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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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.

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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 ℃, 40 ℃, 60 ℃, 80 ℃ and 100 ℃). 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-N_2), and zeta potential. The MOCH 3:1 agreed with the Langmuir, indicating monolayer formation and pseudo-second-order kinetic models, suggesting chemisorption, with R^2 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℃. 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 keberkesanannya sebagai adsorben semula jadi untuk menyingkirkan bahan pencemar daripada sisa kumbuhan. 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-N_2), dan potensi zeta. MOCH 3:1 mematuhi model Langmuir (monolayer) dan model kinetik pseudo-tertib kedua (kemisorpsi) dengan nilai R^2 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℃. 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 *Moringa oleifera*
FTIR	Fourier Transform Infrared Spectroscopy
MF	Microfiltration
MOCH	Combination of *Moringa oleifera* 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 alkalinization 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 ℃, 40 ℃, 60 ℃, 80 ℃ and 100 ℃). 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