

Review article

A Brief Review of Physico-Chemical Treatment Techniques for Palm Oil Mill Effluent (POME)

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Abstract

Keywords

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It is known that palm oil is one of the top selling raw materials that are widely used in both food and non-food industries. It is also widely used as a feedstock for biofuel production. However, the palm oil industry produces a huge amount of effluent that normally contains undesirable contaminants and suspended solids (SS), with high chemical oxygen demand (COD) and biochemical oxygen demand (BOD) values. Hence, it is imperative to ensure the sustainable management of palm oil production and to prevent serious environmental pollution. Various techniques are available to treat palm oil mill effluent (POME). This review summarizes the compilation of articles published in the period of 2010 to 2020 that focused on the development of treatment techniques for POME. Academic databases including Scopus, ScienceDirect, Web of Science, and PubMed were used to search for related articles. This review discusses the performance and highlights the optimum operating conditions for each chemical treatment technique. This review is not only relevant to researchers who work on the advancement and innovation of remediation technologies for POME, but it is also significant for researchers who deal closely with environmental pollution, water resource management, environmental protection, and flora and fauna conservation.

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1. Introduction

Large scale palm cropping can be found in Southeast Asian countries, particularly Indonesia, Malaysia and Thailand, and are typically found in rainy tropical lowland regions located within 10 degrees north or south of the equator [1]. Based on the United States Department of Agriculture database, the total production of crude palm oil was reported as 43,500 and 19,900 thousand tonnes for Indonesia and Malaysia, respectively, in 2020 [2]. The demand for palm oil-based products is expected to increase proportionally with population growth, whereby more than half of the products available on the market, especially products in the food and personal care areas, are produced from palm oil. As a matter of fact, an increment in the physical production of palm oil has caused a significant increase in the amount of liquid waste known as POME.

Even though the rules related to effluent discharge limits for the oil palm industry were strictly set and are implemented by the government of each palm oil exporting country, in many cases, palm oil factories do not comply with the standard discharge limits. This issue can be related to the difference in POME properties. It is known that POME characteristics may differ and be greatly influenced by several factors such as treatment method, period of treatment, batch processing technique, quality or type of fruit, chopping season, and weather condition (drought and monsoon season) [3, 4]. It has been estimated that the production of each tonne of crude palm oil (CPO) will normally produce an average of 0.67 tonnes of POME [5]. With the global production of 72,271,000 tonnes of palm oil in 2019, approximately 48,422,570 tonnes of POME were generated worldwide.

In recent years, various single or hybrid methods have been developed in order to alleviate environmental pollution caused by the palm oil industry. Therefore, in this review, an extensive list of physico-chemical treatment techniques such as coagulation/flocculation, electrocoagulation, membrane filtration, advanced oxidation process, photocatalytic degradation, ozone-based process and Fenton-based process for POME are compiled. The advantages, disadvantages and operational cost for selected physico-chemical treatment techniques are summarized in Table 1.

2. Chemical Treatment Techniques for POME

2.1 Coagulation/flocculation

Coagulation/flocculation is an essential and effective step used to separate suspended solids in the remediation of industrial effluent. The purpose of this process is to destabilize colloidal material using coagulants agents by neutralizing the negative charges on non-settable solids. Once the neutralization process is completed, the target pollutant will clump together in microflocs. Flocculant agents are then added to form bridges between the microflocs to increase the particle size and strengthen the flocs. Flocs are allowed to reach their optimum size and strength prior to sedimentation for settling out particles in suspension from water. Following stabilization and sedimentation, separation processes like filtration, straining or floatation are used [6].

Aluminum sulphate (alum), ferric chloride, polyaluminum chloride (PAC) and polyacrylamide (PAM) are coagulants and flocculants that are widely used in wastewater treatment. As presented in Table 2, coagulants and flocculants demonstrated excellent potential in removing TSS and turbidity of POME with up to 85% reduction. It is apparent that the application of bio-coagulants and flocculants particularly at high dosage rates caused a similar effect to TSS and turbidity, producing a significant reduction percentage. TSS consists of small solid particles that remain in suspension in water, and are made up of organic and inorganic materials such as algae, silt and sediment. In general, the higher the TSS value, the higher the turbidity value of POME is.

Table 1. Advantages, disadvantages and operational costs for selected physico-chemical treatment techniques [6-8]

Technique	Advantages	Disadvantages	Operational Cost*
Coagulation/flocculation	<ul style="list-style-type: none"> • Easy and economically feasible technique • Efficient and high capacity treatment method for different types of particle 	<ul style="list-style-type: none"> • Produce toxic sludge (depending on the type of coagulant used) • Poor removal of arsenic 	Polyaluminum chloride (0.3805 USD/ m ³) for raw water [9]
Electrocoagulation	<ul style="list-style-type: none"> • Produce minimal sludge and are non-hazardous • Required a low energy consumption 	<ul style="list-style-type: none"> • Electrodes have a limited lifetime • Required an active fine-tuning 	Al electrode (1.19 US/m ³) for metalworking fluid wastewater [10] Al electrode (0.86 \$/m ³) for domestic wastewater [11]
Membrane filtration	<ul style="list-style-type: none"> • Small footprint required • Higher selectivity for small molecular particles 	<ul style="list-style-type: none"> • Expensive and high energy demand • Produces concentrated sludge that causes membrane fouling 	Reverse osmosis polymeric membrane (RM 7.03/m ³) for POME treatment [12]
Advanced oxidation process	<ul style="list-style-type: none"> • Required small footprint with rapid reaction rates • No sludge production • Easily automated and controlled 	<ul style="list-style-type: none"> • Relatively high operating and maintenance costs • Complex chemistry tailored to specific contaminants • Removal of residual peroxide may need to be considered. 	1.34 USD/1000 gallons treated [13]

*The cost might be different based on the type of chemical and the concentration of wastewater

In recent years, the use of metal coagulant/flocculant for POME purification has received great attention from researchers. The current trend in metal coagulant/flocculant research is mainly focused on enhancing the coagulant/flocculant properties by modification of process and finding solutions. In addition, it is aimed at overcoming the drawbacks related to metal leakage. For example, ferrous sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$) is one of the main by-products obtained during the production of titanium dioxide. Hossain *et al.* [14] investigated the ability of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ waste in eliminating TSS, COD and BOD from POME. They reported that $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ waste coagulant was able to eliminate TSS, COD and BOD up to 84%, 75% and 80%, respectively, during a single stage of the coagulation treatment. However, the efficiency of coagulation or/and flocculation was greatly affected by several important factors such as effect of pH (which determines the solubility of metal hydroxide species and charge density), temperature (which determines the dispersion rate or kinetic energy of the adsorbate towards the boundary line of coagulants) and adsorbent dosage (which determines the zeta potential of the surface of colloidal and suspended particles). They noted

that the optimum conditions of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ waste occurred at pH 7, temperature of 70°C and coagulant dosage of 2.0 g/l with 45-6 min coagulation time.

Additionally, Noor *et al.* [15] evaluated the performance of magnetic cellulose grafted with polyacrylamide (PAM-g-MagCell) via microwave radiation reaction, as a novel flocculant. The optimum operation and removal percentage of PAM-g-MagCell are given in Table 2. They found that the carbon and hydrogen content of the new grafted flocculants was increased, which enhanced the flocculation performance efficiency. This was related to the presence of PAM long branching chains on the MagCell backbone, which enlarged the molecular weight, radius of gyration and branching of grafted polymer. Moreover, the grafted polymer had a high density charge to neutralize the negative charge of the pollutant that was present in the palm oil wastewater. They also pointed out that by using magnetic material in coagulation/flocculation, the sedimentation time could be cut with the aid of external magnetic material to settle down the magnetic flocs. The operational cost was estimated as USD 92.57 per m^3 for PAM-g-MagCell and USD 166.90 per m^3 for conventional flocculant PAM.

2.2 Electrocoagulation

Theoretically, electrocoagulation and chemical coagulation are used for similar purposes; however, they differ in terms of dosing method, i.e. in the case of electrocoagulation, electric current must be supplied between metal electrodes for generation of in situ coagulant dose [20]. The dissolution of the sacrificial anode, commonly made of aluminum or iron, involves hydrolysis that destabilizes pollutants generating flocs, which are carried out to the surface by the hydrogen bubbles forming at the cathode, to then be skimmed off. For example, Aris *et al.* [5] observed the highest removal efficiency of color from a POME sample (up to 64%) at pH 5, 20 V applied voltage, 9.5 cm plate gap and at 8-h operating time by electrocoagulation process using an aluminum electrode.

Meanwhile, Nasrullah *et al.* [21] studied the effectiveness of a steel wool anode for treatment of raw POME. The results revealed that the treatment with the steel wool anode yielded removals of COD, BOD, SS and turbidity up to 94.29%, 96.87%, 89.31% and 97.80%, respectively, under the optimum operating parameters: 60 min of electrolysis time, 20 mm of inter-electrode distance, initial pH of 4.6 and 20 A of current intensity. They emphasized that the treatment efficiency was influenced by several factors including pH, current intensity, inter-electrode distance and electrolysis time, that might determine the size and structure of the flocs formation. They also mentioned that it is important to form bulky, large and dense flocs that have a high tendency to settle at the bottom of the reactor resulting in maximum pollutant removal by precipitation.

In another study, Nasrullah *et al.* [22] designed and investigated the optimum operating parameters (electrolysis time, inter-electrode distance and initial pH) in one factor at one time study. In their study, four pieces of steel wool electrode were arranged in vertical orientation with a monopolar series arrangement that was dipped in a POME sample. Results obtained from this innovation indicated that SS, COD, and BOD were effectively reduced by 96%, 95%, and 94%, respectively. The optimum parameters identified from the study for electrolysis time, current intensity, inter-electrode distance and initial pH were found to be 50 min, 20 A, 10 mm and pH 4, respectively. In the electrocoagulation process, gas bubbles will simultaneously be generated in the process in order to eliminate the pollutant by floatation. However, they noted that an excess in gas bubbles production reduced the treatment performance due to the reduction in active specific area of the electrode.

Table 2. POME treatment by coagulation/flocculation

Coagulant/ Flocculant	Characteristics of Materials	Mixing Rate	Operation Conditions	Performance	Ref.
Zeolite	Particle size: 63 μm	Rapid: 250 rpm (3 min) Slow: 30 rpm (30 min)	Settling time: 60 min pH: 4.51 (raw POME) Dosage: 1000mg/l Temperature: ambient	TSS: 96.82% Turbidity: 91.98% NH ₃ -N: 93.54% COD: 61.89% O&G: 95.67% Color: 86.5%	[16]
Chitosan	NR	Rapid: 250 rpm (3 min) Slow: 30 rpm (30 min)	Settling time: 60 min pH: 4.51 (raw POME) Dosage: 400 mg/l Temperature: ambient	TSS: 98.95% Turbidity: 98.35% NH ₃ -N: 95.88% COD: 68.31% O&G: 94.86% Color: 96.44%	[16]
FeCl ₃	NR	Rapid: 250 rpm (3 min) Slow: 30 rpm (30 min)	Settling time: 60 min pH: 4.51 (raw POME) Dosage: 1000 mg/l Temperature: ambient	TSS: 99.09% Turbidity: 95.99% NH ₃ -N: 93.74% COD: 72.04% O&G: 95.73% Color: 86.51%	[16]
FeSO ₄ .7H ₂ O	Iron content: 18.6 % Purity: > 92%	Rapid: 200 rpm (1 min) Slow: 50 rpm (60 min)	Settling time: 60 min pH: 5 Dosage: 2 g/l Temperature: 70°C	COD: 75% BOD: 80% TSS: 84%	[14]
Alum	NR	Rapid: 150 rpm (5 min) Slow: 10 rpm (15 min)	Settling time: 30.32 min pH: 3.79 Dosage: 2.57 g/l	TSS: 92.02% COD: 54.55%	[17]
Magnetic cellulose	Optimum ratio: 1:1 (w/w) (magnetite: cellulose)	Rapid: 200 rpm (3 min) Slow: 30 rpm (15 min)	Settling time: 30 min	Turbidity: 74.50% Color: 63.90% TSS: 77.20% COD: 55.70%	[18]

Table 2. POME treatment by coagulation/flocculation (contd.)

Coagulant/ Flocculant	Characteristics of Materials	Mixing Rate	Operation Conditions	Performance	Ref.
Cellulose	NR	Rapid: 200 rpm (3 min) Slow: 30 rpm (15 min)	Settling time: 30 min Dosage: 1 g	Turbidity: 52.85% TSS: 48.90% Color: 13.14% COD: 12.55%	[18]
Polyacrylamide grafted magnetic cellulose	Magnetisation saturation: 55.31 emu/g	Rapid: 200 rpm (3 min) Slow: 30 rpm (15 min)	Settling time: 30 min pH: 8 Dosage: 1.5 g/l Temperature: 27-30°C	Turbidity: 88.62% Color: 91.76% TSS: 82.97% COD: 53.23%	[15]
Ferric chloride + polyDADMAC	NR	NR	Settling time: 120 min pH: 8 FeCl ₃ dosage: 5800 mg/l polyDADMAC Dosage: 8 mg/l	Color: 83.4%	[19]

NR not reported, BOD biochemical oxygen demand, COD chemical oxygen demand, TSS total suspended solids, O&G oil and grease, NH₃-N ammoniacal nitrogen

The effects of three different electrodes (aluminum, iron and steel) with three arrangement types of electrode (monopolar series, monopolar parallel and bipolar) on POME treatment were also studied by Nasrullah *et al.* [23]. The studies were carried out in order to evaluate the removal efficiency of COD, BOD and SS regarding electrode type and design. They conducted the experiments for 120 min by using 5 A initial current intensity and 20 mm of inter-electrode distance. The effectiveness of electrode arrangement on POME treatment was reported to be in the order of monopolar series (65% COD, 62% BOD, 60% SS) > monopolar parallel (61% COD, 58% BOD, 59% SS) > bipolar (56% COD, 56% BOD, 51% SS), while the electrode material order was steel (74% COD, 70% BOD, 66% SS) > iron (72% COD, 67% BOD, 63% SS) > aluminum (65% COD, 62% BOD, 60% SS) for 120 min of electrocoagulation process. It was also noted that the treatment by monopolar series design produced a high amount of anodic dissolution with lower consumption of current intensity.

Nwabanne *et al.* [24] evaluated the effectiveness of electrocoagulation with iron electrodes in removal of total dissolved suspended particles. They reported that under optimum operation conditions at current density of 3 Amps, electrocoagulation time of 60 min, pH of 10, and temperature of 50°C, 65% removal of total dissolved suspended particles was obtained. In another study, Bashir *et al.* [25] used hydrogen peroxide as an oxidizing agent to boost the production of hydroxyl radicals thus enhancing the electrocoagulation process for post treatment of POME. Based on the analysis, the optimum conditions for current density, contact time, initial pH and concentration of H₂O₂ were 40.21 mA/cm², 45.67 min, pH 4.4 and 0.5 g/l, respectively, and under these conditions, removals of COD, color and TSS were 71.3%, 96.8% and 100%, respectively. They also found that the electrocoagulation process with the presence of oxidizing agent could cut down the reaction time and current density by 30% and 28.2%, respectively, for POME treatment.

2.3 Membrane filtration

Membrane technology is a pressure driven separation process that has become one of the most competitive alternative technologies in wastewater treatment. In the membrane process, the separation system involves the separation of single inflow stream into two outflow streams known as the permeate and retentate [26]. Generally, there are four categories of membranes basically used in water treatment processes, namely microfiltration (MF), nanofiltration (NF), ultrafiltration (UF) and reverse osmosis (RO), which are defined based on the pore size range of the membranes. POME treatment using membrane technology is still mostly at the lab scale due to the high suspended solid content of POME which normally causes fouling and high flux on the membrane during the treatment process.

The removal efficiency of three different types of commercial membranes, namely NF270, UF 10 kDa and UF 30 kDa for total organic carbon (TOC) and total nitrogen (TN) from POME was studied by Amat *et al.* [27]. From their study, NF270 membrane was reported to remove almost 100% and 84% of TOC and TN, respectively, which was a higher result than 82.1% and 65.5% for the UF 10 kDa membrane, and 63.0% and 56.1% for the UF 30 kDa one. Although the NF membrane demonstrated satisfactory separation performance, the permeate water flux was reported at 6.6 l/m², which was lower than the other two UF membranes (10.7 l/m² for 30 kDa and 9.7 l/m² for 10 kDa). This was probably due to the dense polyamide surface that was formed on the top layer of the microporous substances of NF membrane. They also reported that NF membranes tended to cause fouling at high pressure where the foulants blocked and narrowed the pores of the membrane, increasing water transport resistance. For reusability, they recommended performing a chemical cleaning process together with periodic backflushing in order to recover water flux and remove foulants that were adhered to the membrane matrix.

Ho *et al.* [28] made an advancement in membrane technology to treat POME by adding graphene oxide (GO) and multi-walled carbon nanotubes (MWCNTs) into polyvinylidene fluoride (PVDF) membrane matrix through a phase inversion method. A response surface methodology using central composite design was applied to determine the synergistic combination of the nanocomposite conductive membrane formulation, and to identify the optimal operating parameters for the membrane process based on internal time and time of electric field of the electrically-enhanced filtration. It was proposed in the cubic model of normalized flux for continuous mode study that the optimum concentration formulation of nanocomposite conductive membrane with CNT nanofiller was 4.22 wt.% with 221.00 V/cm of electric field. The interval time reported of 32.00 min and 6.00 min of application time for intermittent mode study produced the highest normalized flux. In their study, experiments showed that the values of the normalized flux for optimum conditions for continuous study mode and intermittent study mode were reported as 0.7778 and 0.7983, with smaller percentages of error of 9.94% and 11.20%, respectively.

L-Histidine doped TiO₂-CdS photocatalytic nanocomposite merged with a polyethersulfone (PES) nanofiltration membrane was synthesized by Zangeneh *et al.* [29]. This innovation was aimed to enhance the membrane performance related to membrane fouling. The results show that the membrane with 0.5 wt.% of NPs gave high permeation flux at 34.7 kg/m²h with complete COD removal and 99% of flux recovery ratio during POME filtration with concentration of COD at 1000 mg/l, 150 l/h of feed flow rate, and pressure set at 5 bar. The photocatalytic degradation of organic compounds and superhydrophilicity of the polymeric membrane structure at 0.5 wt% of NPs was reported to minimize the membrane fouling. Besides, they also observed that with the increase in cross flow velocity, separation capacity and self-cleaning properties of the membrane were also increased. They also noted that the hydrophobicity property and surface roughness are essential to prevent membrane fouling.

The evaluation of two different thicknesses of hydrophilic thin films of hydroxypropyl methacrylate (HPMA) 50 and 100 nm onto a PVDF membrane surface was studied by Subramaniam *et al.* [30] using an initiated chemical vapor deposition method, utilizing ditert-butyl peroxide as radical initiator. After HPMA was coated onto the PVDF surface, the hydrophilicity was reported to improve and the surface roughness was reduced. The results indicated a membrane flux and rejection for 100HPMA-PVDF at 50.8 l/m²h and 83.1%, respectively. Besides, they also studied the long-term flux and rejection potential of the coated membrane and showed that 100HPMA-PVDF could maintain the stability at elevated permeation level. From their observation, after four filtration cycles, the rejection properties showed only 3.5% of flux drop, and did not show any cake layer formation that could cause membrane fouling.

Table 3 summarizes recent studies on the application membrane filtration techniques for POME treatment as well as their performance capacities. In terms of operational costs for membrane filtration techniques, Ahmad *et al.* [31] made a comparative study on type and orientation of membrane system for POME treatment. They reported that the orientation of two RO polymeric membrane systems could meet the permissible limits for POME with total cost of MYR 7.03 per m³, followed by (UF polymer + RO polymer) membrane at MYR 23.64 per m³ and (UF ceramic + RO polymeric) membrane system at MYR 115.11 per m³ at optimum operating conditions (1 USD = 3.50 MYR as at 22 May 2009).

Table 3. POME treatment by membrane filtration

Membrane	Membrane Process	Membrane Characteristics	Operation Conditions	Performance	Ref.
UF 30 kDa	Ultrafiltration	Water permeability: 99.1±1.4 l/m ² hbar Contact angle: 56.0±2.1° Roughness: 20.3 nm	Pressure: 5 bar	Permeate flux: 10.7 l/m ² h TOC: 63% TN: 56%	[27]
UF 10 kDa	Ultrafiltration	Water permeability: 18.5±0.8 l/m ² hbar Contact angle: 66.2±1.4° Roughness: 14.6 nm	Pressure: 5 bar	Permeate flux: 9.7 l/m ² h TOC: 82% TN: 66%	[27]
NF270	Nanofiltration	Water permeability: 14.2±0.5 l/m ² hbar Contact angle: 14.1±5.9° Roughness: 3.8 nm	Pressure: 10 bar	Permeate flux: 6.6 l/m ² h TOC: 100% TN: 84%	[27]
Graphene oxide (GO) and multi-walled carbon nanotubes (MWCNTs) into polyvinylidene fluoride (PVDF)	NR	Thickness: 200 µm Roughness: 24.70-31.79 nm Carbon nanomaterial conc.: 4.22 wt%	Electric field: 221.00 V/cm Interval time: 32 min Application time: 6 min	Normalized flux Continuous mode: 0.7778 Intermittent mode: 0.7983	[28]
L-Histidine doped TiO ₂ -CdS/PES nanocomposite membrane	Nanofiltration	Contact angle: 47° Pure water flux: 42.1 kg/m ² h Roughness: 11.18 nm	Pressure: 5 bar Stirring rate: 400 rpm Feed conc.: 1000 mg/l Flow rate: 150 l/h Treatment time: 150 min	COD: 100% Colour: 100% Flux recovery: 79.4% Permeate flux: 34.7 kg/m ² h	[29]
Hydroxypropyl methacrylate thin film coating on PDVF hollow fiber membranes	Ultrafiltration	Porosity: 63.762±0.13 g/cm ³ Contact angle: 23.63±3.04° Thickness: 142.7±31.5 nm Roughness: 34.14 nm Pure water flux: 110.49 l/m ² h	NR	Permeate flux: 39.46±0.77 l/m ² h for lignin and 21.73±0.77 l/m ² h for tannic acids Permeate rejection: 83.10±0.53%	[30]

Table 3. POME treatment by membrane filtration (contd.)

Membrane	Membrane Process	Membrane Characteristics	Operation Conditions	Performance	Ref.
Sodium based lignosulfonate	Forward osmosis	Active area: 8.4 cm ²	Pressure: 4.67 bar Stirring time: 100 rpm Flow rate: 100 ml/min Pressure: 0.2 bar Feed conc.: 150 g/l	Permeate flux: 2.4 LMH	[32]
Silver nano-particle coated hydroxyapatite nano-composite membrane	Ultrafiltration	Thickness: 250 µm Water content: 90.81% Contact angle: 65.1° Porosity: 0.901 Intrinsic resistances: 14.0 kPahm ² /l	Pressure: 8 bar Temperature: 26°C Permeate was collected every 10 min	BOD: 89.3% TSS: 91.3%	[33]
K-B-N triple doped-TiO ₂ /PES membrane	Nanofiltration	Contact angle: 52.6° Pure water flux: 37.3 kg/m ² h	Pressure: 4.5 bar Stirring rate: 300 rpm Feed conc.: 5000 mg/l Treatment time: 90 min	Flux recovery: 71.7% Permeate flux: 19.4 kg/m ² h Color: 98% COD: 88.7%	[34]
Zinc iron coupled oxide (ZIO) incorporated into polyvinylidene fluoride (PVDF)	Ultrafiltration	Layer thickness: 142.05 nm Roughness: 57.745 nm Surface area: 28.76 m ² /g Pore size: 18.63 nm Porosity: 44.62%	NR	Permeation flux: 40.14 Lm ² /h Color: 69.53% COD: 60%	[35]

2.4 Advanced oxidation process (AOPs)

AOPs are treatment technologies aimed at eliminating and degrading recalcitrant organic contaminants into biodegradable compounds through reaction with hydroxyl radicals. The general principle of AOPs is the production of extremely reactive oxygen species, namely hydroxyl radicals ($\text{HO}\cdot$), hydrogen peroxide (H_2O_2) and superoxide ($\text{O}_2\cdot^-$) to oxidize organic contaminants and to mineralize products such as CO_2 , H_2O and inorganic ions. Photocatalytic degradation, the Fenton-based reaction and ozone-based process are some types of advanced oxidation process that will be discussed in this section.

2.4.1 Photocatalytic degradation

Photocatalytic degradation is a process involving photochemical reactions to produce reactive species through irradiation of a catalyst with light source. When catalysts absorb light at appropriate wavelengths and possess photon energy, transition of electrons takes place from the valence to the conduction band and leave gaping holes behind in the valence band. The holes in valence band (h_{ν}^+) are then able to react with water molecules to produce $\text{HO}\cdot$ radicals. Meanwhile, photo-generated electrons in the conduction band (e_{cb}^-) react with molecular oxygen to create superoxide radicals ($\text{O}_2\cdot^-$) and undergo further reaction to form $\text{HO}\cdot$ radicals. Both oxygen and hydroxyl radicals that are produced are capable of degrading organic matter in wastewater [36]. Several types of photocatalysts that have been used for treating POME wastewater are shown in Table 4.

The effects of different synthesis routes (co-precipitation and auto-combustion) and calcination temperature (550°C and 700°C) in the preparation of novel photocatalyst CaFe_2O_4 was investigated by Charles *et al.* [37]. They evaluated photocatalytic degradation of POME under visible light irradiation. By comparing four samples of CaFe_2O_4 , the sample prepared using co-precipitation was reported to demonstrate a higher S_{BET} and pore volume when compared to the sample prepared using the auto-combustion method. The highest photocatalytic degradation ability with 56% of COD removal after 8 h and 49% of COD removal after three consecutive cycles was recorded for CaFe_2O_4 photocatalyst prepared by co-precipitation at $550^\circ\text{C}/\text{CP550}$ (S_{BET} $27.28 \text{ m}^2/\text{g}$). However, for the 700°C co-precipitation sample, the S_{BET} was drastically reduced to $9.73 \text{ m}^2/\text{g}$ due to the growth of crystallite size with increase in sintering temperature. The excellent performance of CP550 was primarily related to its electronic structure with a band gap of 1.52 eV of catalyst that was lower enough to be activated by visible light but high enough to produce hydroxyl radicals with sufficient oxidation power to degrade the recalcitrant pollutant in POME successfully.

Sin *et al.* [38] investigated $\text{WO}_3/\text{Nb}_2\text{O}_5$ -decorated hierarchical porous ZnO microspheres (WO_3/ZnO and $\text{Nb}_2\text{O}_5/\text{ZnO}$) that were prepared via a facile surfactant-free method. The examination of photocatalytic ability showed that both $\text{Nb}_2\text{O}_5/\text{ZnO}$ and WO_3/ZnO composites demonstrated excellent photodegradation of POME at 91.7%, and 68.3%, respectively, after 240 min irradiation under UV ray. This was due to the success in avoiding charge carrier recombination and enhanced interfacial charge transfer. They also explained that more photogenerated electrons and holes were generated and participated in the redox reactions to form highly reactive hydroxyl radicals ($\text{HO}\cdot$), and thus the photocatalytic activity was significantly improved.

To enhance the effectiveness of photocatalyst, Ghazali *et al.* [39] synthesized a photocatalyst derived from cockle shell waste and doped it with lanthanum (La). The introduction of dopants established energy states in the band gap, and thus increased the trapping recombination rate and the surface area of photocatalyst. These increments were reported to increase the degradation of the photocatalytic system. They reported that La doping cockle shells waste showed

Table 4. POME treatment of AOPS by photocatalytic degradation oxidation

Type of Catalyst	Source of Energy	Characteristics of Catalyst	Operation Conditions	Performance	Ref.
CaFe ₂ O ₄	Visible light (500 W Xenon lamp with light intensity 50 mW.cm)	Specific surface area: 27.28 m ² /g Average pore size: 11.2 nm Average pore volume: 0.077 cm ³ /g	Catalyst loading: 0.75 g/l Irradiation time: 8 h	COD: 69%	[37]
WO ₃ /ZnO	UV light (15 W)	NR	Catalyst loading: 1.0 g/l Irradiation time: 240 min	Photocatalytic degradation: 68.3%	[38]
Nb ₂ O ₅ /ZnO	UV light (15 W)	NR	Catalyst loading: 1.0 g/l Irradiation time: 240 min	Photocatalytic degradation: 91.7%	[38]
La/CaO	UV light	Specific surface area: 4.08 m ² /g Average pore diameter: 85.22 nm	Catalyst loading: 3 g/l Irradiation time: 240 min	Photocatalytic degradation: 54.09%	[39]
Pt/TiO ₂	Visible (100 W Xenon lamp) /UV light (100W)	Specific surface area: 11.85 m ² /g Average pore diameter: 23.05 nm	Catalyst loading: 1.0 g/l Irradiation time: 8 h	Degradation conversion: 90%	[40]
Ag/TiO ₂	Visible (100 W Xenon lamp) /UV light (100 W)	Specific surface area: 9.23 m ² /g Average pore diameter: 22.90 nm	Catalyst loading: 1.0 g/l Irradiation time: 8 h	Degradation conversion: 85%	[41]
WO ₃	UV light (100 W)	Crystal size: 45.6 nm	Catalyst loading: 0.5 g/l Irradiation time: 240 min	Photocatalytic degradation: 51.15% Color: 96.21%	[42]
Nb ₂ O ₅ /ZnO	UV light	NR	Irradiation time: 240 min	Photocatalytic degradation: 91.7% Color: 100%	[43]
PVDF-TNT	UV light (8 W)	Effective surface area: 188.95 m ² /g Cumulative pore volume: 0.26 m ² /g Average pore width: 52.17 Å	pH: 3 Irradiation time: 240 min	Color: 67.3%	[44]

Table 4. POME treatment of AOPS by photocatalytic degradation oxidation (contd.)

Type of Catalyst	Source of Energy	Characteristics of Catalyst	Operation Conditions	Performance	Ref.
ZnO-PEG	UV light (15 W)	Particle size: 25- 150 nm	pH: 6.5 Catalyst loading: 0.5 g/l Dilution of POME: 50%	Color: 84% Turbidity: 94% BOD: 99 mg/l COD: 94%	[45]
TiO ₂	UV light (20W)	Average particle size: 100nm	pH: 5.5 Catalyst loading: 4.84 mg Reaction time: 42.86 min	COD: 66.5% BOD: 68.18% Color: 73.17%	[46]
ZnO	UV light (light intensity 5400 $\mu\text{W}/\text{cm}^2$)	Specific surface area: 18.4 m ² /g Average diameter: ~280 nm Average length: ~ 17 μm	pH: 8.7 Catalyst loading: 1.0 g/l Irradiation time: 120 min	COD: 96.0%	[47]

NR not reported, *BOD* biochemical oxygen demand, *COD* chemical oxygen demand, *TSS* total suspended solids, *O&G* oil and grease, *RSM* response surface methodology

a surface area and pore diameter of 4.08 m²/g and 85.22 nm, higher than that of 3.66 m²/g and 28.75 nm for the precursor cockle shells waste, respectively. Besides, they evaluated the optimum stirring speed related to COD removal efficiency of POME under UV light irradiation of 240 min. The results revealed that the maximum stirring speed was 300 rpm with photocatalytic degradation activity efficiency at 54%.

2.4.2 Ozone-based process

Ozonation is an effective oxidation technology for the removal of inorganic and organic substances as well as microbiological contaminants including bacteria and viruses from water. Ozone contains three oxygen atoms in its structure and is an unstable gas that can either directly react with organic compounds or first be decomposed to hydroxyl radicals. However, some compounds such as amides, aliphatic carbon chains, and nitroso are hardly oxidized due to their low reactivity with ozone. Therefore, hydrogen peroxide can be introduced to improve the oxidation of ozone-resistant compounds, whereby it acts to increase OH fabrication by accelerating ozone decomposition [48].

Ahmad [49] experimentally investigated the ability of ozone to treat POME containing high amount of organic matter. Their preliminary study demonstrated that the volatile fatty acids (VFA) and BOD were reduced by up to 70% and 89%, respectively, while the COD degradability efficiency was reduced by 90% within 70 h of ozonation at 5 g O₃/l. Besides that, the BOD/COD ratio of 0.871 was also reduced to 0.110 under anaerobic conditions. Furthermore, they highlighted that ozonated POME was faster to biodegrade (with 73.9% efficiency) than non-ozonated POME.

Chaiprapat and Laklam [50] found that treatment by the ozonation process increased the ratio of BOD/COD by 37.9% with slight COD loss of 3.3% at an ozone dose of 0.4 g O₃/l POME. The ozonated POME showed a total chemical oxygen demand (TCOD) removal of 64.2±2.3% and 54.8±2.0%, which were obtained at hydraulic retention times of 10 and 7 days, respectively, at organic loading from 6.52 to 11.67 kg COD/m³/d. The reduction was due to the breakdown of long-chain fatty acids into smaller pieces. They emphasized that the POME treatment by pre-ozonation was not only capable of removing organic matter but also had excellent system stability that could be achieved without pH adjustment.

Facta *et al.* [51] studied the removal of color in treated POME and successfully achieved obvious color changes by ozonation from 100 to 20 mg/l Pt.Co when ozone injection was set up to 800 ppm within 5 min. This was a great achievement because water with color reading below 40 mg/L Pt.Co. can be considered as clear water. By extending the time of ozone treatment up to 15 min, the color of POME was reduced to below 15 mg/l Pt.Co. Their ozonizer was able to produce ozone concentration more than 1800 ppm at 1.23 kVrms within 1 litre per min flow rate at ambient temperature and normal atmospheric pressure.

The highest removal (44%) of COD at POME concentration of 15,000 mg/l under mesophilic condition with the optimum pH of 6 at 37°C was reported by Pisutpaisal *et al.* [52]. They found that ozone was able to break down the volatile fatty acid to acetic and butyric acids as well as a small number of valeric and propionic acids. The removal of COD efficiencies pattern was proportionally increased with the increase of POME concentration from 5,000 to 15,000 mg/l, however at concentrations ranging from 15,000 to 30,000 mg/l the COD removal decreased due to partial ozonation of POME, thereby improving the biodegradability of soluble substrate in the POME.

Tanikkul *et al.* [53] conducted ozonation of pre-treated POME under mesophilic condition and figured out pretreatment by ozone enhanced the biodegradability process rate of the complex organic matter in POME at concentration range of 3,000-15,000 mg COD/l. They reported that the ratio of BOD/COD could be increased to 0.76 after 27 min, with an optimum COD: Ozone (w/w) of 94. This finding indicated a progressive increase in biodegradable reaction of the ozonated solutions. The agglomeration of volatile fatty acid occurred gradually when the POME

concentration increased from 15,000 to 40,960 mg COD/l, which can be toxic and hinder the activity of methanogens in methanogenesis.

2.4.3 Fenton-based process

Theoretically, highly oxidative hydroxyl radicals are produced during the reaction of hydrogen peroxide (H_2O_2) with Fe^{2+} ion under irradiation with sunlight or an artificial light source. The hydroxy radicals attack and degrade harmful organic pollutants into carbon dioxide and water. There are two categories of the Fenton-based process, namely the homogeneous and heterogeneous Fenton processes [54]. However, there are several limitations associated with the homogeneous Fenton process that led to the use of the heterogeneous Fenton process. These have been widely studied by researchers and mainly are: (i) narrow pH range, (ii) generation of a large amount of secondary iron sludge, and (iii) the requirement of large catalyst dosage [55]. Therefore, the heterogeneous Fenton process received great attention for its value in alleviating organic contaminants in wastewater, and especially for POME remediation. However, most studies related to the heterogeneous Fenton process have been conducted at the lab scale and have drawn a number of limitations and disadvantages from the industrial viewpoint such as relatively complicated synthetic conditions, complex synthesis routes and high costs in synthesis of catalysts material, and intricate design treatment process [54].

Lim *et al.* [56] compared the performance of the conventional Fenton process and the electro-Fenton process towards the deprivation of COD. The outcomes showed that the optimum conditions of retention time, concentration of H_2O_2 and FeSO_4 catalyst for achieving better degradation of organic matter in POME were found to be in 4 h, 0.05 M and 0.10 M, respectively. The results indicated that up to 94% of the maximum COD removal capability was achieved by the electro-Fenton under 1.5 V of power input as compared to the conventional Fenton process, which had 48% removal from the same initial concentration of COD in the POME solution.

Meanwhile, Gamaralalage *et al.* [57] investigated the degradation behavior of raw POME by applying Fenton oxidation and identified the organic compound content. The existence of the predominant carboxylic acid, acetic acid, which was difficult to be decomposed naturally, was the main factor behind the high TOC value in POME. The findings showed that the highest TOC reduction, 91%, was achieved with a TOC: H_2O_2 : Fe^{2+} molar ratio of 1:3.7:0.6 within 90-180 min under the reaction conditions of pH 3 at 25°C with an initial concentration of TOC at 0.19 g/l. They also noted that an excess of H_2O_2 might have a negative impact on the degradation of organic matter due to the self-decomposition and scavenging of $\text{HO}\cdot$. The Fenton reaction is favored in acidic condition (pH 3), possibly due to the presence of higher amounts of the $\text{Fe}(\text{OH})^{2+}$ and $\text{Fe}(\text{OOH})^{2+}$ species that are generated during the process. In addition, analysis of phosphorus content revealed that 99.9% of total phosphorus (TP) was removed from the liquid phase within 15 min.

Kanakaraju *et al.* [58] treated POME with the aim to reduce COD level using solar and indoor photo-Fenton techniques by varying the Fe^{2+} : H_2O_2 ratio. They indicated that the solar photo-Fenton was more efficient in removing COD when compared to indoor photo-Fenton due to reduction of $\text{Fe}(\text{OH})_3$ formation. They also reported that the highest removal was recorded at 89% in the presence of 1:30 ratio of Fe^{2+} : H_2O_2 under acidic pH ~2.8 after 3 h of solar exposure. The concentration of H_2O_2 greatly influenced the COD removal, as it affected the production by hydroxyl radicals that attacked the pollutant.

In order to design and optimize experimental parameters for POME treatment, Saeed *et al.* [59] used RSM via a central composite design model. Four independent variables, namely: (i) Fe^{2+} ions concentration, (ii) H_2O_2 concentration, (iii) reaction time, and (iv) initial solution pH and two dependent responses (COD and final solution pH) were studied. The optimum conditions to achieve COD removal of more than 85% were at pH 3.5 with reaction time of 90 min and concentrations of H_2O_2 and Fe^{2+} were set at 3.99 g/l and 2.8 g/l, respectively. Their experimental study demonstrated

that the Fenton oxidation process was able to eliminate organic and inorganic compounds in POME wastewater.

Zaied *et al.* [60] developed a pre-treatment technique for POME by the Fenton oxidation process at different hydrogen peroxide concentrations in order to increase the production of bioenergy. They reported that marginal reduction in COD and TOC of 33.80% and 28.31%, respectively, was achieved at hydrogen peroxide dose of 1.00% with 1 mM Fe³⁺, contact time of 30 min and temperature of 298 K. They also increased the ratio of BOD/COD up to 0.72, which resulted in production of 46.00% and 64.83% of biogas and methane bioenergy, respectively. They noted that the reduction occurred above the optimum dosage was due to the rupture of the aromatic rings by the powerful hydroxyl radicals which turned them into smaller atomic fatty acids such as acetic acids and formic acids.

In summary, the Fenton-based process shows that pH range 3 to 4 gives the best degradation of organic pollutants in POME for all studies mentioned above. In addition, the pH for raw POME is normally in the pH range of 4 to 5, which means that the amounts of the chemicals used for adjusting pH of organic wastewater can be minimized. So, it can be concluded that the Fenton-based process is an environmentally friendly, sustainable, and effective technique that can be applied for the degradation of POME.

3. Conclusions

Liquid waste from palm oil production can significantly lead to the deterioration of the environment if proper effluent management is not implemented and regulations are not strictly enforced. Despite the fact that many studies have been carried out by researchers for the remediation of POME, there is still a need to innovate and perform further studies on POME treatment, particularly to overcome issues related to conventional ponding system treatment techniques. Various treatment techniques have been employed to produce and discharge POME that meets the permissible limits set by the government of each palm oil exporter. However, each technique possesses inherent advantages and limitations. These limitations include the scale of study for the treatment, for which most of the experimental findings summarized in this review are based on lab-scale studies. Therefore, the efficiency of these techniques for the degradation of contaminants in POME in real application, which produces 0.67 tonnes of effluent for each tonne of crude palm oil, is still unclear. Hence, more pilot scale studies are needed for future research to identify the feasibility of the techniques in real-world industrial applications. Besides, extensive studies on the possibilities for converting sludge generated in coagulation/flocculation, electrocoagulation, membrane filtration and AOP treatment into organic fertilizer are useful to reduce the amount of sludge waste dumped in landfill. In addition, plant flexibility, reliability, capital expenditure and operating expenses are crucial factors in the selection of treatment processes as well as the environmental tolerance of the material used in the process. For this reason, it is therefore imperative to conduct further research studies, particularly on the effectiveness of the process at the industrial scale, and also on the potential of POME sludge to be recycled into organic fertilizer.

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