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PERFORMANCE OF CHITOSAN FROM MUSHROOM AS BIOCOAGULANT AGENT FOR KAOLIN AND PALM OIL MILL EFFLUENT WASTEWATER

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By

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Chairman : Associate Professor Zurina binti Zainal Abidin, PhD
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This project aims to investigate the potential use of commercial chitosan produced from mushroom (CMs) as bio-coagulants which are water-soluble chitosan (WSC) and acid-soluble chitosan (ASC) in wastewater treatment. Palm Oil Mill Effluent (POME) is wastewater generated from the palm oil milling industry must be treated properly before being discharged into the environment. The use of inorganic coagulants like alum can potentially lead to the spread of chronic diseases due to the residual content of the coagulants in the treated wastewater. Thus, this study searched for an alternative coagulant using (CMs) for coagulation process. In this study, the optimum condition for synthetic water kaolin treatment when WSC was at pH 8, with a dosage of 10 mg/L, and with almost 100% turbidity removal. While ASC optimum conditions were at pH 11, dosage 10 mg/L and almost 100% turbidity removal. On the other hands, the performances of CMs coagulants as bio-coagulants were compared to aluminum sulphate (alum) coagulant. The results showed that alum gave 93% turbidity removal at optimum pH 4 and with an optimum dosage of 1200 mg/L.

The treatment of POME wastewater by using CMs as a bio-coagulant was successfully able to reduce COD with up to 73% in value, 70% of BOD reduction and 99% of TSS removal, whereas alum coagulant resulted in 71% of COD, 65% of BOD reduction and 86% of TSS removal. The second objective of this study was to examine the characterization of the WSC and ASC by using Fourier Transform Infrared (FTIR) analysis, Zeta Potential, and antimicrobial activity. The results of FTIR confirmed the existence of amino groups in the backbone of CMs. The results of Zeta Potential analysis showed that the surface charges for WSC and ASC were 40.46 mV ± 1.01 and +70.24 mV ± 3.69, respectively. The antimicrobial activity analysis showed that ASC is more active than WSC where the concentration used of CMs was 10%. The ASC was active to inhibit microbes within around 7 mm zone which is considered clear zone of inhibition. The estimated cost for treatment using WSC, ASC and alum was RM 5.5, 4.52 and 84 per cubic meter respectively of POME treated wastewater.

In conclusion, this study has proven that chitosan mushroom possesses a significant potential to be used as a bio-coagulant for wastewater treatment and also economical.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk Ijazah Master Sains

KECEKAPAN CHITOSAN MUSHROOM SEBAGAI AGEN BIO KOAGULAN ON KEKERUHAN DIANGKAT DARJAT SISA

Oleh

ODAY ADNAN IRHAYYIM

Januari 2017

Pengerusi : Profesor Madya Zurina binti Zainal Abidin, PhD
Fakulti : Kejuruteraan

Projek ini bertujuan untuk mengkaji potensi chitosan yang dihasilkan oleh cendawan (CMs) sebagai agen bio-penggental iaitu chitosan larut-air (WSC) dan chitosan larut-asid (ASC) dalam rawatan air sisa. POME merupakan air sisa yang terhasil daripada industri minyak sawit dan mesti dirawat dengan sebaiknya sebelum dilepaskan ke persekitaran. Penggunaan penggental bukan organic seperti tawas telah membawa kepada penyebaran penyakit kronik akibat kesan kandungan sisa bahan penggental dalam air sisa yang dirawat. Oleh itu,kajian ini adalah bertujuan untuk mencari alternatif menggunakan CMs dalam proses penggentalan. Dalam kajian ini, keadaan optimum untuk rawatan air kaolin sintetik menggunakan WSC pada pH 8, dengan dos sejumlah 10 mg/L, dan dengan hampir 100% penyingkiran kekeruhan. Manakala keadaan optimum menggunakan ASC adalah pada pH 11, dengan dos sejumlah 10 mg/L, dengan hampir 100% penyingkiran kekeruhan. Pada masa yang sama, kebolehan penggental CMs sebagai bio-penggental telah dibandingkan dengan penggental aluminium sulfat (tawas). Keputusan menunjukkan bahawa tawas hanya memberikan 93% penyingkiran kekeruhan pada keadaan optimum pH 4 dengan dos optimum 1200 mg/L. Rawatan POME dengan menggunakan CMs sebagai bio-penggental telah berjaya mengurangkan COD sehingga 73%, 70% pengurangan BOD dan 99% pengurangan TSS manakala bio-penggental tawas menunjukkan hanya 71% COD, 65% pengurangan BOD dan 86% pengurangan TSS.

Objektif kedua kajian ini pula adalah untuk memeriksa ciri-ciri WSC dan ASC dengan menggunakan analisis Spektroskopi Fourier Menukar Sinaran Merah (FTIR), Potensi Zeta dan aktiviti antimikrob. Keputusan FTIR telah mengesahkan kewujudan kumpulan-kumpulan amino dalam tulang belakang CMs. Keputusan analisis Potensi Zeta menunjukkan bahawa cas permukaan untuk WSC dan ASC masing-masing ialah 40.46 mV ± 1.01 dan +70.24 mV ± 3.69. Analisis aktiviti antimikrob pula menunjukkan bahawa ASC lebih aktif daripada WSC dimana kepekatan CMs yang digunakan adalah 10%. ASC aktif untuk merencat mikrob dalam zon lingkungan 7 mm yang dikira sebagai zon bebas perencatan. Anggaran kos untuk rawatan

menggunakan WSC, ASC dan tawas adalah RM 5.5, 4.52 dan 84 bagi setiap satu meter padu air sisa POME yang dirawat. Kajian ini telah membuktikan bahawa cendawan chitosan mempunyai potensi yang signifikan dalam pengunaannya sebagai bio-penggental serta kos yang efektif untuk merawat air sisa.

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I certify that a Thesis Examination Committee has met on 31 January 2017 to conduct the final examination of Oday Adnan Irhayyim on his thesis entitled "Performance of Chitosan from Mushroom as Biocoagulant Agent for Kaolin and Palm Oil Mill Effluent Wastewater" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Master of Science.

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This thesis was submitted to the Senate of the Universiti Putra Malaysia and has been accepted as fulfillment of the requirement for the degree of Master of Science. The members of the Supervisory Committee were as follows:

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This is to confirm that:

- the research conducted and the writing of this thesis was under our supervision;
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LIST OF ABBREVIATIONS

CMs	Water-soluble chitosan and acid-soluble chitosan
WSC	Water-soluble chitosan
ASC	Acid-soluble chitosan
POME	Palm Oil Mill Effluent
Alum	Aluminum Sulphate
COD	Chemical Oxygen Demand
BOD	Biological Oxygen Demand
TSS	Total Suspended Solids
FTIR	Fourier Transform Infrared
TS	Total Solids
N	Total Nitrogen
P	Phosphorus
VOC	Volatile Organic Compounds
SS	Suspended Solids
DO	Dissolved Oxygen
Kw	Dissociation Constant
NOM	Natural Organic Matter
DADMAC	Diallyl Dimethyl Ammonium Chloride
PAC	Polyaluminium Chloride
MW	Molecular Weight
DD	Degree of Deacetylation
Ctnp	Mushroom Chitosan Nanoparticle

CHAPTER 1

INTRODUCTION

1.1 General Overview

Water is definitely the main secret of life. Global water consists of approximately 95.6% oceans, 2.5% fresh water, and the rest is saline water; in addition, the surface water is only about 1.2% of the total freshwater of the earth. (figure 1.1) (Gleick, 1993). The water crisis has become one of the current issues nowadays. The population growing led to a reduction in clean water sources, thereby threatens a numerous people; for instance, the report of (Unicef, 2008) estimated that one billion people or more are unable to obtain safe water.

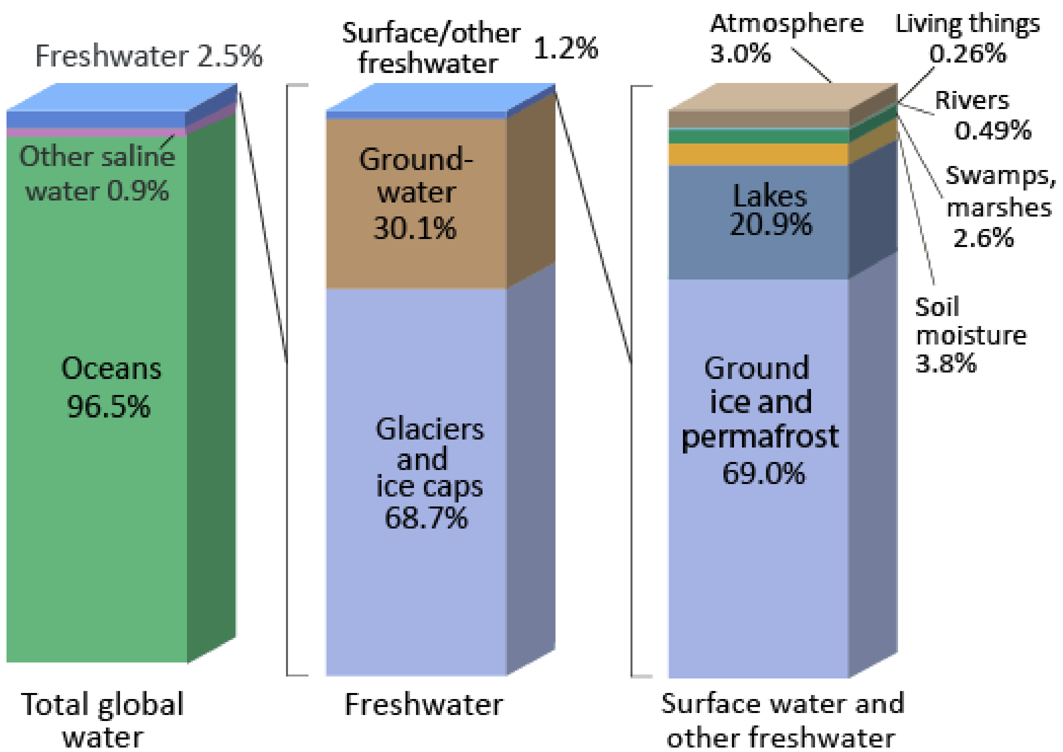

Figure 1.1 : The distribution of total world water
(http://water.usgs.gov/edu/earthwherewater.html)

Discharging the wastewater without remedy into the environment is no longer accepted choice because it carries a high load of organic and inorganic pollutants, pathogenic, and serious microorganisms. When the wastewater released without treatment into water bodies, that generates accumulated contamination in the aquatic environment (Riffat, 2012), thus, it may cause many diseases. The report issued by

(WATER, 2014) had stated that approximately 502,000 diarrheal deaths occurred in low and middle-income countries due to the inadequate and unsafe drinking-water.

Many of recent studies have focused on water control and wastewater treatment (Punmia et al., 1998). In fact, there are key methods for wastewater treatments; namely, physical, and chemical and biological treatment. Physical Treatment consists of preliminary, primary, and advanced primary process. Preliminary treatment aims to remove the gross solids such as large particles, and grit which destroy the apparatus. The primary treatment, which is a physical operation such as sedimentation, is typically used to harvest the floating particles of the wastewater. Subsequently, advanced primary treatment is used to boost the elimination process of suspended solids and to increase the precipitation rate. Secondly, chemical and biological treatment that includes secondary and advanced (or tertiary) treatment. Secondary treatment is applied to remedy the majority of the organic matters. Afterward, the advanced treatment uses supplementary unit operations and processes to deal with the residual suspended solids and other elements which were not removed considerably after the secondary treatment (Tchobanoglous, 1979).

Coagulation is a pivotal physicochemical process which has been broadly applied as a pre or post process step in wastewater treatment (Saini & Kumar, 2016). Coagulation is a principal process to reduce suspended solids, turbidity, viruses, and any particles within the size (0.1 to 10 µm) away from wastewater (Asano, 1998). This process involves basically, surface charge neutralization of particles and flocculation (i.e. floc creation) (Norulaini et al., 2001). When coagulation process starts, rapid mixing is applied in order to disperse the coagulant particles readily, thereby, destabilization of the colloidal particles is done. Slow mixing is applied to let the destabilized particles slowly aggregate and create settleable flocs. Undoubtedly, coagulation is applied to remedy diverse wastes, for instance, textile wastewater (Han et al., 2016), coal mining wastewater treatment (Galloux et al., 2015), biodiesel wastewater (Daud et al., 2015), and drinking water (Hu et al., 2015).

Coagulation efficacy depends on several factors such as coagulant category, coagulant concentration, sample PH, ionic strength and wastewater characteristics (Norulaini et al., 2001). Coagulants can be inorganic such as aluminum sulfate and magnesium chloride or polyelectrolyte (e.g., synthetic coagulants and natural coagulant).

1.2 Problem Statement

Alum coagulants are the most common agents used for water remedy (Okuda et al., 1999). However, recent studies approved that the use of aluminum salts showed extreme weakness in affecting the wastewater (Miller et al., 1984; Ndabigengesere et al., 1995). Abidin et al. (2011) mentioned that the use of alum salts may potentially caused many Alzheimer related disease cases. When these metals applied for water therapy purpose, a high aluminum concentration can be detected in the sludge residual of treated water as well as their efficacy is extreme pH- sensitive (Z. Yang et al., 2013). Subramonian et al. (2015) stated that the inorganic coagulants have serious side effects on mankind; also their efficiency in cold water is low. Moreover, these types of coagulants are rarely applied in the developing countries due to their high prices. In addition, alum forms a large amount of residual sludge and gives dewatering problem. (Özacar, 2000). The use of the synthetic organic polymers such as acrylamide and poly aluminium chloride exhibited many adverse consequences (e.g. neurotoxicity and strong carcinogenic impacts) (Choy et al., 2014).

To manage above mentioned issues, many researchers have turned their faces toward safer coagulants that can be:1) more effective, 2) human health friendly, 3) economic, and 4) biodegradable (Abidin et al., 2013; Choy et al., 2014; Shamsnejati et al., 2015). For this effect, natural coagulants have attracted considerable attention recently. Biocoagulants can be extracted from the sustainable sources (e.g. plants, animals, and microorganisms) (Abidin et al., 2011; Abidin et al., 2013; Zurina et al., 2014). In addition, polyelectrolytes are natural coagulants and they are able to condition the sludge and solve the dewatering problem (Özacar, 2000). Natural coagulants such as Ocimumbasilicum, Moringa Olivera, chitosan, and other polymers are used in wastewater treatment as they are rich in polyelectrolytes in their contents (Bolto & Gregory, 2007; Oladoja, 2015; Shamsnejati et al., 2015).

Natural coagulant has proven to demonstrate a high efficacy in water treatment. For instance, (Chi & Cheng, 2006) studied the chitosan efficiency on an effluent that produced high fat and protein content in sludge where authors found out chitosan possess high potential. On the contrary of conventional coagulant, chitosan is more feasible, lower cost, low toxic and produce reasonable sludge volume. Generally, invertebrates (e.g. shrimp and crab shell) are the main source of the commercial chitosan. However, the raw materials required are seasonal and variable thereby leads to a difficult process. Hence, chitosan produced from this process is heterogeneous based on its physiochemical characteristics. Also, chitosan production rate is low due to the long time required to complete the reaction. In this scenario, exploiting the fungi is recommended as an alternative source for chitosan as it is rich in chitin and chitosan can be easily obtained from their wall cell (Pochanavanich & Suntornsuk, 2002).

The previous studies have dealt with the chitosan extracted from invertebrates; therefore, it is necessary to explore alternative source of chitosan of the new origin. In this study, chitosan of mushroom origin is studied to explore its ability as a bio-coagulant for wastewater treatment technology. From our knowledge, chitosan from mushroom can be produced by utilizing the waste accumulated due to mushroom production and harvest. This in turn will help to reduce waste accumulation and disposal to landfill which represents environmental issue for the producers (Tao et al., 2004).

1.3 Research Objectives

The current study presents an advanced water treatment through bio coagulant agent based on water-soluble chitosan mushroom (WSC) and acid-soluble chitosan mushroom (ASC). Therefore, there are three objectives of this study which are:

1. To characterize the ASC and WSC from mushroom as bio coagulant agent.
2. To investigate the coagulation performance of ASC and WSC for Kaolin and POME and compared with Alum.

1.4 Scope of Research

In this work the efficiency of chitosan mushroom (WSC and ASC) on kaolin wastewater and POME was investigated. This work included observing the turbidity reading, pH alteration, chemical oxygen demined (COD), biological oxygen demined (BOD), and total suspended solids (TSS). Furthermore, characterization of chitosan mushroom was limited to involve zeta potential, Fourier Transform Infrared (FTIR), and antimicrobial activity. Actually, the performance of chitosan was studied by test the alteration in the pH of treated water as well as by observing the effects of coagulant concentration on the coagulation process. The pH range in all pH optimization tests was from 2 to 12 while the dose of WSC or ASC ranged from 5 to 60 mg/L. This work involved a comparison between WSC and ASC as well as a comparison with alum. Moreover, this project included a comparison between the results obtained from the application of chitosan of mushroom origin with the results reported by other investigators in the literature which associated with chitosan produced from invertebrates (e.g. shrimp and crab shell).

1.5 Thesis Organization

The current manuscript is partitioned into five parts starting by the preface of this investigation which it affirms on the background of the study as well as the research problem statement with the research targets. Regarding chapter two, literature review discloses the important previous studies and their findings, potential, and the weakness, which mainly centering on the coagulant-flocculation process of wastewater therapy. On the other hand, chapter three indicates steps are taken throughout the study for data collections besides the provision steps which were deemed within the study. Chapter four confirms the data acquired and argues the outcomes with the previous studies. Chapter five includes the comprehensive conclusions and recommendations for next studies. Finally, this thesis is ended with the references and appendices associated with this thesis.

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