



Effective coagulation-flocculation treatment of highly polluted palm oil mill biogas plant wastewater using dual coagulants: Decolourisation, kinetics and phytotoxicity studies



A.Y. Zahrim^{a,b,c,d,*}, Z.D. Dexter^a, C.G. Joseph^{b,e}, N. Hilal^f

^a Chemical Engineering Programme, Faculty of Engineering, Universiti Malaysia Sabah, Jalan UMS 88400 Kota Kinabalu, Sabah, Malaysia

^b Water Research Unit, Universiti Malaysia Sabah, Jalan UMS 88400 Kota Kinabalu, Sabah, Malaysia

^c Sustainable Palm Oil Research Unit, Universiti Malaysia Sabah, Jalan UMS, 88400 Kota Kinabalu, Sabah, Malaysia

^d Energy Research Unit, Universiti Malaysia Sabah, Jalan UMS 88400 Kota Kinabalu, Sabah, Malaysia

^e Industrial Chemistry Programme, Faculty of Science and Natural Resources, Universiti Malaysia Sabah, Jalan UMS 88400 Kota Kinabalu, Sabah, Malaysia

^f Centre for Water Advanced Technologies and Environmental Research (CWATER), College of Engineering, Swansea University, Swansea SA2 8PP, UK

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ABSTRACT

The performance of several chemical coagulants including ferric chloride, calcium lactate, magnesium hydroxide, aluminium chlorohydrate, and polydiallyldimethylammonium chloride (polyDADMAC) were investigated in removing colour of palm oil mill biogas plant wastewater (POMBWP). The results show that ferric chloride as a sole coagulant can achieve high colour removal of more than 80% without needed for pH adjustment, which indicates the effectiveness of the coagulant to treat palm oil mill biogas plant wastewater (POMBWP). However, dual coagulants i.e. ferric chloride-anionic polyacrylamide (APAM) shows better performance than ferric chloride-polyDADMAC in terms of colour removal, pH, with shorter sedimentation time. The addition of polymer to system not only reduces the ferric chloride dosage, but also increases the colour removal of more than 20%. Comparison between APAM and polyDADMAC as flocculant aids shows that APAM can achieve stable removal at wider pH range and lowest sedimentation time at 20 min while polyDADMAC was at one hour. Both dual coagulants were followed second order kinetic and APAM shows the highest rate over polyDADMAC i.e. $3 \times 10^{-5}/\text{PtCo.min}$ and $2 \times 10^{-5}/\text{PtCo.min}$ respectively. Addition of polymers reduced phytotoxicity of generated sludge and the sludge has potential to be reused for land application.

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1. Introduction

Most of palm oil mills have to deal with their highly polluting wastewater that is known as palm oil mill effluent (POME). This has led to the emergence of a new treatment technology with environmental friendly approach. Palm oil mill has adopted anaerobic digestion system technology known as biogas plant where it can convert the organic carbon i.e. chemical oxygen demand (COD) and biochemical oxygen demand (BOD) to biogas with the help of anaerobic bacteria. This renewable energy generated from this plant can be used to generate electricity and can save tones of fossil fuel. However, the performance of biogas plant is primarily depending on the bacteria that readily exist in the wastewater.

This bacteria can be sensitive to their environment, namely pH and temperature. It has been reported that biogas plant can remove up to 80% of COD from the POME after being anaerobically digested [1]. Nevertheless, the treated wastewater from this plant has high level of polluting contents, which do not comply with the discharge requirement limit set by Department of Environment (DOE). Moreover, the wastewater colour turned from brownish (POME) to blackish after POME was digested. This colour transformation might be due to Maillard reaction of natural condensation between carbonyl groups and free amino groups in POME that produce melanoidin, which is an anaerobic fermentation by product [2]. As a result, this highly coloured wastewater can create aesthetic problem and affect the photosynthesis activity by reducing the sunlight penetration if it is discharged into the watercourse [3–6]. Bunnung et al. [7] have studied the decolourisation of palm oil mill biogas plant wastewater (POMBWP) by using physical method and also combined with biological method. They found out that the decolourisation of more than 80% can be achieved when using palm

* Corresponding author at: Chemical Engineering Programme, Faculty of Engineering, Universiti Malaysia Sabah, Jalan UMS 88400 Kota Kinabalu, Sabah, Malaysia.
E-mail address: zahrim@ums.edu.my (A.Y. Zahrim).

ash as adsorbent and after pre-treated with biological method of mixed cultures [8,7]. However, the characteristics of POMBPW in their studies were lower as compared in this study; especially the colour and COD. In the study, the decolourisation increased with increase in agitation time, as well as in mixed cultures cultivation [8,7].

Typically, coagulation-flocculation process is regarded as one of the most important treatment processes of industrial wastewater [9] and raw water [10] due to its simplicity, effectiveness, and low energy consumption. Coagulation-flocculation process is widely used in industry in the past few decades. In addition, this process usually used is used as a pre-treatment to other integrated treatment process such as membrane filtration [11–13], biological treatment [14,15] and advanced oxidation processes [16–18]. The attribute of wastewater plays an important factor in choosing the most suitable coagulant for the process treatment. Inorganic coagulants such as aluminum and iron salts are commonly used in water treatment. However, they can alter the pH of the treated water, increase toxicity level and sometimes can increase the wastewater colour, which can make secondary contamination [19,9]. Synthetic organic polymers such as polyacrylamide (PAM) and poly(diallyldimethylammonium chloride) (PolyDADMAC) have been widely used not only as flocculant aids but also as direct flocculation in most industries [20,21]. Generally, polymers will bind the flocs via bridging mechanism and produce larger, denser, stronger, and rapid-settling flocs. The formation of larger and denser flocs is mainly due to the higher molecular weight of polymer coagulants which improve interparticle bridging and provide more adsorption sites [22–24].

Ferric salt was used to further studied due to problems associated with aluminium salt. High concentration of aluminium species in water may lead to Alzheimer's disease upon consumption [9,25]. Therefore, the residue of Al^{3+} content in the discharge should be below 10–15 mg/L according to Malaysian standard [26].

Over the years, the usage of dual coagulants (inorganic coagulant and polymer based coagulant) has benefits over the sole use of inorganic or polymer coagulants. These benefits are mostly related to the reduction of the inorganic coagulant dosage while producing larger, denser, and stronger flocs [20]. Several studies have reported the effectiveness of dual coagulants application when dealing with various types of wastewater where it can reduce up to 90% of COD, total suspended solids (TSS), turbidity, and colour. Furthermore, it can reduce the consumption of coagulant demand, thus reduce the overall cost of the coagulation-flocculation process [20,27–33].

The concern in accumulation of large amount of sludge from this process has led to finding the best way to manage the sludge in economically and environmentally approach. To the best of our knowledge, there were small amount of work in the literature concerning studies of phytotoxicity from sludge generated through the coagulation-flocculation process alone. Most researchers combine the coagulation-flocculation with another treatment process in which to further degrade the organic compound to lower level. Bedekar et al. [34] reported that the coagulated dye sludge was fermented by using consortium-BBA and found that the phytotoxicity was decreased after coagulation and degradation mechanism [34]. In another study, combination of coagulation, acid cracking, and Fenton-like process could result the phytotoxicity of final olive oil mill wastewater to decrease [35]. Another combination was reported by [36], where they found high toxicity removal up to 90% in their phytotoxicity test of landfill leachates. The combination was coagulation-flocculation-Fenton and powder zeolite adsorption [36].

Lee et al. [37] has used a ferric chloride–polyacrylamide inorganic–organic hybrid polymer which was synthesized using a ferric chloride/polyacrylamide ratio of 1:1 via free radical solution polymerization. The hybrid polymer was then tested on kaolin sus-

pension flocculating activities and Terasil Red R dye wastewater and the results showed it is capable to decolourise up to 99% of colour [37]. Another study reported by Yang et al. [38] used polyferric chloride (PFC) and poly(epichlorohydrin-dimethylamine) [P(ECH-DAM)] to test for the coagulation treatment of reactive dye suspensions and found that composite PFC-P(ECH-DAM) can improved characteristics, such as increased efficient polymeric speciation concentration, improved zeta potential, and enhanced flocculation performance [38]. In summary, both of these studies were using hybrid polymer and focusing on dye wastewater. The hybrid polymer synthesizing is time consuming due to the complicated preparation [39]. To the best of our knowledge, there is no publication in the literature dealt with dual coagulants before in POMBPW decolourisation.

In this study, the ability of various coagulants in removing colour of POMBPW was investigated by various metal coagulants such as ferric chloride, calcium lactate, magnesium hydroxide, and aluminium chlorohydrate. Then, the addition of polymers i.e. poly-DADMAC and PAMs as flocculant aids was studied. Last but not least, the coagulation kinetic order was predicted and phytotoxicity test of the filtered sludge was carried out.

1.1. Coagulation kinetics

The kinetics of coagulation process can be described as [19,40];

$$\frac{dC}{dt} = -kC^n \quad (1)$$

where “C” is the total mass of particle per litre, “t” is the coagulation time, “k” is the nth order coagulation rate constant, and “n” is the order of the coagulation process. The turbidity concentration decrease as a function of increasing time (t), is represented as a negative sign in Eq. (1).

During the coagulation process, the rate of removal of colour is proportional to the initial colour concentration and amount of coagulant used. For a first order (n = 1) coagulation process, the rate equation is shown as;

$$\frac{-dC}{dt} = k_1 C \quad (2)$$

and the integral form is written as follows;

$$\ln\left(\frac{C_0}{C}\right) = k_1 t \quad (3)$$

where “ C_0 ” is initial colour of the POMBPW in PtCo, “C” is final colour of the POMBPW at time “t” and “ k_1 ” is the first order rate constant in (1/min). Eq. (3) shows that the plot of $\ln(C_0/C)$ against time (t) should be a straight line passing through the origin and the slope is k_1 . However, if the line does not pass through the origin but intercepts along the Y-axis line, it follows second order coagulation process (n = 2) which is shown as;

$$\frac{-dC}{dt} = k_2 C^2 \quad (4)$$

and the integral form is written as follows;

$$\frac{1}{C} = k_2 t + \frac{1}{C_0} \quad (5)$$

where “ k_2 ” is the second order rate constant in (1/PtCo.min).

2. Methodology

2.1. Materials

The biogas plant wastewater was collected from the Lahad Datu, Sabah. In this mill, the palm oil mill effluent (POME) from the cooling pond is fed into three anaerobic digester tanks which are located

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