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Research paper



Recent Advances in Photocatalytic Treatment of Palm Oil Mill Effluent (POME): A Review

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Abstract

Palm oil mill effluent (POME) pollution is a serious issue that has gained the attention of an international audience including policy makers, mill owners, NGOs and academicians. With a production rate of 74 million metric tonnes a year, Malaysia is in dire need for new technologies able to address this mounting problem. While previous research has attempted to modify existing microbial driven organic degradation methods to optimize the organic load removal, new studies recognize the futility in this endeavour as mills are unwilling to change the way they operate and adopt these disruptive technologies. Instead current research aims at tertiary polishing technologies of which photocatalysis has emerged as a forerunner. Recent advances indicate photocatalytic treatment of POME is a viable solution to the problem but for it to be cost effective, future studies should focus on visible light driven processes and new reactor configurations to improve the photocatalytic activity of existing systems.

Keywords: *photocatalysis; POME; solar; UV-visible light; wastewater treatment.*

1. Introduction

Palm oil is the most lucrative and therefore most cultivated commercial crop in Malaysia. As of 2017 Malaysia has a production rate of 21 million metric tonnes of palm oil per annum from a total of 453 operating mills across Peninsular Malaysia, Sabah and Sarawak [1]. This accounts for 39% of world palm oil production and 44% of global palm oil exports [2]. Every tonne of crude palm oil (CPO) produces approximately 2.5 to 3.5 tonnes of palm oil mill effluent (POME) which puts the total POME discharged in 2017 in Malaysia at a staggering 74 million metric tonnes [3, 4]. As such, there has been a growing academic interest in the treatment of palm oil mill effluent (POME) given its far reaching economic and environmental implications. This is also in part due to increasing global scrutiny of environmental issues worldwide which has not overlooked the palm oil industry. Since 2004, the Roundtable on Sustainable Palm Oil (RSPO), a World Wildlife Fund (WWF) initiative has been monitoring closely the environmental impact of the palm oil industry and engaging stakeholders in an effort to reduce pollution deriving from palm oil of which POME is a large contributor [1]. In response to this, albeit at a much later date, Malaysia came up with the Malaysian Sustainable Palm Oil (MPSO) standard in 2014 aimed at achieving some form of middle ground between the RSPO demands and what local palm oil millers were willing to go for [5]. Unfortunately, local millers have been slow to comply with these new regulations and enforcement of these standards has been rather loose primarily due to the lack of practical technologies that can address the problem of POME pollution in an economic wayally. Many methods have been proposed to deal with POME such as adsorption [6], coagulation/flocculation [7-9], microbial fuel cells [10, 11], ultrasonic-assisted membrane anaerobic system (UAMAS) [12], up-flow anaerobic sludge fixed film (UASFF) reactor [13, 14], modified anaerobic baffled bioreactor (MABB) [15], continuous stirred tank reactors (CSTR) [16], up-flow anaerobic sludge blanket (UASB) reactors [17, 18], expanded granular sludge bed (EGSB) reactors [19, 20], membrane filtration [21-23] and advanced oxidation processes (AOPs) [24-31]. It is clear that there is a much larger volume of research dedicated to AOPs than other forms of POME treatment. This is primarily due to the general opinion that the open ponding method employed by more than 85% of palm oil millers is too well entrenched and hence resistant to disruptive technologies that require a complete revamp of the mills' waste treatment processes [19, 21, 32, 33]. Instead much of the research over the past two years have been dedicated to polishing or tertiary treatment methods of the POME final discharge or POME secondary effluent [34]. Of these studies, many have employed the traditional Fenton and photo-Fenton systems [28, 31] but more research has been dedicated to photocatalytic oxidation systems [24, 25, 32, 35-39] as this technology offers a more cost effective solution to the problem of POME pollution. This article therefore presents a review of the new and emerging photocatalytic technologies that have shown promise in terms of their ability to provide a long term solution to the issues plaguing palm oil milling in Malaysia.

2. Characteristics of POME

POME is a thick brownish, acidic, foul smelling colloidal wastewater. POME is a complex mixture containing various cellulosic materials, oils and fatty acids as well as lignin, pectin, carotene and phenols [1, 40]. Despite going through multiple stages of

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biological treatment, POME final discharge still has high colour intensity in excess of 500 ADMI (American Dye Manufacturers Index)[34]. The high colour intensity of POME is due to the high organic loading especially organic substances like lignin, tannin, humic acids and carotene which are known colour causing agents [41]. Since POME final discharge is typically channelled to open waterways, this dark brown colloidal wastewater will significantly affect the water clarity of rivers. Besides being unpleasant aesthetically, reduced water clarity also affects permeability of sunlight which in turn adversely affects aquatic plant life and subsequently endangers fragile aquatic ecosystems. This problem is further compounded by the presence of oil (which forms a layer on the water surface preventing dissolution of oxygen) and the high chemical oxygen demand due to organic species present in the POME waste. All of these create an oxygen diminished aquatic environment that can easily become unsustainable for less robust aquatic life forms creating a cascade of events that lead to catastrophic ecosystem failure [42]. Table 1 presents the average values for various water quality parameters of POME final discharge compared to the Department of Environment (DOE) standard for those same parameters. It is quite clear that chemical oxygen demand (COD), biological oxygen demand (BOD) and colour are parameters that still fall short of DOE regulatory limits. COD particularly falls short by a rather significant margin, indicating this to be a parameter of primary concern and validating the need for tertiary and polishing treatment technologies. However, this great margin may be due in part to nature of these measurements. The values in Table 1 are obtained from averages presented in several studies [1, 34, 43] who have also reported values averaged from other studies. There have been researchers reporting POME final discharge values that are orders of magnitudes higher than the DOE discharge limit. Some researchers have reported values in the range of 3000-5000 ppm of COD which indicates a gross mismanagement of waste treatment [44]. These high values for COD final discharge are an alarming indication of the severe shortcomings of current POME treatment methods.

Table 1: Characteristics of POME Final Discharge [1, 34, 43]

Parameters	Average Values for POME Final Discharge	DOE Discharge Limit
COD (ppm)	300-800	50
BOD (ppm)	200	100
pH	8.4	5.0-9.0
Temperature (°C)	25-30	45
Color (ADMI)	>500	200
TSS (ppm)	130	400
Total Nitrogen (ppm)	127	200
Oil and Grease	NA	50

3. Conventional POME Treatment Technology

Currently a vast majority of palm oil plantations (> 85%) employ aerobic and anaerobic digestion as the preferred method for the removal of much of the COD, BOD and total suspended solids (TSS) in raw POME [45, 46]. This is usually preceded by pre-treating the raw POME via mechanical and physiochemical processes of which coagulation/flocculation followed by sedimentation and filtration are commonly used [43]. The main purpose of pre-treatment is some removal of COD, total suspended solids (TSS) and turbidity prior to mode advanced treatment geared at reducing the bulk of the organic load. For the purpose of pre-treatment, typical coagulants and flocculants used in municipal wastewater treatment such as poly-aluminium chloride (PAC), aluminium sulphate, aluminium chloride, ferric chloride and ferric sulphate are used with PAC and aluminium sulphate (alum) being used more frequently than others due to ease of access and relatively low cost [8, 43, 47]. The pre-treated POME is then subjected to a series of open ponding stages comprising the cooling and mixing, anaerobic, facultative and aerobic ponding. The cooling and mixing pond is set up to prepare the POME mixture for anaerobic digestion. To achieve this, the pH and temperature of the mixture must be carefully regulated as optimal conditions are essential to ensure the sufficient growth and survival of the necessary microorganism species that will be responsible for the bulk organic load removal [48]. Anaerobic ponds are typically 5-7 m deep ponds dug into the ground in which POME is digested by microbes in the absence of aeration or oxygen; hence the name "anaerobic". These ponds can digest large quantities of POME inexpensively but the drawback is the large amount of land required (1-5 ha), high hydraulic retention time (approximately 20 days) and occasional solids accumulation due to inefficiencies in the pre-treatment process [46, 49]. At the anaerobic and facultative ponding stage, methane, carbon dioxide and trace amounts of hydrogen sulphide are released which contribute to green-house gas emissions. Although there has been some limited research into harnessing these gasses for biogas or other fuel purposes, this currently remains an area of untapped potential with limited commercial viability [50, 51]. The facultative and aerobic ponds serve to degrade the remaining organic content in POME such that it meets the water quality parameters stipulated by the DOE. In recent years, there has been research into modifications of the more conventional open ponding favouring instead of various reactor configurations to optimize organic degradation. Of these, noteworthy modifications are the up-flow anaerobic sludge blanket reactor (UASBR) [17, 18, 52], anaerobic fluidized bed reactor (AFBR) [53], expanded granular sludge blanket (EGSB) [19, 20], and up-flow anaerobic sludge fixed film reactor (UASFF) [13, 14]. A further modification of conventional POME degradation methods involves the introduction of membranes either as part of the initial pre-treatment process prior to microbial assisted degradation [23] or integrating microbes and membranes into hybrid system to achieve superior POME degradation [3, 12, 21, 22]. One example of an extremely successful hybridization is the zero liquid discharge pilot plant in Labu, Malaysia which incorporates pre-treatment, patented anaerobic (EGSB) and aerobic (BioAX) microbial systems and membrane separation technology to completely convert all influent POME into harmless by-products that meet the most stringent DOE criteria [54]. However, the cost effectiveness of this zero liquid discharge plant is not discussed at length hence casting doubt on the likelihood of adoption by the vast majority of palm oil mills. Over this same time period, photocatalysis has emerged as a promising new polishing technology geared at further degradation of organics present in POME final discharge.

4. Advances in Photocatalytic Treatment of POME

Photocatalysis refers to the process of degrading organic materials using a suitable semiconductor in the presence of light of an appropriate wavelength and possessing photon energy (hv) that is at least equal to the bandgap of said semiconductor. When these conditions are met, the photon energy from the incident light creates photoexcitation of electrons in the valence band thus promoting them to the conduction band. The electrons in the conduction band and the resulting holes left behind in the valence band are able to react with water molecules to produce oxygen radicals and hydroxyl radicals respectively as represented in the following equations.

$$photocatalyst + hv \rightarrow e_{cb}^{-} + h_{vb}^{+} \quad (1)$$

$$e_{cb}^{-} + O_2 \rightarrow O_2^{-*} \quad (2)$$

$$h_{vb}^{+} + H_2O \rightarrow OH^{*} + H^{+} \quad (3)$$

Both oxygen and hydroxyl radicals are capable of degrading organic matter but of the two, several studies have reported hydroxyl radicals to be more reactive and able to attack a wide range of organic species indiscriminately [24, 25, 35, 38, 39, 55]. A semiconductor material having a bandgap between 1.7 eV and 3.1 eV would be considered visible light active i.e. able to achieve photoexcitation within a visible light regime. In contrast, a semiconductor with a bandgap greater than 3.1eV would be UV light active. Fig. 1 shows the photon energy for various spectral parts.

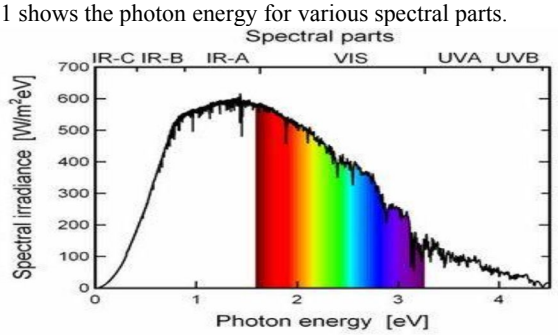

Fig. 1: Photon energy for various spectral components [56]

Currently, a vast majority of research on photocatalysis involves the use of titanium dioxide (TiO_2) given that it is inexpensive, non-toxic and chemically stable [57, 58]. However, titanium dioxide has a band gap of 3.2 eV which means it is only active in the UV region of the spectrum. Since visible light is a much larger portion of the available spectrum and is easily accessible (sunlight comprises more than 46% visible light), much of recent research has been geared towards reducing the band gap of TiO_2 to allow it to be visible light active. Table 2 provides a synopsis of recent research involving photocatalytic treatment of POME. It can be seen from Table 2 that of the TiO_2 remains the photocatalyst of choice for POME remediation albeit there is some deviation from the conventional TiO_2/UV photocatalytic system to accommodate visible light driven photocatalysis. However the studies reporting on visible light driven photocatalysis of POME indicate very feeble results (11-26.8% COD removal) which highlight this as a viable area of new research [35, 38, 39]. All the remaining studies reported using UV light to achieve photomineralization with much higher degrees of success. This is not surprising as UV light is more energetic (> 3.2 eV) and hence capable of producing reactive oxygen species (ROS) with greater oxidative potential. A majority of studies also employed a slurry photo-reactor system in which a powdered form of the catalyst of a specific loading is suspended in the POME liquor with the help of stirring. While this configuration provides intimate contact between the incident light, organic matter and photocatalyst, it suffers from the drawbacks of requiring an additional separation stage after the photocatalytic degradation process as well as impermeability of light at high catalyst loadings especially for fine particles. One study used an innovative approach in which a polyvinylidene fluoride/titanate nanotube (PVDF/TNT) photocatalytic membrane was submerged in a typical Pyrex jacketed photo-reactor. Although there was a clear trade off in terms of % colour removal (67.3%), the improved recyclability of this system, ease of use and lack of need for additional separation processes should be taken into consideration when judging the economic viability of this method [59]. The cylindrical column immobilized photo-reactor was also used and shown to achieve 95% COD removal and 83% colour removal [60] indicating it to be a superior mode of operation especially for TiO_2/UV photocatalytic system. A majority of current research on photocatalytic degradation of POME involve the use of TiO_2 and/or ZnO in either pure or doped forms. Only one study reported the use of an entirely new catalyst, tungsten oxide, WO3 for UV light photodegradation of POME. Typical optimal catalyst loadings for photodegradation of POME range from 0.5 g/L to 1 g/L with irradiation times ranging from 2-8 hrs. High COD and colour removal efficiencies have been reported with the use of UV systems especially with the more tried and tested TiO_2 and ZnO photocatalysts.

Table 2: Summary of recent research on photocatalytic treatment of POME

System	CL[1] (g/L)	t[2] (h)	COD Removal (%)	Colour Removal (%)	ROS[3]	References
ZnO-PEG/UV	0.5	-	92	73	-	[32]
3% wt Nb_2O_5/ZnO/UV	1g/L	4	91.7	100	-	[61]
Ag/TiO_2 /Vis	1.5	8	26.8	-	$OH^{\bullet}$	[35]
ZnO/UV	1.0	4	50	-	$OH^{\bullet}$	[62]
0.5% wt Pt/TiO_2 /UV + Vis	1.0	8	90 (UV) 11 (Vis)	-	-	[38]
0.5%wt PVDF-TNT/UV	-	4	-	67.3	-	[59]
WO_3/UV	0.5	4	51.2	96.2	$OH^{\bullet}$	[37]
0.5% Ag/TiO_2/UV + Vis	1.0	8	25 (UV) 16 (Vis)	-	-	[39]
ZnO/UV	1.0	2	96.0	-	$OH^{\bullet}$	[63]
TiO_2/UV	-	6	95	83	-	[60]

[1]CL: Catalyst loading
[2]t: reaction duration
[3]ROS: Reactive oxygen species

5. Conclusion

Palm oil production is a huge industry in Malaysia and Indonesia and as such is accompanied by a sizable waste management problem. Current methods for treating POME, while economical, are unable to achieve DOE discharge limits for COD, BOD and colour. Many new technologies have been introduced aimed at modifying existing practices to optimize organic load removal but these have not been adopted by the industry due primarily to economic concerns as most of these technologies involve completely changing the way the waste is handled in the mill. Instead new research has been levelled at adding a polishing stage to the POME final discharge so as to remain undisruptive to the entire mill operation. Of these new technologies, photocatalytic degradation has emerged to take the spotlight with reported COD and colour removal efficiencies of upwards of 75% for most studies. There have also been studies attempting to use the more abundantly available visible light spectrum to drive photodegradation but these report more modest findings indicating an area for potential future research. There is also clearly a need to improve modes of operation such that the contact between catalyst-substrate-light can be improved without trading off on cost, ease of operation and need for additional separation stages. Photocatalysis is clearly a revolutionary technology for wastewater treatment but the practical applications of current research do not extend far past the laboratory stage and much more needs to be done before pilot testing and subsequent commercial application can take place.

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