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A review on possible approaches of anaerobic biological processes for palm oil mill effluent: Process, quality, advantages, and limitations

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Abstract. Palm oil mills play an essential role in the economic development of many countries. Though, it is the primary source of environmental pollution and degradation. Water scarcity and the high cost of new water supply technologies are the two major factors responsible for the increasing recognition of the importance to conserve water resources by wastewater treatment and reuse. Sustainability of sanitation systems should be related to low requirements in cost, energy consumption, and maintenance. Anaerobic biotreatment is the preferred industrial choice for mediating high strength wastewater. Anaerobic biotreatments for wastewater are increasingly being researched as cost-effective alternatives to deliver low sludge accumulation, efficient biodegradation and mineralisation, microbes' reduction, and solids-free effluents. In the last decade, many studies investigated various types of anaerobic reactors in combination with membranes. This review shows the potential of anaerobic bio mediations for palm oil mill effluent. Also, the paper discusses the impact of various factors on both biological and filtration performances and identifying strengths and limitations.

1. Introduction

Human activities directly pollute water courses and the environment by toxic compounds, nutrients, microorganisms, and pathogenic substances. Luckily, global industries, such as heavy industry, livestock farming, mining, and plantation are turning into the economic mode. Despite that, the pollution amount is still increasing per time due to unsuccessful willing. Pollutants involve liquid and solid wastes that are harmful, and danger on the healthy life, and the lifestyle. Hence, a significant call for the need of marvellous treatment to decrease pollution amount, but we have not accomplished that yet because of numerous difficulties. For that, pollution awareness is an excellent alternative caution against water contamination [1–6].

Wastewater requires robust developed technology to handle the massive contamination loading with no significant issues. Thus, operating, and the capital cost will increase. Malaysian palm oil industry provides thousands employment with USD 20 billion revenue [6]. Palm oil mill effluent (POME) is a discharged wastewater from palm oil industry. POME is observed as a thick brownish liquid containing a high concentration of biochemical oxygen demand (BOD), chemical oxygen demand (COD), oil &



grease, and solids, resulting in severe worldwide issues like global warming. The generated POME amount in 2011 was around 60 million ton, while it was around 30 million ton in year 2004 and 44 million ton in year 2008 [7–9]. Usually, aerobic and anaerobic ponds are utilized for treating POME within 100 days. Although, the effluent has not met the enforced rules and regulations by the environment department of Malaysia DOE for industrial discharges [10–13].

Wastewater could be processed under physical (mechanical forces), chemical (reactions), or/and biological (microorganisms) [14]. Physical and chemical treatments have not stood well for processing high strength wastewater i.e. high concentration of ammonia, suspended solids, heavy metals, high COD and BOD ranges, and shock loadings. Therefore, tertiary treatment is needed for discharging high-quality effluent. Numerous researches have been conducted on the potential of biological wastewater treatment, such as bacteria, yeast, and microalgae [15–17]. Membrane, anaerobic, and aerobic are the current treatments for POME [9]. Table 1 shows the advantages and disadvantages of using aerobic, anaerobic, or membrane processing for wastewater.

Table 1. Benefits and drawbacks of using membrane, aerobic, and anaerobic systems

Type	Benefits	Drawback	References
Membrane	1) Produce consistent and good water quality after treatment, 2) smaller space required, 3) can disinfect wastewater effluent	1) Short lifetime, 2) fouling, 3) expensive	[18]
Aerobic	1) Short retention time, 2) efficient in handling toxic wastes	1) Unsuitable for land application, 2) low pathogen inactivation, 3) high energy consumption	-
Anaerobic	1) Low energy consumption, 2) no aeration, 3) biogas production, 4) low sludge generation, 5) high COD removal	1) Large area required, 2) slow startup, 3) long retention time	[19–22]
Evaporation	1) The remaining solids can be applied as fertiliser	1) High energy consumption	-

It has been proposed to employ anaerobic treatment for POME due high COD removal, low sludge production (5-20%), capital cost, biogas recovery, and energy requirement. Additionally, methane gas is generated during POME anaerobic treatment, which is a valuable revenue if its captured. Although, anaerobic techniques can barely handle the strength of slaughterhouse wastewater [21]. Hence, anaerobically treated water requires extra processing to achieve complete removing of pathogenic organisms, total phosphorus (TP), and total nitrogen (TN) [23–25]. Finally, it was found that using a combination system (e.g. UV/H₂O₂, hybrid anaerobic baffled bioreactor) can discharge a satisfying effluent quality [21,26].

2. Anaerobic Biological Treatment

Microorganisms break down different kinds of contaminates in POME under anaerobic conditions, this process is named as anaerobic biological treatment. Organic matters and nutrients are considered as significant food for microbes in POME to increase their population and size. Anaerobic biological treatment is an essential process for POME to achieve high removal amount of pollutants with low costs. It is considered friendly and economical because it produces biomass which could be used as fertiliser, and biogas source. Although, not all anaerobic treatments capture and store the discharged biogas. The below section reviews possible POME anaerobic mediations showing their mechanisms and pointing to their drawbacks that require more investigations.

2.1 Anaerobic Contact Digestion (ACD)

ACD is considered kind of anaerobic digester. ACD comprises of sedimentation tank and digester where digester sludge is discharged to the sedimentation tank to settle, while the effluent is pumped to the digester. It has been occupied for POME, fermented olive mill wastewater, alcohol distillery wastewater, and ice-cream wastewater [27–29]. If the digester and the sedimentation tank are not combined, it is named as conventional method, but if they are combined, it is termed as non-conventional system. ACD

treatment is considered an appropriate method for highly contaminated wastewater remediation. ACD is stable, and quick start treatment compared to anaerobic filter method because it delivers sufficient mixing [29,30]. During using pilot plant ACD for POME treatment, a scum layer was produced [27]. Although, it delivered about 80% COD reduction. Despite that, it was found requiring long solid retention time (SRT) and can process only half of the influent.

2.2 Microbial Fuel Cell (MFC)

Microorganisms oxidize organic solids within oxygen existence in the anodic chamber which produce electrons and protons that travel to the cathode side [31]. In another definition, MFC is a biochemical device that utilizes microorganisms as biological tool to transform organic solids' chemical energy (e.g. glucose) into electricity [32,33]. Usually, anode and cathode sections are separated by membrane layer (e.g. proton exchange membrane) or salt bridge which permits proton (H^+) transfer from the anode to the cathode side to form water by combining with oxygen [34]. Studies had reported on electricity generation, microbial communities, electrodes materials, degradation process, and membrane material in MFC treatment [31,35]. Its worth mentioning that cathode, anode, and membrane materials are still expensive. MFC has been required to have good scalability, low cost membrane, high quality electrodes, and high electricity generation to suit human demand of energy. Despite that, most of the anode materials are built up from high resistivity carbon which largely contributes in wasting energy [36–38]. Luckily, using Si, Ni, TiO_2 , and Cu as edged metals for producing low resistivity composite based carbon as MFC anode electrode may overcome the negative factors. Currently, carbon felt, carbon paper, carbon cloth, graphite fiber brush, and graphite rod are the common electrodes for MFC, but carbon felt is the most preferred.

Many sorts of microorganisms were investigated inside MFC treatment such as *Shewanella Putrefaciens*, *Pseudomonas Aeruginosa*, and *Geobacter Metallireducens*. Electrogens such as *Shewanella Putrefaciens*, and *Geobacter Metallireducens* which are pure cultures able to transport electrons for current generation might be less or more than mixed culture. Hence, organic converting process into electricity is made in MFC by electrogens (pure cultures) and mixed bacteria cultures [32,39–42]. Electrical conductive pili, electron mediators, and direct outer membrane c-type cytochrome transfer are the common ways of electron transfer in MFC by extracellular electron transfer mechanism [43,44].

Attachment process of microbes with MFC electrode is important to achieve stable electricity generation. Communications are performed among microorganisms during immobilization stage at the biofilm or extracellular polymeric substance (EPS). High energy conversion, efficient operation during low and ambient temperatures, and no aeration is required are the major advantages of using MFC [45,46]. It has been utilized for textile wastewater containing azo dyes, mustard tuber wastewater, chocolate industry wastewater, starch processing wastewater, food processing wastewater, domestic wastewater, and paper recycling wastewater [47–54]. POME is an appropriate substrate for MFC due high COD, and BOD concentrations. A study achieved 45 Mw/m^2 power generation and 45% COD reduction during 15 days of employing POME as influent for double chamber-MFC [55]. Another research achieved 3004 Mw/m^2 power generation from using synthetic wastewater containing acetate in double chamber-MFC, while it produced only 622 Mw/m^2 from utilizing POME [56].

MFC has been integrated with other technologies to avoid system weaknesses like Up flow membrane less microbial fuel cell (UML-MFC) combination. Successfully, UML-MFC delivered higher microorganisms removal than using anaerobic digestion system [57]. In addition, ammoniacal nitrogen, and COD removal were 93.6, and 96.5%, respectively. The main drawbacks of scaling up MFC are limited power output, membrane fouling, high operation cost, and high internal resistance which make MFC being long-term Laboratory scale [58].

2.3 Anaerobic Ponds (AnPs)

AnPs are neither mixed nor aerated ponds, they are structured to process high organic loading rates with lack of dissolved oxygen (DO). It is a traditional way employed for wastewater treatment sludge digestion, especially in the tropical countries [59]. AnPs are not installed in low or moderate temperature countries because it requires a long time and ends up in delivering unsuccessful digestion and low gas generation. Strong wastewater containing high pollutants amount is fed to AnPs for solids settlement

and BOD reduction. Anaerobically, solids are digested and settled at the pond bottom, while the partially clarified supernatant liquor is pumped to another treatment stage. Methanogenic (methane production) and acid-forming bacteria (acid formation) are related phases working groups in AnP to deliver biological degradation. Biodegradation process leads to CH₄ and CO₂ emission by anaerobic microbes. Complex organic solids are being transformed at the acid phase by acid forming bacteria to short-chain volatile organic acids. Then, the bacteria utilise short chain organic acids to produce CO₂, hydrogen gas, and acetate. Finally, methanogenic bacteria transform the final components to methane, as shown in equation 1, and 2.



Population and activity balance is important to accomplish high performance of wastewater pollutants degradation as dynamic equilibrium. Also, pH over 6, and temperature higher than 15 °C are the main circumstances impacting the biological activities [23]. Therefore, the optimal pH and temperature ranges were 6.6-7.6, and 25-40 °C, respectively [60]. Furthermore, it was reported that sludge amount is minimal due to high-performance bioactivity. Organic molecules reduction, sludge digestion, and solids settlement are achievable through using longer hydraulic retention time (HRT). Usually, AnPs operation should be within 5-50 days retention time, 2-4 meters depth, and 2 m/d hydraulic surface loading [24,61]. It is expected to deliver 80-90% reduction of BOD₅, require none or low energy, generate energy, be high flexible, accumulate low sludge, handle high organic loading rate, and be long lifetime operation [60]. Despite that, AnPs produce hydrogen sulphide, odorous compounds, and require maintenance.

2.4 Up Flow Anaerobic Sludge Blanket Reactor (UASB)

Anaerobic treatments became primary treatment among other methods for domestic, mixed, high strength i.e. contains great amount of organic compounds, oil and grease, heavy metals, ammonia, and fats (e.g. industrial, agro-industrial, sugar beet, coffee, pharmaceutical, POME, ice cream, slaughterhouse, and potato wastewater) [62–70]. Frequently, UASBs have been installed to vastly reduce suspended solids (SS), and organic loading ranges. In year 1970, UASB reactors were developed to become high rate anaerobic digesters for tropical and subtropical countries such as India, Colombia, and Brazil [71]. It is worth mentioning that UASB reactors are still not recommended for non-tropical regions due to limited digestion, slow biodegradation, sludge deterioration, and sludge flotation unless there are suspended solids pretreatment and two stages of anaerobic treatment [72-73].

UASB treats wastewater within 6-12 h of retention time by distributing wastewater influent across the reactor base, then flowing through the sludge layer to the upper level to assure high contact performance between the anaerobic bacteria and the wastewater pollutants (see Figure 1). UASB reactor is separated into upper (settling) and lower (digestion) zone. Biogas bubbles are produced due to the biodegradation process, where UASB's deflectors prevent delaying in sedimentation process by stopping gas bubbles entering the settling layer. Also, phase separators in UASB reactor are employed for gas collecting. About 0.2 kg sludge is generated for every 1.0 kg reduced of BOD which is higher than anaerobic ponds, and less than conventional activated sludge treatment. In some cases, UASB reactor having low HRT (2-5 h) can be used to prevent methanogenesis reaction, its named as hydrolytic up-flow sludge blankets (HUSB). Despite that, hydrolysis process requires over 15 days to deliver high performance in UASB reactors.

At 25 °C, AnP can remove 70% BOD in one-day retention time, while UASB reactor produces 70% BOD removal in 6 h. Despite that, UASB reactors are considered smaller and higher cost than AnPs, but AnPs generate odours. Additionally, UASB reactors suffer from sludge granulation, long startup time, methane production, reactor clog and biomass aggregation [65,68,74,75]. Although, it has been recommended to implement toxic and inhibition detection, organic loading rate determination, and survey sludge development to prevent sludge accumulation.

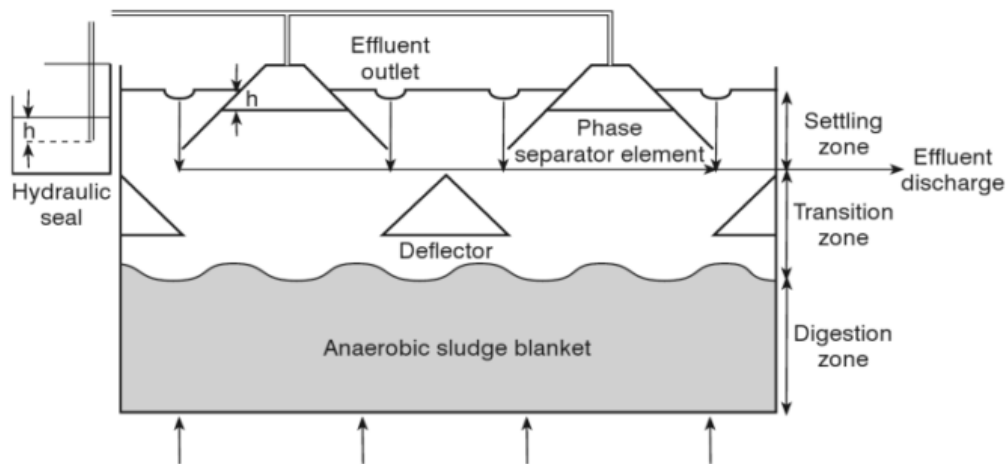


Figure 1. Schematic of UASB reactor [4]

A study proved that UASB reactors deliver more performance in removing suspended solids, COD, and organic solids than settling tanks. Also, two stages UASB system delivered 71% COD removal at 15 °C. About 98.4% COD removal was achieved by treating POME through UASB reactor with 10.63 kg COD/m³ day organic loading rate (OLR) [64]. Although, treating wastewater containing high volatile fatty acid overloads and destabilizes the UASB reactor after 15 days of processing. Hence, UASB structure is endorsed to be fitted with high organic loading rates, and capable of processing POME effluent. For POME processing, a researcher investigated granule formation from high OLR by two-staged UASB reactor [76]. Hence, the study showed an importance to employ a pair of UASB reactors for segregating acidogenesis, and methanogenesis stages. Also, it was reported that high methane conversion with 90% COD removal is possible for 30 kg COD/m³ day of OLR. Also, another study proved that reactors seeding process with granulated sludge deliver short startup time with high reduction performance, and require less time to process gradual rise in influent OLR [74,77].

2.5 Up Flow Anaerobic Sludge Fixed Bed Reactor (UASFF)

UASFF is a combined system between the anaerobic filter and UASB reactor. It is designed to achieve high performance, eliminate drawbacks, and combine benefits. Many advantages are reported by using UASFF such as clog elimination, prevent biomass washout, handling high OLR, stability, and excellent biomass retention [30]. A study proved that using UASB or anaerobic filter provide lower performance than UASFF for wood fibre wastewater [78]. Also, UASFF was examined for POME, brewery, coffee, virgin olive oil, slaughterhouse, dairy, and sugar wastewater [79–86]. It was reported that methane emission from UASFF is acceptable, and the removal efficiency is at least 70% of COD concentration except wood fibre wastewater because it is considered difficult to be composed. The stability of UASFF for POME processing is decided by the internal packing and effluent recycle ratio [79].

2.6 Anaerobic Baffled Bioreactor (ABR)

The development of ABR has been initiated in the 1980s. It is applied widely for wastewater due to having an anaerobic filter, low bacterial washout, up-flow anaerobic sludge blanket, no fixed media required, low up flow liquid velocity, high contact time, high stability towards toxic, no requirement for particular sludge or gas separator, and low energy required [87,88]. Also, ABR is presented as series of Up flow anaerobic sludge blanket (UASB) reactors [89]. ABRs behave partially as trickling submerged fixed, activated sludge reactor, UASB reactor, septic tanks, as well as fluidised bed reactor. ABRs comprise from series of baffles and compartments where the influent flows under and over.

The significant attention towards ABRs is to produce capable bioreactors of high capacity of solids retention. Therefore, vertical baffled to a plug flow was illustrated for treating high solids slurry [90]. The design of ABR provides sufficient time for influents to be decomposed and discharge acceptable effluents. Well-structured and designed ABR brings a considerable contact between microorganisms and substrates in short time and deliver high removal rate without occupying massive volumes. A series

of vertical baffles force the influent to move over and under which helps microorganisms to thrive (see Figure 2).

Vertical ABRs presented good ability in retaining biomass and methane gas generation. Upflow chamber is the Methanogenesis zone, while the downflow chamber is acidification zone. Compared to continuous stirred tank reactors (CSTRs), less ABR's volume can achieve higher reaction rate per unit reactor volume by strongly retaining the biomass separately in the reactor from the inlet wastewater [91]. The physical design of ABR makes it capable of treating contaminated water in one unit with using circulation pattern and requires low capital costs. Figure 2 shows ABR structure and procedure for wastewater treatment. Unlike UASB, ABR does not require granulation technique to increase the bioreactor efficacy.

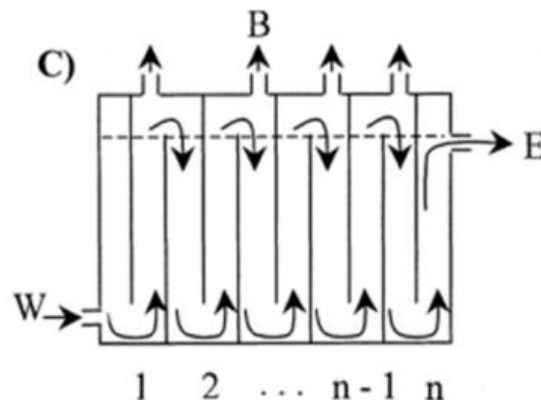


Figure 2. Vertical Baffled Reactor (W= wastewater, B= biogas, E= effluent) [92]

In year 1987, hybrid ABRs (HABRs) were introduced. HABRs involve suspended and attached growth initiated by boosting flocculent and granular biomass growth by using 0.46 m/h of liquid up-flow velocities, 0.97 kg COD/kg VSS d of low initial loading rate, and 4.01 g VSS/l [93]. A stable formation of 0.5 mm granules presented in all HABRs after the one-month duration. Despite that, the flocs were found weak and less than 1.5 mm, but it reached 3.5 mm after three months and it is determined by the substrate kind. Acetoclastic methanogens (*Methanosarcina* cluster) were the granules content and it raises to the reactor surface because it is full of gas cavities and being low-density.

ABR got a further modification from Boopathy and Sievers [94] for treating swine wastewater containing a high range of small particulate materials. Another study occurred on ABR modification by using two compartments, where the first section size was 10 L, and the second section was half the first compartment size. It was recorded that the three-chamber ABR collected half the solid amount (10.45 g/L) of the two compartments ABR. Although, three-chamber ABR showed higher treatment efficiency and solid washout than two compartment ABR [94]. COD and BOD removal were 90% at the up-flow compartments [95–97]. Laboratory scale ABR was examined for slaughterhouse wastewater (SWW) characterised by 63.38 mg/l of TN, and 183.35 mg/l of total organic carbon (TOC) [98]. The highest reduction percentage was 51.52% of TN and 88.88% of TOC. It was found that operating costs increase with TOC range, according to Bustillo-Lecompte et al. [26]. Another research reported about ABR and UV/H₂O₂ combination for treating SWW containing 973 mg/l of TOC in laboratory scale [99]. It is concluded that using hybrid system delivers higher removal efficiency than employing an individual method. After three days of treatment, the combined system delivered 95% TOC reduction. Table 2 presents ABR treatment for various sorts of wastewater.

Table 2. Anaerobic baffled bioreactor treatment for different sorts of wastewater [92]

Wastewater	HRT (h)	Influent COD (mg/L)	COD Removal (%)	OLR (kg/m ³ .d)	Gas produced (L/day)
Dilute wastewater	80	50	80	-	-
Brewery wastewater	15	-	92	5.6	24.01
Soybean wastewater	39.5	2000	97	1.2	-
Heavy oil produced water	60	50	65	-	-
Domestic wastewater	48	305.18	74	-	-
Municipal wastewater	6	350	86	2.62	0.34
Palm oil mill wastewater	3	16000	77.31	1.60	27.4
Undiluted brewery wastewater	15	5500	31.82	2.75	55.7
Synthetic substrate wastewater	24	3000	82	3.00	-
Pulp and paper mill black liquor	48	4020	68	5.00	2.95
Penicillin reduction wastewater	64	8.00	65	2.64	-
Domestic wastewater	22	716	72	-	-
High sulphate wastewater	240	6.6	82.71	0.66	0.29
Low strength wastewater	12	550	89	1.69	-
Soybean protein processing wastewater	39.5	10000	97	6	-
Complexed wastewater	8	500	88	2	0.31
Synthetic wastewater	10	501	90.7	1.2	0.36

2.7 Anaerobic Fluidised Bed Reactor (AFBR)

AFBR is a reactor that can be occupied to handle different sorts of multiphase chemical reactions. The operation of AFBR has minimal issues in gas hold up, plugging, and channelling matters [8,100,101]. Moreover, it possesses large surface area and requires low HRT to handle high OLR, and high polluted wastewater [100–103]. Anaerobic expanded bed development got initiated to convert diluted organic wastes to methane at high hydraulic and organic loading rates, and low temperatures. During year 1988, anaerobic expanded bed followed by post-treatment was scaled up into pilot scale 10,000 gal/day and studied. It was reported that the assessment occurred for two years on two AFBR, where sand was the carrier in the first reactor, and the second reactor used granular activated carbon (GAC) [104]. It was shown that using sand as carrier delivers less attached biomass with slow biofilm growth and higher BOD removal.

Contact time between the inlet and the bed is deciding by influent up-flow velocity. Therefore, attachment and growth of biomass will occur on the reactor support media [30]. AFBR treatment efficiency can be manipulated by bed material sort that can impact the whole process [102,103]. The performance of AFBR has been examined to treat POME, pharmaceutical effluent, slaughterhouse wastewater, ice-cream wastewater, brewery wastewater, wine and distillery wastewater, real textile wastewater, and cutting-oil wastewater [63,100,102,103,105–110]. In comparison to up flow configuration, a study showed that inverse flow AFBR is capable of high OLRs, with high stability even at overload situations [107]. Even AFBRs are capable of high OLRs, it is preferred to occupy less AFBR size for lower OLRs [30].

A study combined ovoid saponite with AFBR. It resulted 94.4% COD removal, while using granular activated carbon produced 60% COD reduction [8]. AFBR has been determined as a suitable method for POME mediation process than anaerobic filters because of the good strength to treat high OLRs. Also, shorter HRT 6 h is required for AFBR to mediate POME than anaerobic filters HRT which is 1.5 to 4.5 days [30]. AFBR treatment for different wastewater kinds with COD removal and OLR ranges are tabulated in below. From the organised data in the table, it declared that 65-95% is the COD removal range by AFBR.

2.8 *Continuous Stirred Tank Reactor (CSTR)*

CSTR is a closed tank acts as a digester with a mixer. It has been occupied to treat coke wastewater, jam wastewater, and dilute dairy wastewater, but in some cases, it was operated under aerobic circumstances [111–113]. CSTR operation is a continuous flow of outlets, and inlets. Gas generation through the biological activity increases due to agitation process which delivers high contact in short time.

CSTR capability was investigated for removing pollutants from POME [114]. It produced effluent with range 93.6 to 97.7% of COD reduction. In Masai, Malaysia, Keck Seng Berhad utilised CSTR as a mill for POME mediation [115]. It delivered 83% COD removal while occupying CSTR for dairy wastewater resulted 60% COD reduction. Keck Seng Berhad got different results from [114].

Operating conditions like temperature have high impact on wastewater treatment performance. Another factor known as insufficient mixing properties led Keck Seng Berhad to receive low COD removal. Another research worked on combining CSTR with biofilm support system (BSS) [116]. BSS was implemented by using low-density nylon mesh as supporting material for biomass growth. The hybrid system proved high ability towards discharging effluent with lower COD ranges.

2.9 *Constructed Wetlands*

Wetlands are usually land areas which are kept wet during part or all of the times. They are shallow earthen tanks involve a soil layer for plants growth which performs attached growth biological wastewater mediation. Often, wetlands change among deeply flooded or uplands and continuously systems [117]. It can treat industrial wastewater, mine wastewater, animal wastewater, domestic wastewater, and municipal wastewater.

Historically, natural wetlands have received wastewater discharge as a convenient method for waste treatment in southeastern United States coastal plain areas, and poorly drained fens and marshes of the north. For instance, Florida cypress domes, or Michigan Houghton Lake fen were extensively examined and recognised as a treatment method for wastewater [118]. During the 1950s, the first practices of wetland vegetation were performed to reduce wastewater contamination, while the first full-scale free water surface wetland was structured in between 1967 to 1969 in the Netherlands for camping site wastewater treatment. An advantageous development occurred on wetlands performance by occupying coarse materials (washed gravel) instead of wetlands soil in the late 1980s [119]. Also, during the 1980s, a thorough understanding had been made for improving numerous wetland benefits and weaknesses. Hence, it got strong global attention as a robust technology for wastewater mediation [118]. Then, vertical flow constructed wetlands have been employed in the 1990s as a significant process for ammonia and nitrogen reduction since it provides massive oxygenation for the nitrification process. After few years, it showed the importance of using a hybrid system involving constructed wetlands to accomplish nitrification and denitrification process [119].

In the northern and the central part of Europe, constructed wetlands were found popularly employed for thirty years in wastewater treatment plants for small populated cities (see Figure 3). Developing countries consider constructed wetlands more attractive method in southern and northern countries than conventional wastewater techniques. In addition, constructed wetlands are considered flexible, low operation duty, less susceptible to OLR variation, low sludge formation, low maintenance and construction cost, and require low experienced workers than conventional method [20,21,120,121]. Despite that, employing massive land area, and low performance at low temperatures are considered significant drawbacks, but it is still a concrete way for tropical regions [121,122].

Constructed wetlands are considered unique and ecofriendly among other technologies [117]. It was reported that full scale constructed subsurface flow wetland able to achieve 30% organic matter reduction [95,123]. Also, the enforced regulations and laws are not satisfied with the discharged effluent quality by constructed wetlands where TSS, COD, and BOD₅ reduction percentage are about 85, 89, and 91%, respectively. Constructed wetland efficacy for mediating slaughterhouse wastewater (SWW) was investigated by Soroko[124] via utilising three basins, where the first one was horizontal flow (HFCW), and the other two were vertical flow (VFCW). They produced 78.20, 99.90, and 97.40% of TN, BOD, and COD removal with using gravel and sand beds, where SWW influent contained 500, 2500, and 3188 mg/l of TN, BOD, and COD. *Typha latifolia* was combined with constructed wetland which had 89% active volume and 111 days HRT for SWW treatment [125]. The hybrid system

accomplished 87, 88, 72, and 95% removal performances of TN, TP, TSS, and BOD, respectively. Differently constructed wetlands were examined by [126] for SWW treatment. SWW influent was characterised by 56-64, 79-87, and 293-314 mg/l of TN, BOD, and COD, respectively. It delivered 5.20-25.40, 9.27-71.40, and 28.28-75.03% removal ranges of TN, BOD, and COD, respectively [126].

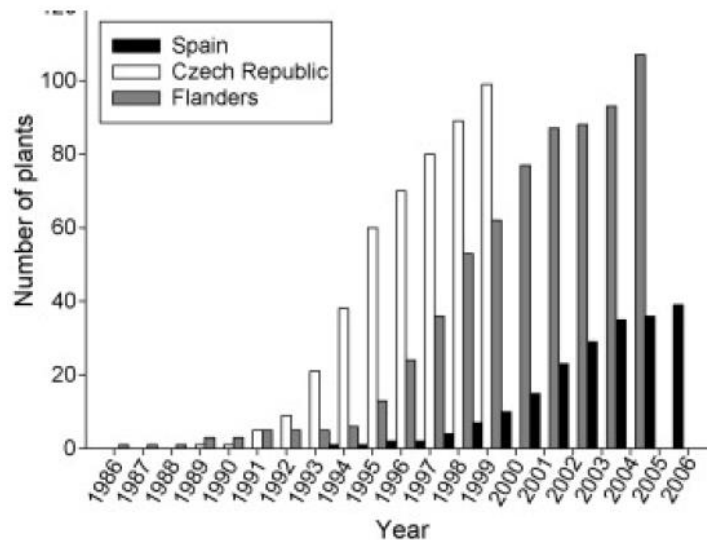


Figure 3. Cumulative number of WW treatment plants based in CWs over the last years in various European regions [127]

2.10 Anaerobic Filters

Anaerobic digestion has been utilised for wastewater treatment since over 100 years ago. The development started from airtight vessel and septic tank to full mixed digester equipped with temperature control and ended as high rate biological reactor containing active biomass. The advancement of anaerobic wastewater digestion led to variety of changes such as growth technique, fluidised bed (attachment on mobilised carriers), and anaerobic filter (attachment on static carriers) [104]. To date, all these advancements are still in developing stage.

The anaerobic filters were employed to treat various types of wastewater such as ice-cream manufacture wastewater, beet sugar water, distillery wastewater, drug wastewater, slaughterhouse wastewater, brewery wastewater, municipal wastewater, landfill leachate, wine vinases, and soybean processing wastewater [104]. It is preferred in some cases because it delivers high pollutants reduction, handles high loadings, produces high contact rate between liquid medium and biomass without impacting the process performance, needs inexpensive construction, requires short hydraulic retention time, and demands small reactor volume [8]. Although, clogging is a significant drawback for continuous run anaerobic filters [128–130]. During processing POME with 20 g COD/l/day of OLR, and processing SWW with 6 g COD/l/day of organic loading rate OLR, the anaerobic filter got clogging. Anaerobic filters utilise the packing surface for biomass attachment, development, and growth. Influent enters the process from the bioreactor bottom, while the generated biogas and the effluent discharge from the top.

POME was processed by the anaerobic filter, according to Borja and Banks [110,131]. Anaerobic filters were found able to discharge effluents with at least 70% COD removal [30]. Empirical examinations on improving anaerobic filters efficacy got occurred. For instance, it was reported that optimal recycle ratio differs based on OLR range which will produce enhanced COD removal and biomass high retention time. Thus, increasing the optimal recycle ratio delivers higher methane amount [132]. Open-pored support media which involve high porosity were suggested by Bala et al. [8] for biomass retention optimisation by trapping it. Moreover, continuous fed anaerobic filters were found able to accomplish great biological decomposition, and stability [133]. The highest record of POME

COD removal was 94% with 63% of methane at 4.5 kg COD/m³/day of OLR, while 90% was overall COD removal with 60% of methane gas composition [131].

2.11 Membrane Bioreactor (MBR)

Between the late 1960's and early 1970's, membrane bioreactor (MBR) was introduced for processing wastewater. Then, it faced massive evolution and extensive usage since the early 1990s. In Mansfield, Ohio, United States, the first large MBR was structured. Also, in the 1990s, the first submerged MBR was built, and it was found requiring anaerobic or aerobic circumstances, and lower operational cost to deliver higher active biomass settling, higher mixed liquor, higher suspended solids concentration and efficient longer processing in comparison to other MBR types [134]. Later, the first large-scale internal MBR system was built during 1998 in North America for industrial food wastewater treatment [135]. However, high capital cost and difficult maintenance reduced the overall research on MBR.

MBR process comprises from biological treatment (aerobic or anaerobic suspended growth) and membrane separation (like ultrafiltration, and microfiltration) [136]. The membrane layer is a thin film porous structure employed as a filter which rejects anything larger than the pores open. The pressure difference between membrane layer (transmembrane pressure) generates potential energy allowing soluble components and treated wastewater to pass, while particulate matters (organic compounds, graphite, glass, metals, ceramics) are retained based on membrane pore size, and membrane structure. Activated sludge process is part of MBR treatment where microporous semipermeable pressure-driven rejection membranes are retaining biomass on their surfaces.

There are two types of MBR, the first type includes immersed MBR in Bioreactor, and they co-occur, while the second type recirculates part of the effluent for the filtration system after the biological treatment, while the rest is pumped back to the bioreactor for extra treatment. Thus, it is a high-cost operation due to the enormous need of efficient pumping system [137].

During membrane process, multiple mechanisms are happening in removing antibiotics. At the initial filtration stages, adsorption can occur by membranes to the hydrophobic antibiotics (who are possessing strong hydrogen bonding). In the other side, stable rejection can deliver removal process due to uncharged solutes steric effects or charged solutes combined electrostatic and steric effects. These mechanisms are varying based on the membrane features (pore size, surface morphology, material), the solution (ionic strength, pH), and the compound (hydrophilicity/hydrophobicity, pKa, molecular weight cut-off (MWCO)) [138]. During wastewater treatment, reverse osmosis (Pore size > 0.001 mm), and nanofiltration (Pore size less or equal 0.0001 mm) are substantial membranes in removing low molecular weight (pharmaceutical, antibiotics), while ultrafiltration and microfiltration are large enough to be micropollutants passages (see table 3). Additionally, many examinations presented antibiotics removal involving trimethoprim, tetracyclines, sulfonamides, and quinolones. Hence, there is a demand to combine reverse osmosis (RO) with nanofiltration (NF) membranes to achieve overall high efficacy [139,140]. Fouling (By inorganic matter, SMP, EPS) can lead to rejection improvement of micropollutants due to the transforming to negative surface charge which contributes the ionic species electrostatic rejection [141,142]. Hence, non-ionic solutes adsorptive capacity has increased eventually.

MBR biological treatment is similar to the conventional wastewater treatments. For instance, cell growth, metabolism, nitrification, denitrification, and phosphorus removal are occurring inside MBR system. However, MBR system permits accumulation of better nitrification rates, very high sludge volume index, lower sludge production, high amounts of predators like metazoan, and protozoa, high biomass concentration, and slow-growing microorganisms [134,143]. It was proposed that metazoan, and protozoa grazing on the nitrifying bacteria can decrease MBR nitrifying capacities.

MBR nitrification and denitrification rates are affected by sludge age. Previously, it was shown that the removal efficiency of total nitrogen increased from 49 to 73% by increasing sludge age from 20 to 60 days during treating black water [144]. Also, the treatment occurred under low COD/nitrogen ratio, high nitrogen amount, and low dissolved oxygen (0.1-0.2 mg/l). The process delivered 100% reduction during denitrification process, and 40% reduction within nitrification process. In contrast, another researcher found the different outcome in decreasing sludge age from 29 to 16 days which resulted higher total nitrogen removal from 89 to 91%. The study had occurred by three stages anaerobic anoxic and aerobic system of the pilot plant [145].

Pre-denitrification is universal wastewater system for biological nitrogen removal. It includes anaerobic and anoxic tank followed by aeration tank for nitrification process. The aeration process generates nitrate which is pumped back to the anoxic tank for denitrification. Then, the provided electron donors are employed with the nitrate to achieve denitrification. Sometimes, different recirculation rates are used to improve MBR performance.

Traditionally, MBRs are used in conventional activated sludge process (CASP) as a replacement for the final sedimentation process or the secondary clarifier [146]. For that, micro or ultrafiltration MBR maintain higher concentration of mixed liquor suspended solids (MLSS) compared to conventional activated sludge process (ASPs). It resulted 99% solids removal and almost complete clarification. Additionally, semipermeable MBR is capable of being a good barrier against bacterial cells and colloids. It was reported 2-5 log removals of human enteric viruses, and 2-7 log removals of coliform bacteria for different range membranes and MBR [147,148]. In conclusion, operating conditions (gel layer formation), pore size, membrane type, and membrane material are essential factors in MBR performance against colloidal, viruses, and bacteria retention.

The produced water can be used as an inlet for heat integration, landscape purposes, and industrial sanitary. Hence, A researcher had examined ultrafiltration MBR efficacy against organics and nutrients removal from SWW [149]. SWW was characterised by having 102, 16, and 571 mg/l of TN, TP, and COD, respectively. The process resulted 97, 96, 65, and 44% removal of COD, TOC, TP, and TN, respectively. Also, UF MBR produced a successful organics reduction, but high nitrate amount passed with the effluent. For that, the produced effluent requires a complete denitrification.

Table 3. Comparison of different membrane dimensions and pore size exclusion used in SWW treatment [95].

Membrane type	Pore size (mm)	TOC removal (%)	COD removal (%)	BOD removal (%)	TN removal (%)
Microfiltration (MF)	0.080-0.550	44.81	90.63	-	45.22
Ultrafiltration (UF)	0.030	75.00-96.00	83.00-97.00	-	27e44
Ultrafiltration (UF)	0.010-0.100	-	94.52-94.74	97.80-97.89	-
Reverse Osmosis (RO)	0.001-0.005	-	85.80	50.00	90.00

2.12 Membrane Bioelectrochemical Reactor (MBERs)

MBERs are combined system involving Microbial fuel cell (MFC) and Membrane filtration (MF or UF). The membrane system can be installed within either the cathodic or anodic compartment. MBERs Hybrid treatment could be a remarkable option for wastewater treatment evolution and competitive among conventional aerobic MBRs (AeMBRs) or anaerobic MBRs with successful nutrients removal, low dissolved methane, no aeration, minimal maintenance requirement, and low energy requirement [150]. Additionally, scaling up (over 1 m³), construction cost (like ion exchange membrane, and catalyst), discharging high suspended solid concentration, no effective treatment against nutrients (like nitrogen, and phosphorus), and extended treatment period (11 h are required to achieve 65-70% degradation) are considered weaknesses for Bioelectrochemical systems (BES). In processing wastewater, MFCs approach is an advantageous method. Also, MFC-MBR integration is an approach for sustainable wastewater treatment. Cathodic and anionic compartments are semi-separated by involving anion exchange membrane (AEM). Its named as microbial desalination cell (MDC), if used as desalination solution.

The bioelectrochemical concept can be defined as the transformation of organic chemical energy to hydrogen gas or bioelectrical energy by microbial electrolysis cells (MECs) or MFCs. MFC does not require aeration system, for that sludge production, is less than CAS process [45]. Biodegradation is occurring for organic matters in the anodic MFC section. Then, electrons are generated and transferred to a solid electrode through NADH/NAD⁺, then to the serial cytochrome agents at the outer cell

membrane. Finally, ferricyanide, nitrate, or oxygen in the cathodic compartment are accepting the transferred electrons through an external circuit.

If the theoretical open circuit voltage (OCV) is about 1.1 V, the carbon source is sodium acetate in the anodic compartment, and oxygen is employed as an electron acceptor in the cathodic chamber, then 0.8 V was observed as the low potential due ohmic loss, electrolyte diffusion resistance, and overpotential.

It was found a significant reduction in fouling matter via using granular activated carbon (fluidized bed support material), and membrane in the cathodic section. Also, AEM film can be employed for nutrients removal and make MBER versatile process.

MFC ability to direct transfer of chemical energy into bioelectrical energy with flexibility and low carbon footprint has shortened long way of using treatments for biodiesel production (see figure 4). Also, applying an external voltage inside MFC leads to gaseous hydrogen evolution or hydrogen peroxide in the cathodic compartment, known as MEC process.

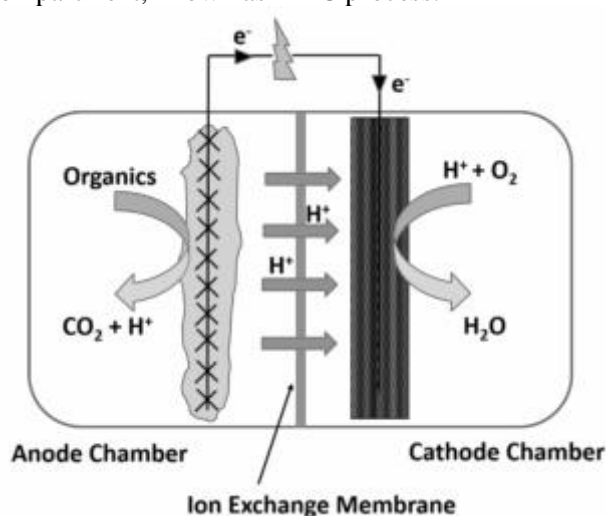


Figure 4. schematic of two chambers MFC

2.13 Fungi

Varying circumstances are not huge matter for fungal organisms because it has significant capability for augmenting their metabolism based on the environmental conditions. Their presence is vigorously reliant on metabolism climatisation feature. Also, fungal metabolic activity is supported by extra and intracellular enzymes.

Textile wastewater treatment starts with dyes decomposition to another chemically forms by fungal enzymes. Laccase, manganese peroxidase (MnP), and lignin peroxidase (LiP) are utilised enzymes by fungal organisms for biodegradation process [151]. Also, the used fungal species for azo dyes decomposition is the white rot fungal cultures. Hence, wastewater COD concentration faced a removal process with dyes decomposition by white rot fungus *Pleurotus eryngii*, *Penicillium simplicissimum*, and White rot fungi *Corioloropsis sp* [152–154]. Though, long holding time, unstable treatment, large reactor, unreliable enzyme generation, nitrogen necessity, and long growth are a significant drawback of wastewater fungal treatment [155]. Also, after 20 to 30 days of fungal wastewater treatment, bacteria dominate the medium by their fast growth while fungi predominate wastewater medium with idle dyes degradation [156]. Despite that, it was found that *azoreductase* enzyme can provoke Green macroalgae *Cladophora* species to degrade wastewater azo dyes [157-158].

2.14 Anaerobic Digestion

Its degradation process of wastewater organic compounds under anoxic or anaerobic circumstances. Also, it can be defined as the engineered methanogenic anaerobic decomposition of organic matter. In 1859, the first anaerobic digester was built in Bombay, India by leper colony. Then, sludge processing and sedimentation occurred with a dual basin in 1904 [159]. Initially, anaerobic digestion process was

used in treating sludge, industrial wastewater, and municipal by organic matter decomposition approach. Time is the matter with high or standard rate anaerobic digestion process because the responsible bacteria consortia and other microorganisms require an undetermined period for the bioaugmentation, adaptation, and organic matter biodegradation [8,104]. Anaerobic digestion requires long retention time, stable operation, and large reactor size to accomplish full digestion process (see table 4). Hence, it was proposed to develop high rate anaerobic bioreactors via biogas capturing, delivering shorter retention time, and employing less reactor volume [104]. Standard rate anaerobic digestion is processing wastewater with no requirement for mixing or heat, while high rate digester requires heating and mixing. As a result of that, high rate digestion process needs 15 days or less of hydraulic retention time (HRT), while standard process requires 30 to 60 days [160].

Hydrolysis, acidogenesis (including acetogenesis), and methanogenesis are theoretical biological digestion phases of POME, which discharge treated water, carbon dioxide, and methane [161]. First of all, proteins, lipids, carbohydrates, nucleic acids and other complex molecules (high molecular mass) are processed into substrates such as amino acid, fatty acids, simple carbohydrates, purines and pyrimidines, and sugar by Hydrolysis process. Then, lower molecular mass like organic acids, lactate, succinate, methylamine, acetate, carbon dioxide, hydrogen gas, methanol, and other fermentation products are produced through utilizing acidogenic bacteria via breaking amino acids, fatty acids, sugar, and other compounds where carbon dioxide, hydrogen, and acetic acid (from acetogenesis) are main components of the generated organic acids. Finally, acetolactic methanogens utilise carbon dioxide and acetic acid for methane production as the final product, while carbon dioxide and hydrogen are metabolised by hydrogenotrophic [95,161]. In comparison with alternative technologies, anaerobic digestion process claims to be cost-effective, significant waste stabilisation, low energy requirement, high COD removal, and low sludge generation for POME treatment, also it does not require aeration system. Additionally, methane gas is generated during the anaerobic digestion process, which can be captured, and utilised for additional worthy revenue. In the seventeenth century, biogas from organic matter biodegradation got scientific attention.

Table 4. Comparison between anaerobic and aerobic digestion [21].

Characteristic	Aerobic	Anaerobic
Organic loading rate	Moderate	High
Organic removal efficiency	High	High
Nutrient requirement	High	Low
Energy requirement	High	High
Alkalinity requirement	Low	Low to moderate
Sludge production	High	Low
Temperature sensitivity	low	High
Odor production	Less opportunity for odours	Potential odour problems
Bioenergy and nutrient	No	Yes
Startup time	2-4 weeks	2-4 months

It was reported that POME and high strength wastewater (high organic carbon concentration) could be processed under anaerobic digestion method [76,99].

2.15 Biological Activated Carbon (BAC) Filtration

BAC filtration involves biodegradation process and sorption for removing CECs (Recognized as potential hazardous) [162,163]. Microorganisms development and growth are supported by a granular activated carbon (GAC) fixed bed. Eventually, bacteria initiate their evolution on the bed surface.

Previously, BAC presented a bioremediation process by biodegradation and sorption course on secondary treated wastewater where nitrogen and dissolved organic carbon (DOC) concentrations were decreased [162]. Treatment procedure starts with sorption process by activated carbon at the beginning, then biodegradation process dominates the medium by the attached bacteria on the GAC bed. According to Neptune project, biologically activated coke was utilised as sorbent material with the fixed bed system

in BAC filtration succeeding to conventional bioactivated sludge treatment. It produced 70 to 90% removal rates for numerous pharmaceutical components including the hardly degradable compounds, such as diclofenac, and carbamazepine (unpublished data, <http://www.aqua-biocarbon.de/aktuelles.html>). After ozonation treatment, BAC was employed instead of UV treatment for removing NDMA, thus its cost-effective process [164]. Though, it is recommended to check transformation products (TPs) rate to stop their passing through activated carbon (AC) filter [165]. In comparing to many wastewater technologies, AC produces no TPs because of its dependent on sorption process, but its limited capability. Hence, AC is relevant for exchanging or regenerating during determined durations. Also, pollutants removal can be varied with the AC sort, amount, sorption mechanism, and quality. The biological performance and AC efficacy are highly dependent on the medium pH level. Hence, sulfamethoxazole, sulfamethazine, and acetaminophen compound sorption and biodegradation can be significantly varied [166]. It was reported that neutral materials like carbamazepine were processed in constant removal rate, while negatively charged compounds like diclofenac, naproxen, and ketoprofen had advanced reduction [167]. Another research showed a race on different spots of AC surface to accomplish DOC sorption [164]. High polar compounds like anionic organic compounds and X-ray contrast media are used to indicate AC loading during wastewater treatment to avoid unrequired consequences. Despite that, removing high polarity compounds, and examination are hard challenges. In comparison to liquid chromatography (LC) analysis, ion exchange chromatography (IC) technique offers more efficient separation capacity for cationic and anionic compounds. Hydrophilic interaction liquid chromatography (HILIC) is used for high polar CECs like illicit drugs, pesticides, and pharmaceuticals [168]. AC filter operation can be applied before the oxidation for removing potentially toxic compounds generated by oxidative treatment, like chlorine, or ozone [169]. Though, AC has a contribution in N-nitrosamines generation from secondary amines. Also, hydrophilic natural organic matter (NOM) fractions might pass through AC filter. However, GAC performance is highly based on the matrix efficiency and GAC particle size if NOM is presented [170].

3. Conclusion

This review study showed the weaknesses of anaerobic treatments towards treating high strength POME, like clogging, specific species domination, instability, high retention time, expensive, and employing large surface area. On the other hand, they are promising methods, since they can capture biogas, and generate low sludge amount. Majority of wastewater industries use ponding systems due to their low cost. Despite that, it produces greenhouse gases and causing soil clogging. The advanced concept of anaerobic treatments is the combined system like MBR. It is a promising way to pass numerous drawbacks and discharging reusable water. Finally, applying the sustainable approach, and zero waste energy can develop, and produce eco-friendly treatment.

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