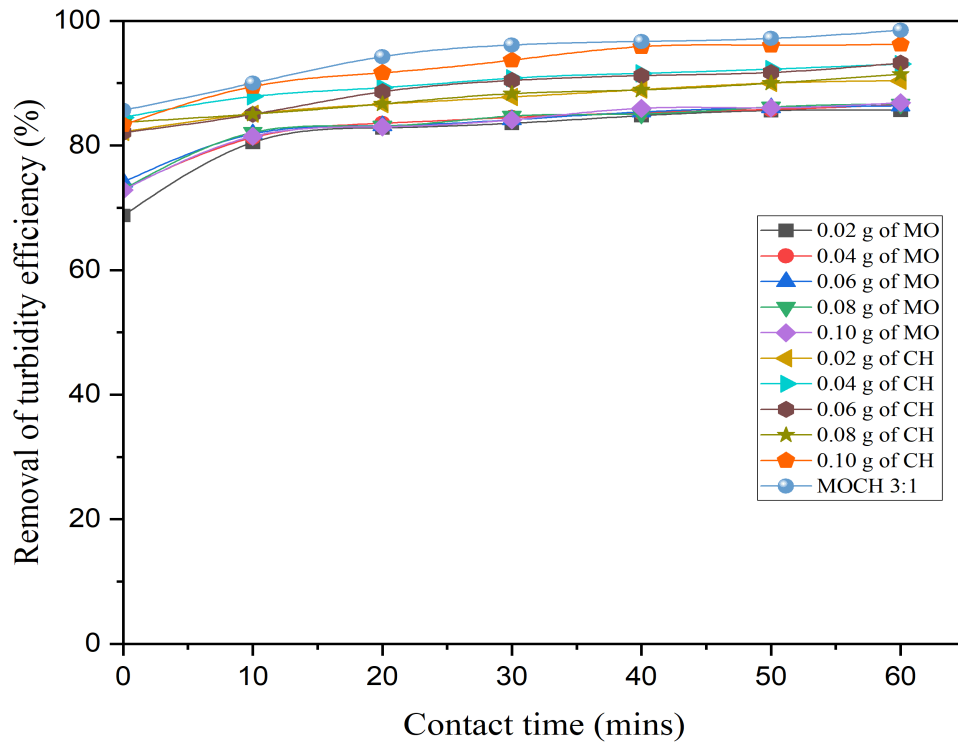


Figure 3: Turbidity removal efficiency using individual coagulant and MOCH 3:1

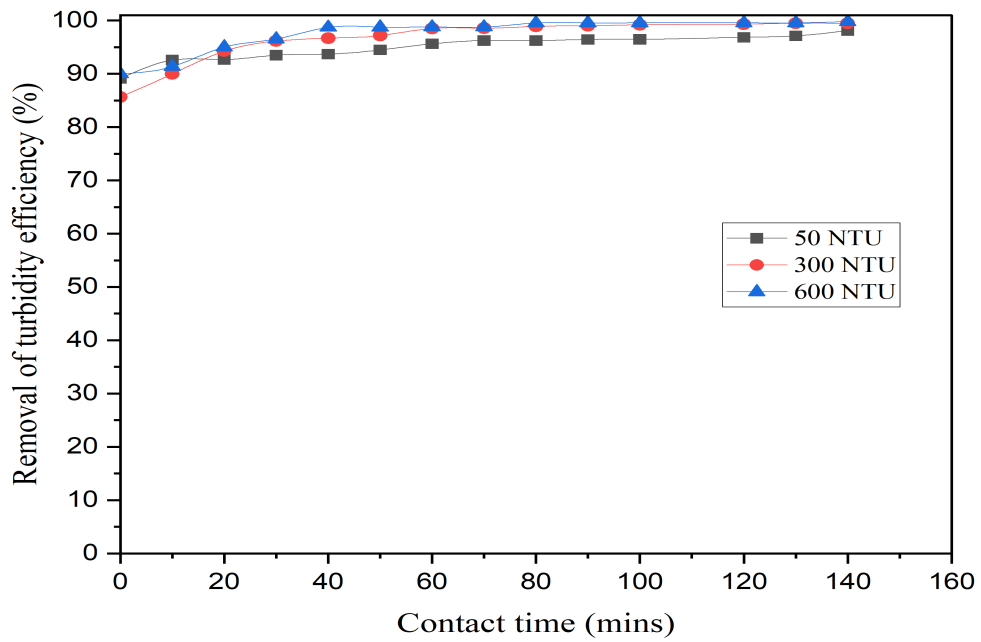


Figure 4: Turbidity removal efficiency using MOCH 3:1 at 50 NTU, 300 NTU and 600 NTU

APPENDIX H2: TDS removal using MOCH

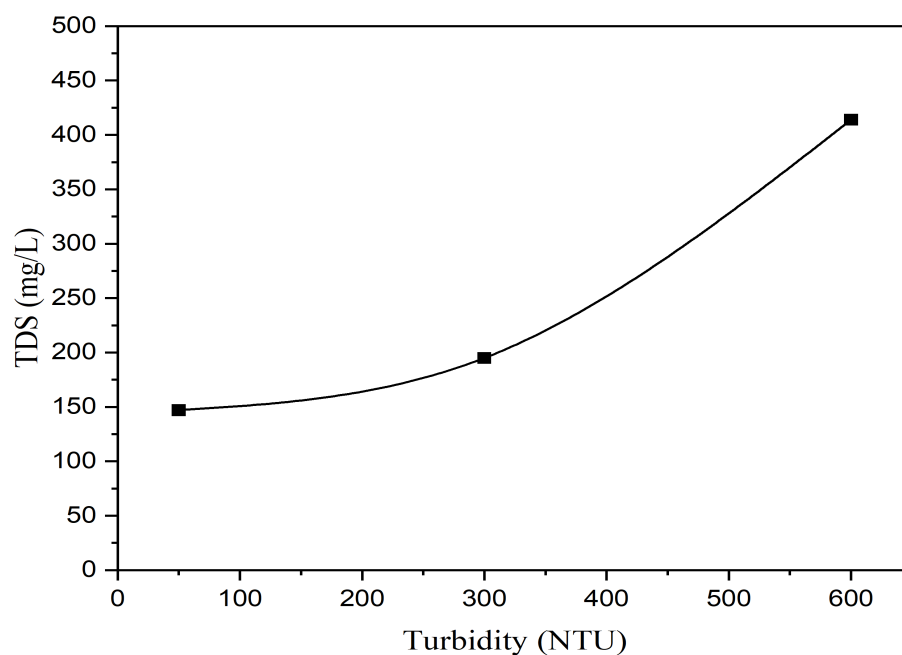


Figure 1: Total dissolved solids removal by using MOCH 3:1 at 50 NTU, 300 NTU, 600 NTU

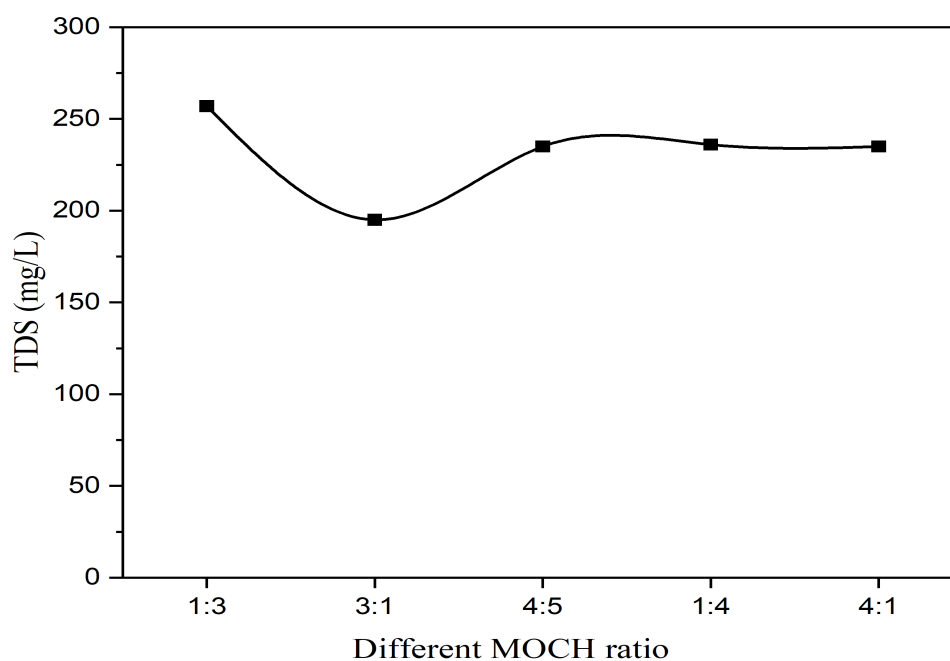


Figure 2: Total dissolved solids removal by using different MOCH ratio

APPENDIX H3: EC removal using MOCH

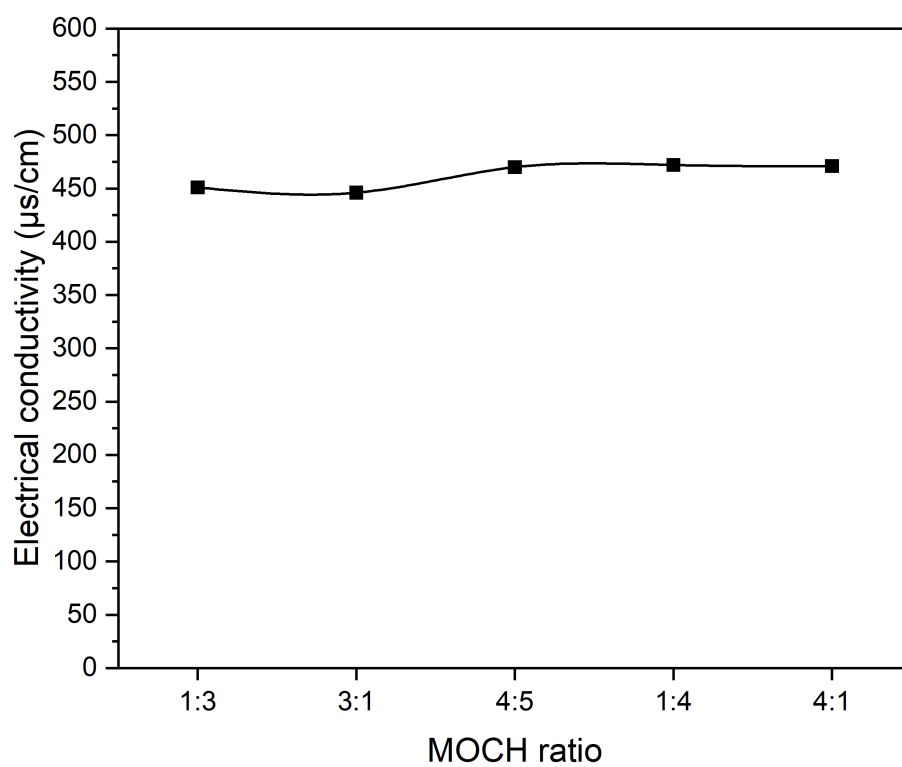


Figure 1: Electrical conductivity removal using different MOCH ratio

APPENDIX H4: Effect of temperature on turbidity removal using MOCH 3:1

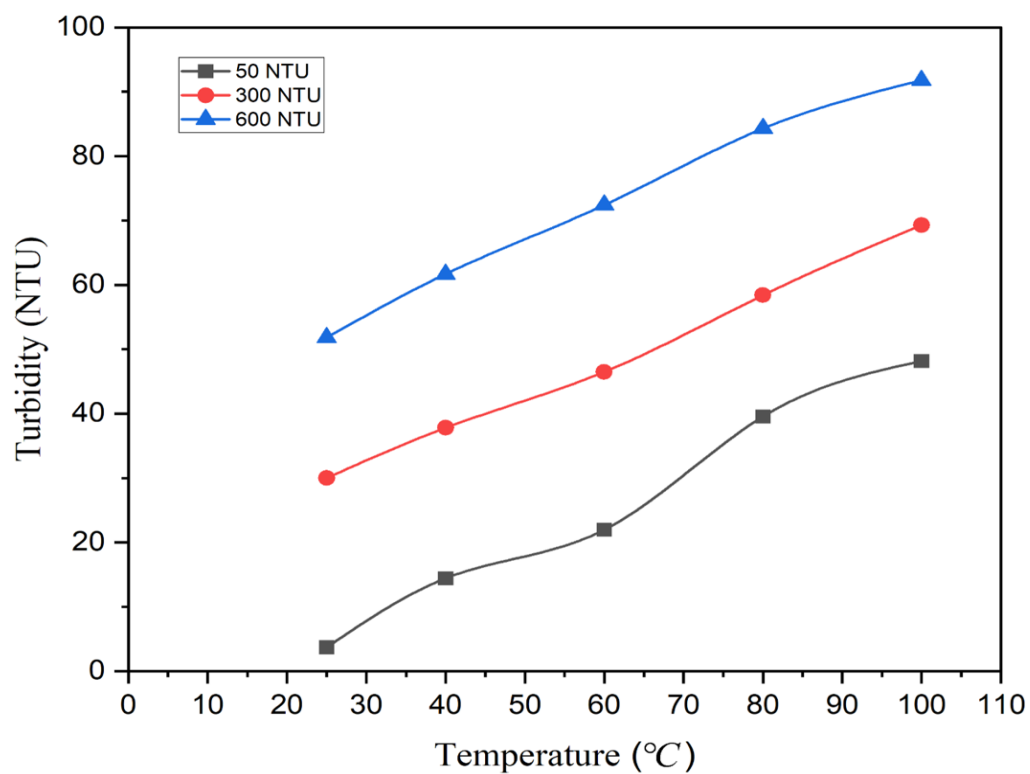


Figure 1: Effect of temperature on turbidity removal using MOCH 3:1

APPENDIX I: ZETA POTENTIAL

APPENDIX I1: Zeta potential analysis of *M.oleifera*

Autotitration Report
v2.0



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Sample Details

Titration Type: pH Titration Measurement Type(s): Zeta
Sample Name: *M.oleifera*
SOP Name: Default pH Zeta 12-2.sop
General Notes:

File Name: Claudeareena250324.dts
Record Number: 35 Measurement Date and Time: Tuesday, March 26, 2024 9:22...

System

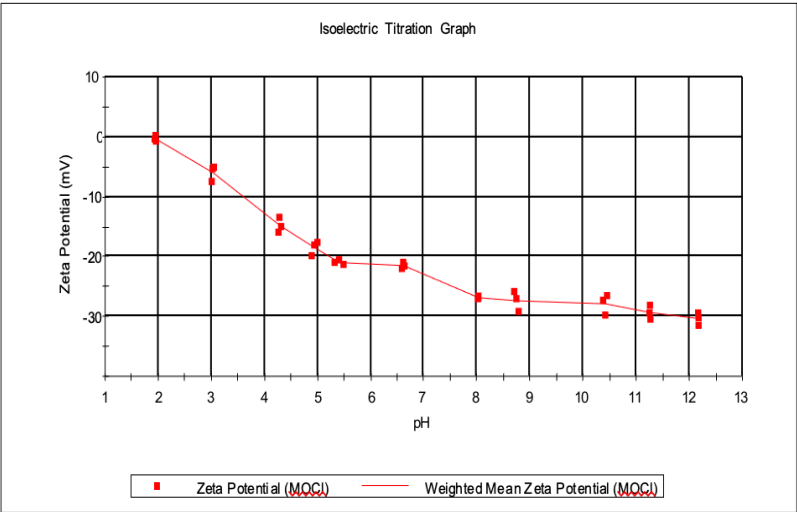
Cell Description: Clear disposable zeta cell

Titants

	Type	Name	Concentration (M)
Bottle 1:	Acid	HCl	0.250
Bottle 2:	Acid	HCl	0.500
Bottle 3:	Base	NaOH	0.500

Results

IEP(s) in (pH): Total Injected Volume (mL): 0.300



APPENDIX I2: Zeta potential analysis of CH

Autotitration Report
v2.0

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Sample Details

Titration Type: pH Titration Measurement Type(s): Zeta
Sample Name: Coconut husk
SOP Name: Default pH Zeta 12-2.sop
General Notes:

File Name: Claudeareena250324.dts
Record Number: 1 Measurement Date and Time: Monday, March 25, 2024 12:5...

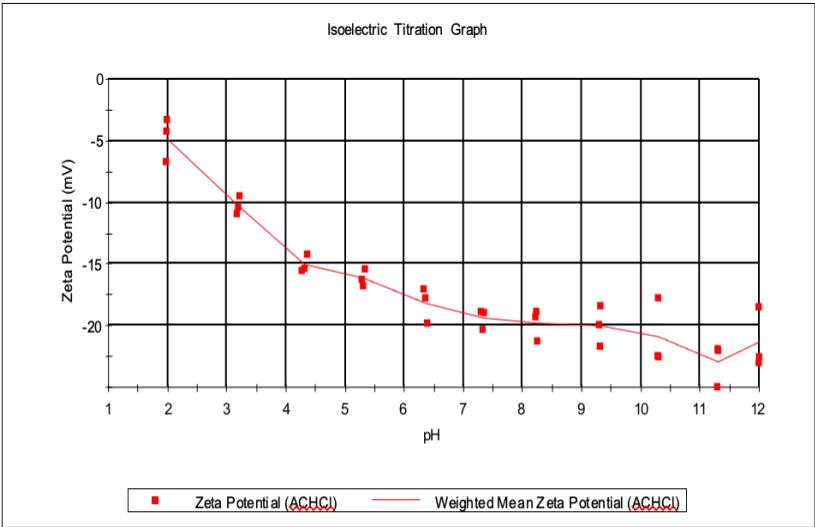
System

Cell Description: Clear disposable zeta cell

Titriments	Type	Name	Concentration (M)
Bottle 1:	Acid	HCl	0.250
Bottle 2:	Acid	HCl	0.500
Bottle 3:	Base	NaOH	0.500

Results

IEP(s) in (pH): Total Injected Volume (mL): 3.65



APPENDIX I3: Zeta potential analysis of MOCH 3:1

Autotitration Report
v2.0



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Sample Details

Titration Type: pH Titration Measurement Type(s): Zeta
Sample Name: MOCH 3:1
SOP Name: Default pH Zeta 12-2.sop
General Notes:

File Name: Claudeareena250324.dts
Record Number: 69 Measurement Date and Time: Tuesday, March 26, 2024 3:59...

System

Cell Description: Clear disposable zeta cell

Titrants

	Type	Name	Concentration (M)
Bottle 1:	Acid	HCl	0.250
Bottle 2:	Acid	HCl	0.500
Bottle 3:	Base	NaOH	0.500

Results

IEP(s) in (pH): Total Injected Volume (mL): 0.344

