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Abstract

This study proposes a new approach for integrated technology of ultrasonic and membrane for a palm oil mill effluent treatment. This study evaluated the performance of the new design of ultrasonic membrane anaerobic system (UMAS) when a palm oil mill effluent (POME) introduces this approach. To fit kinetic study, six steady states were investigated and the results have shown that the mixed liquor volatile suspended solids (MLVSSs) range from 10,400 to 17,350 mg/l while the mixed liquor suspended solids (MLSSs) range from 13,800 to 22,600 mg/l. Three kinetic models of Monod, Contois, and Chen and Hashimoto were used to evaluate the integrated system at organic loading rates ranging from 1 to 15 kg COD/m³/day. The percentage efficiency of COD removal was from 92.8 to 98.3%, and hydraulic retention time (HRT) was from 500.8 to 8.6 days. The influent COD concentrations of the POME ranged from 70,400 to 90,200 mg/l. The integrated technology of UMAS is a more attractive one as it avoids membrane fouling problems.

Keywords: membrane, ultrasonic, POME, methane, CO₂, UMAS

1. Introduction

The palm oil industry has grown tremendously in the recent years and accounted for the largest percentage of oil and fats production in the world in 2011. Over the last few decades, the palm oil industry has been growing rapidly. Palm oil has risen to become the most produced and consumed vegetable oil in the world, widely used in food, cosmetic and hygienic
products due to its affordable price, efficient production and high oxidative stability [1]. Palm oil is the most produced vegetable oil in the world with a global production of almost 60 million tons and a global vegetable oil market share of more than 35% by weight in 2015 as reported by Hansen et al. [2] and MPOB [3]. The industry continues to generate huge revenues for the producing countries. Accordingly, it is not surprising that the oil palm industry is expected to grow further in the coming years as shown in Figure 1.

Over the long term, global palm oil demand shows an increasing trend as an expanding global population gives rise to increased consumption of palm-oil based products world consumption of palm oil [5]. [6] Stated that palm oil industries have been significantly contributing towards the economic growth and increase standard of living among the South East Asian countries. Nowadays, the global production and demand for palm oil are increasing rapidly where the plantations are spreading across Asia, Africa and Latin America. The five leading palm oil producing countries are Indonesia, Malaysia, Thailand, Colombia and Nigeria [7] as shown in Figure 2.

The development of palm oil industry in Malaysia has turned into a phenomenal in which the area of plantation expanded from year to year. The country is experiencing a robust development in new oil palm plantations and palm oil mills. This commodity plays a significant role in the Malaysia economic growth [8]. Throughout the year, Malaysia is blessed with favorable weather conditions, which are advantageous for palm oil cultivation [9]. Thus, it is not surprising that the highest yields have been obtained from palms grown in this region, which is far from its natural habitat. Besides, the Malaysian palm oil industry has grown to become a very important agriculture-based industry, where the country is today one of the world’s leading producer and exporter of palm oil.

![Figure 1. Global consumption of palm oil from 1995/1996 to 2014/2015 (USDA, 2016) [4].](image-url)
Figure 3 depicts the statistics production of palm oil superseded soybean oil from 13% in 1990 to 28% of total oil and fats production in 2011. This is because oil palm has higher annual oil yield per hectare than other oil seeds crops including soybean [11] and palm oil has a relatively lower price as compared to the major alternative vegetable oils [12]. POME is highly polluted wastewater if not treated properly; it causes a lot of environment issues. POME is a colloidal suspension of 95–96% water, 0.6–0.7% oil and 4–5% total solids including 2–4% suspended solids originating from mixture of a sterilizer condensate, separator sludge and hydrocyclone wastewater [13]. The conventional treatment technology of POME employed in most of the palm oil mills in Malaysia is the ponding system of biological treatment [14–16]. However, coping with the increasing production in most palm oil mills, the undersized biological treatment system is unable to cope with the increased volume of POME [17]. Thus, proper POME treatment is urgently needed to ensure the sustainable economic growth of palm oil industry in Malaysia besides protecting the environment. Several researchers have proposed other biological treatments.

The main objective of this study was to evaluate the performance and kinetics of the new designed ultrasonic membrane anaerobic system (UMAS) in the treatment of palm oil mill effluent (POME) based on three models [22–24]. Table 1 shows mathematical expressions for specifics substrate utilization rate for three kinetic models (Monod, Contois, and Chen and Hashimoto).
1.1. Mechanisms of anaerobic digestion

In anaerobic degrading of POME, biogas is formed when microorganisms, especially bacteria, degrade organic material in the absence of oxygen. Biogas consists of 50–75% methane (CH\textsubscript{4}), 25–45% carbon dioxide (CO\textsubscript{2}) and small amounts of other gases [25–27]. A simplified schematic representation of anaerobic degradation of organic matter is given in Figure 4. The AD process can be subdivided into the following four phases, each requires its own characteristic group of microorganisms.

The sequence of reactions involved in the mechanisms of AD is hydrolysis, acidogenesis, acetogenesis and methanogenesis [28]. Hydrolysis is conversion of nonsoluble biopolymers to soluble organic compounds. Acidogenesis is summarized as a conversion of soluble organic compounds to volatile fatty acids (VFA) and CO\textsubscript{2} while acetogenesis is the conversion of VFAs to acetate and H\textsubscript{2} [29]. Methanogenesis represents conversion of acetate and CO\textsubscript{2} plus H\textsubscript{2} to methane and carbon dioxide gas.

### Table 1. Mathematical expressions of specifics substrate utilization rates for known kinetic models.

<table>
<thead>
<tr>
<th>Kinetic Model</th>
<th>Equation 1</th>
<th>Equation 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Monod</td>
<td>$U = \frac{K_S S}{K + S}$</td>
<td>$\mu = \frac{K_S}{K_S + b}$</td>
</tr>
<tr>
<td>Contois</td>
<td>$U = \frac{\mu_{max} \times S}{Y(B \times X + S)}$</td>
<td>$\mu = \frac{\mu_{max} \times S}{\mu_{max} + \frac{Y(1 + a)}{B \times X + S}}$</td>
</tr>
<tr>
<td>Chen &amp; Hashimoto</td>
<td>$U = \frac{\mu_{max} \times S}{Y(K_S + (1 - K) \times Y)}$</td>
<td>$\mu = \frac{Y \times K_S}{Y \times (1 + a) \times \mu_{max}}$</td>
</tr>
</tbody>
</table>
2. Materials and methods

2.1. Raw POME wastewater preparation

The raw POME was collected from a near local palm oil mill in Lebah Hillier, Kuantan, Malaysia. The raw POME was stored in a cold room at 4°C before use. Different dilutions of POME were prepared using tap water. The pH of the feed was adjusted to 7.0 using a 6 N NaOH solution.

2.2. UMAS bioreactor operation and experimental setup

A laboratory scale, with an effective 200-L UMAS reactor (Figure 5), was used in this study. The UMAS reactor consists of a cross-flow ultrafiltration membrane apparatus, a centrifugal pump and an anaerobic reactor. The total volume of the reactor was 200 L, and the working volume was 150 L. Six multifrequency ultrasonic transducers, operated at 25 KHz, are bonded to two sides of the tank chamber and connected to a Crest Genesis Generator (250 W, 25 KHz; Crest Ultrasonic, Trenton, NJ, USA). The maximum operating pressure on the membrane was 55 bars at 70 WC, and the pH ranged from 2 to 12.

2.3. Analytical methods

The following parameters were analyzed: COD, BOD, pH, VSS and TSS.

Methane gas was determined by gas chromatography with a stainless steel column (200 × 0.3 cm) packed with active carbon (30–60 mesh) using thermal conductivity detection. For TSS, VSS, volatile fatty acids and alkalinity were determined according to the standard methods [31]. The COD was measured using a Hach colorimetric digestion method (Method # 8000, Hach Company, and Loveland, CO, USA). The MLSS and MLVSS were determined by drying the sample at 105 and 550 ± 50°C.
2.4. Bioreactor operation

The steady-state performance of ultrasonic membrane anaerobic system (UMAS) was evaluated under different influent COD concentrations (70,400–90,200 mg/l), hydraulic retention time (HRTs) (500.8–14.7 days) and OLR of 1.5–9.0 kg COD/m$^3$/day (Table 2). In this study, the system was considered to have achieved steady state when the operating and control parameters were within ±10% of the average value. The produced biogas contained only CO$_2$ and CH$_4$, so the addition of sodium hydroxide solution (NaOH) to absorb CO$_2$ effectively isolated methane gas (CH$_4$). Table 2 depicts the results of the application of three known substrate utilization models.

![Experimental setup](image)

Table 2. Summary of results (SS: steady state).

<table>
<thead>
<tr>
<th>Steady state (SS)</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
</tr>
</thead>
<tbody>
<tr>
<td>COD feed, mg/L</td>
<td>70,400</td>
<td>73,478</td>
<td>76,200</td>
<td>83,570</td>
<td>86,700</td>
<td>90,200</td>
</tr>
<tr>
<td>COD permeate, mg/L</td>
<td>1197</td>
<td>1617</td>
<td>3048</td>
<td>3343</td>
<td>4508</td>
<td>6494</td>
</tr>
<tr>
<td>Gas production (L/day)</td>
<td>290</td>
<td>310</td>
<td>340</td>
<td>400</td>
<td>480</td>
<td>540</td>
</tr>
<tr>
<td>Total gas yield, L/g COD/day</td>
<td>0.48</td>
<td>0.53</td>
<td>0.58</td>
<td>0.67</td>
<td>0.78</td>
<td>0.81</td>
</tr>
<tr>
<td>% Methane</td>
<td>81</td>
<td>78.5</td>
<td>75.6</td>
<td>73.8</td>
<td>68.6</td>
<td>64.6</td>
</tr>
<tr>
<td>CH$_4$ yield, l/g COD/day</td>
<td>0.39</td>
<td>0.54</td>
<td>0.57</td>
<td>0.60</td>
<td>0.64</td>
<td>0.70</td>
</tr>
<tr>
<td>MLSS, mg/L</td>
<td>13,800</td>
<td>12,400</td>
<td>13,400</td>
<td>14,800</td>
<td>17,648</td>
<td>22,600</td>
</tr>
<tr>
<td>MLVSS, mg/L</td>
<td>10,269</td>
<td>10,751</td>
<td>11,765</td>
<td>13,320</td>
<td>15,530</td>
<td>20,159</td>
</tr>
<tr>
<td>% VSS</td>
<td>74.41</td>
<td>86.70</td>
<td>87.80</td>
<td>90.00</td>
<td>88.00</td>
<td>89.20</td>
</tr>
<tr>
<td>HRT, day</td>
<td>500.8</td>
<td>60.6</td>
<td>22.6</td>
<td>14.7</td>
<td>11.20</td>
<td>8.6</td>
</tr>
<tr>
<td>SRT, day</td>
<td>300</td>
<td>250</td>
<td>180</td>
<td>30.5</td>
<td>20.30</td>
<td>15.80</td>
</tr>
<tr>
<td>OLR, kg COD/m$^3$/day</td>
<td>1.0</td>
<td>3.5</td>
<td>6.0</td>
<td>8.5</td>
<td>11.0</td>
<td>15</td>
</tr>
<tr>
<td>SSUR, kg COD/kg VSS/day</td>
<td>0.164</td>
<td>0.195</td>
<td>0.252</td>
<td>0.263</td>
<td>0.294</td>
<td>0.314</td>
</tr>
<tr>
<td>SUR, kg COD/m$^3$/day</td>
<td>0.023</td>
<td>0.724</td>
<td>2.225</td>
<td>4.576</td>
<td>5.685</td>
<td>7.347</td>
</tr>
<tr>
<td>Percent COD removal (UMAS)</td>
<td>98.3</td>
<td>97.8</td>
<td>96</td>
<td>96.0</td>
<td>94.8</td>
<td>92.8</td>
</tr>
</tbody>
</table>
3. Results and discussion

3.1. The performance of ultrasonic membrane anaerobic system (UMAS)

The operating conditions for the ultrasonic membrane anaerobic system (UMAS) over the 500-day experimental setup are given in Table 2. The performance evaluation of the integrated ultrasonic membrane anaerobic system (UMAS) was generated at different influent COD concentrations and hydraulic retention times (HRTs). Table 3 depicted and summarized the kinetic coefficients. For the system results at influent COD concentrations from 70,400 to 90,200 mg/l and pH (6.7–7.8), UMAS was performed well. The mixed liquor volatile suspended solids (MLVSSs) for the first steady state were 10,400 mg/l, whereas the mixed liquor suspended solids (MLSSs) were 13,800 mg/l, equivalent to 75.36% of the MLSS. This low result can be explained due the palm oil mill effluent wastewater contains very high suspended solids. The volatile suspended solid (VSS) fraction in the reactor at sixth steady state was increased to 89.20%. Results have shown that the long solid retention time (SRT) of UMAS facilitated the decomposition of the suspended solids and their subsequent conversion to methane (CH\textsubscript{4}); these findings found by Nagano et al. [32] and Abdurahman et al. [33]. At organic loading rate, OLR of 15 kg COD/m\textsuperscript{3}/day, the system registered the highest influent of COD 90,200 mg/l at this stage; the UMAS achieved 92.8% COD removal. Figures 6–8 shown that UMAS can be applied and treat POME efficiently. Among the three models applied, the Monod and Chen

<table>
<thead>
<tr>
<th>Model</th>
<th>Equation</th>
<th>$R^2$(% )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Monod</td>
<td>$U = 2025 \frac{S}{1} + 3.61$</td>
<td>99.6</td>
</tr>
<tr>
<td></td>
<td>$K = 498$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$K = 0.350$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$\mu_{\text{max}} = 0.284$</td>
<td></td>
</tr>
<tr>
<td>Contois</td>
<td>$U = 0.306 \times S 1 + 2.78$</td>
<td>99.1</td>
</tr>
<tr>
<td></td>
<td>$B = 0.111$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$u = 0.344$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$a = 0.115$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$\mu_{\text{max}} = 0.377$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$K = 0.519$</td>
<td></td>
</tr>
<tr>
<td>Chen &amp; Hashimoto</td>
<td>$U = 0.0190 \times S 1 + 3.77$</td>
<td>99.5</td>
</tr>
<tr>
<td></td>
<td>$K = 0.006$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$a = 0.006$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$\mu_{\text{max}} = 0.291$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$K = 0.374$</td>
<td></td>
</tr>
</tbody>
</table>

Table 3. Summary of the three known substrate utilization models application.
and Hashimoto models performed better, shown that UMAS reactor performance should consider organic loading rates. These two models suggested that the predicted permeate COD concentration ($S$) is a function of influent COD concentration ($S_0$).

The percentage removal of COD by UMAS at various HRTs was shown in Figure 9. It was observed that COD removal efficiency increased as HRT increased from 8.6 to 500.8 days and it was in the range of 92.8–98.3%. It was found that this value higher than the 85% COD removal is observed for POME wastewater treatment using anaerobic fluidized bed reactors [34] and the 91.7–94.2% removal is observed for palm oil mill effluent wastewater treatment using membrane anaerobic system [35], and the 93.6–97.5% removal is observed for POME treatment using membrane anaerobic system [33]. Interestingly, it was found that there is no much difference in COD removal efficiency between HRTs of 500.8 days (98.3%) and 14.7 days.

Figure 6. The Monod model.

Figure 7. The Contois model.
(96.0%). On the other hand, the COD removal efficiency has declined at shorter hydraulic retention time; at HRT of 8.6 days, the COD removal efficiency was reduced to 92.8%. **Table 2** results show that UMAS result might because of grown of volatile fatty acids inside the reactor. Usually, the hydraulic retention times were mainly effected by the ultrafiltration (UF) membrane influx rates, which directly determined the volume of influent (POME) that can be fed to the reactor.

### 3.2. Evaluation of UMAS biokinetic coefficients

The evaluated biokinetic coefficients based on COD basis by UMAS were analyzed as shown in **Table 2**.
The kinetic coefficients were calculated and summarized in Table 3. The growth yield coefficient, Y, value ranges from 0.32 to 0.68 gm VSS/gm COD, specific microorganic decay rate, b, and maximum substrate utilization rate, K, ranges from 0.350 to 0.374 COD/g VSS.day. Figure 10 depicts the relationship between the substrate utilization rates (SUR) and the specific substrate utilization rate for COD with various hydraulic retention times. The HRTs range from 8.6 to 500.8 days. The biokinetic coefficients of growth yield, Y, and specific microorganic decay rate, b, were calculated from the slope and intercept as shown in Figures 11 and 12. The evaluated maximum specific biomass growth rates, $\mu_{\text{max}}$, range from 0.248 to 0.474 day$^{-1}$.

![Figure 10](image1.png)

**Figure 10.** The specific substrate utilization rate for COD with various hydraulic retention times.

![Figure 11](image2.png)

**Figure 11.** Evaluation of the growth yield, Y, and the specific biomass decay rate, b.
4. Production of methane (CH$_4$) and carbon dioxide (CO$_2$) gases

A semicontinuous operation was conducted to verify the performance of the integrated ultrasonic membrane anaerobic system (UMAS) throughout a different hydraulic retention times (HRTs) and influent COD concentrations. In this study, the influent COD concentration was increased from 70,400 to 90,200 mg/l (for the six steady states). Figure 13 illustrates the gas production rate and the methane content of the biogas. It was clear that the methane CH$_4$ yield decreased with increasing OLRs. Methane gas contents were varied from 64.6 to 81%, and the methane yield was varied from 0.39 to 0.70 CH$_4$/g COD/day. The decreased CH$_4$ yield with increasing OLR was also

![Figure 12. Evaluation of the maximum specific substrate utilization and the saturation constant, K.](image-url)

![Figure 13. Gas production and methane content.](image-url)
noted in many previous studies [36–40]. One of the reasons might be that shorter HRT of the system contributed to more active methanogens that were washed out during the removal of effluent. The gas production has increased from 290 to 540 L per day during the study. Biogas production increased with increasing OLRs from 0.48 l/g COD/day at 1.0 kg COD/m³/day to 0.81 l/g COD/day at 15 kg COD/m³/day. These findings are in line with the results obtained from Refs. [41–43].

5. Conclusions

The kinetic performance of newly designed ultrasonic membrane anaerobic system (UMAS) was evaluated in the treatment of palm oil mill effluent (POME).

The steady-state performance of ultrasonic membrane anaerobic system (UMAS) was evaluated under different influent COD concentrations (70,400–90,200 mg/l), hydraulic retention times (HRTs) (500.8–14.7 days) and OLR of 1.5–9.0 kg COD/m³/day.

Among the three models applied, the Monod and Chen and Hashimoto models performed better, shown that UMAS reactor performance should consider organic loading rates. These two models suggested that the predicted permeate COD concentration (S) is a function of influent COD concentration (S). It was observed that COD removal efficiency increased as HRT increased from 8.6 to 500.8 days, and it was in the range of 92.8–98.3%. The evaluated maximum specific biomass growth rates, $\mu_{\text{max}}$, range from 0.248 to 0.474 day⁻¹.

It was found that the methane CH₄ yield decreased with increasing OLRs. Methane gas contents were varied from 64.6 to 81%, and the methane yield was varied from 0.39 to 0.70 CH₄/g COD/day.

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