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Pollutants analysis during conventional palm oil mill effluent (POME) ponding system and decolourisation of anaerobically treated POME via calcium lactate-polyacrylamide

A.Y. Zahrim [a,b,*], A. Nasimah [a], N. Hilal [c,d]

[a] Chemical Engineering Programme, Faculty of Engineering, Universiti Malaysia Sabah, Jalan UMS, 88400 Kota Kinabalu, Sabah, Malaysia
[b] Sustainable Palm Oil Research Unit, Universiti Malaysia Sabah, Jalan UMS, 88400 Kota Kinabalu, Sabah, Malaysia
[c] Centre for Water Advanced Technologies and Environmental Research (CWATER), College of Engineering, Swansea University, Swansea SA2 8PP, UK
[d] Qatar Energy and Environment Research Institute (QEERI), Doha, Qatar

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ABSTRACT

The conventional palm oil mill effluent (POME) ponding system is unable to fully decolourise the effluent which is aesthetically important. Several parameters, such as low molecular mass coloured compounds (LMMCC), lignin–tannin, ammonia nitrogen (NH_3-N) and chemical oxygen demand (COD) in the cooling pond, are much higher than for the fresh raw POME. The analysis of the POME from each pond revealed that the removal of lignin–tannin is insignificant after anaerobic ponds and hence physicochemical treatment is necessary. The POME colloid repulsion in the aerobic pond is greater than in the anaerobic pond. The coagulation/flocculation process was utilized to destabilize the anaerobically treated POME (AnPOME) colloid and calcium lactate was chosen as a coagulant. The best polymer order was identified based on an overall removal performance. The best polymer can be arranged as QF23912 (58%) > QF25610 (57%) > AN1500 (51%) > QF24807 (50%) > AN1800 (47%). All tested polymers have similarity in removing NH_3-N. It can be concluded that calcium lactate-cationic polymer has potential as a pre-treatment for AnPOME.

1. Introduction

Agriculture has covered more than half of the world's rainforest and the most widespread tropical crop is the oil *palm* (*Elaeis guineensis*). The countries that contribute to the production of palm oil are in South East Asia, especially Indonesia and Malaysia; Papua New Guinea; Colombia; Ivory Coast; Nigeria and Thailand [1]. The operation of a palm oil mill requires large volumes of water and subsequently discharges high volumes of palm oil mill effluent (POME) [2]. Approximately, 1.0–1.5 tonnes of water is required to process 1 tonne of fresh fruit bunches, while each tonne of crude palm oil produced generates approximately 2.5 m^3–3.5 m^3 of wastewater [1]. The POME may vary in quality and quantity by different batches, days and factories, depending on the oil palm cropping seasonal and palm oil mill operation (occasional public holidays, closure of the mill, operation and quality control of individual mills) [3,4]. The conventional treatment consists of anaerobic followed by aerobic ponding system. Upgrading the conventional ponding system is vital since the anaerobically treated effluent (AnPOME) still consist of dark brown colour. The coloured treated POME is often objected to by the public on the assumption that colour is a sign of pollution.

It is believed that the colour is derived from plant constituents such as lignin, tannin, humic and fulvic acid like substance and phenolic compounds as well as re-polymerization of colouring compounds ([2,5,6]). Jamal et al. [7] reported that the palm oil mill effluent contains several phenolics compounds such as gallic, protocatechuic, 4-hydroxybenzoic, 4-hydroxylphenylacetic, caffeic, syringic acids, p-coumaric and ferulic acids [7]. To enhance the AnPOME treatment, we propose an integrated treatment that consists of coagulation/flocculation-depth filtration-biological treatment/membrane separation. In this paper, we focus on the coagulation/flocculation of AnPOME as a pre-treatment.

As a pre-treatment process, coagulation/flocculation can be effective to enhance water/wastewater treatment performance towards water reclamation and reuse [8]. It is simple, inexpensive, has a good removal efficiency and easy on-site implementation [9] and may enhance the biodegradability of the wastewater [10]. Several studies using conventional coagulants i.e. aluminium and

* Corresponding author at: Chemical Engineering Programme, Faculty of Engineering, Universiti Malaysia Sabah, Jalan UMS, 88400 Kota Kinabalu, Sabah, Malaysia. Tel.: +60 88320000; fax: +60 88320348.
E-mail address: zahrim@ums.edu.my (A.Y. Zahrim).

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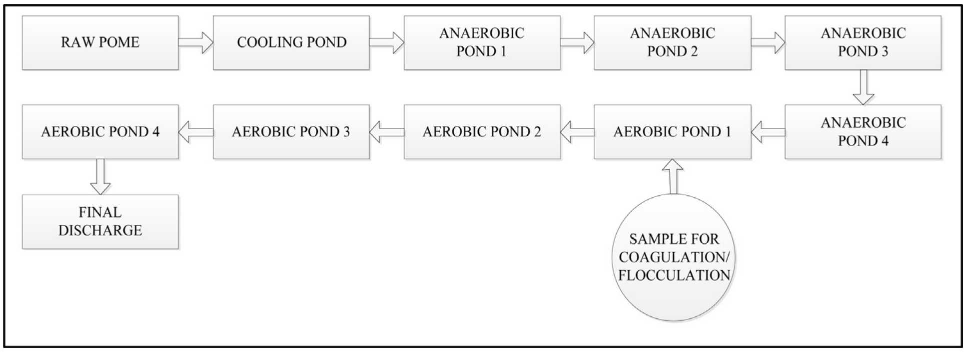

Fig. 1. The flowchart of ponding treatment system of POME at Lumadan Palm Oil Mill, Beaufort, Sabah.

iron based coagulants have been studied for palm oil mill effluent treatment. Ahmad et al. [8] reported that aluminium sulphate was effective at pH 6. Tan et al. [1] combined ferric sulphate-aluminium sulphate-ammonium sulphate as coagulants and found that it reduced turbidity and COD at 97% and 64%, respectively [1]. Recently, Jami et al. [11] reported that ferric chloride is superior to aluminium sulphate during coagulation/flocculation of biological treated POME. However, the residual alum and ferric based coagulants might inhibit the post biological treatment process which is indicated by the reduction of the microorganism respiration rate and low organic matter removal [12] and could also contribute to membrane fouling [13].

Currently, more attention has been given to natural coagulants for their eco-friendliness, biodegradability and freedom from secondary pollution risks, in comparison with conventional metal-based coagulants, particularly for POME treatment [14–16]. Recently, Devesa-Rey et al. [17] reported that calcium lactate, which is a biodegradable and non-toxic coagulant, could reduce synthetic solution turbidity by more than 90%. Devesa-Rey et al. [18] also reported that the performance of calcium lactate is comparable with that of aluminium based coagulants ([18]; [17]).

The objective of this study is to determine the coagulation/flocculation performance during AnPOME treatment. To the best of the authors' knowledge, no published study has previously been carried out for the treatment of palm oil mill effluent using calcium lactate-polymer. Polymer addition improves the size of the destabilized flocs and consequently enhances the settling rate, provided the correct dosage is added [19]. Past investigations reported that the application of polymer has been shown to be effective in removal of POME pollutants [20–22]. Prior to that, the characteristic of anaerobic-aerobic pond during biological treatment of POME will be investigated. It is anticipated that the data obtained in this study will help to identify which polymer would be most effective in the treatment of AnPOME.

2. Methodology

2.1. Materials and analysis

The anaerobically treated palm oil mill effluent (AnPOME) was collected from the Beaufort, Sabah. The POME ponding system contained a cooling/grid pond, four stages of anaerobic ponds and four stages of aerobic ponds (Fig. 1). Generally, the hydraulic retention time (HRT) for the ponding system is 45–60 days [23].

Several parameters, such as low molecular mass coloured compounds (LMMCC), lignin–tannin, ammonia nitrogen (NH_3-N) and Chemical Oxygen Demand (COD), were analyzed. The COD was tested using the Spectrophotometer HACH DR 2010. The absorbance at λ_{max} = 290 nm was used to analyze LMMCC with a V-650 UV/V is spectrophotometer (Jasco). The lignin–tannin and NH_3-N content were tested via the Tyrosine and Nessler Method, respectively [24]. The pH and conductivity was measured by using metre HI 9611-5 (Hanna Instrument). The zeta potential was determined by using Malvern-Zetasizer Nano Series model ZS machine.

2.2. Jar test

During the jar tests, the appropriate volume of AnPOME (without any pH adjustment) was transferred into a round jar. Next, 0.5 g/L of calcium lactate (Molecular mass 218 g/mol) (Merck, Germany) was added to the solution in the jar. The calcium lactate dosage is based on our preliminary study [25], which focused on the application of calcium lactate alone. A standard flocculator apparatus (Phipps & Birds) equipped with stainless steel paddles and stirrer was used for the coagulation/flocculation tests. The aqueous solution was then rapidly mixed at a paddle speed of 258 rpm for 3 min. Predetermined dosage of polyacrylamide (PAM) (supplied by Guangxi Nanning Bonglin Business &Trade Co., Ltd, China) i.e. 50 mg/L was added to the solution in the jar, making the total volume of 500 mL, followed by slow mixing for 20 min at 39 rpm. Since our final aim is to utilize membrane separation in the final stage, the dosage of 50 mg/L polymer was chosen because the polymer dosage was reported to have minimal effect on membrane fouling [26]. After allowing settling to occur (30–120 min), about 25 mL of the liquid was withdrawn using a pipette from a height of about 3 cm below the liquid surface in each jar [19]. The jar test was conducted in room temperature.

3. Results and discussion

3.1. Evolution of several parameters during POME treatment in different ponds

The physicochemical characteristics of POME in each treatment pond are shown in Figs. 2–6. The colour evolution at palm oil mill wastewater treatment plant is shown in Fig. 2(a). As expected, the fresh raw POME and cooling pond contains high concentrations of low molecular mass coloured compounds (LMMCC), lignin–tannin, ammonia nitrogen (NH_3-N), and high chemical oxygen demand (COD) and total suspended solids (TSS). However, several parameters for the cooling pond were measured to be much higher than the fresh raw POME. This might be due to the effect of the combination of the old and fresh POME.

Fig. 2(b) shows the evolution of pH and conductivity during the wastewater treatment. The raw POME was acidic at pH 4.3 and pH values continued to rise slowly during anaerobic treatment until anaerobic pond 2. This might be due to consumption of H^+ during methanogenic digestion [27]. Biomethane is usually produced by methanogenic bacteria from acetic acid, hydrogen and carbon dioxide as well as other substrates such as formic acid

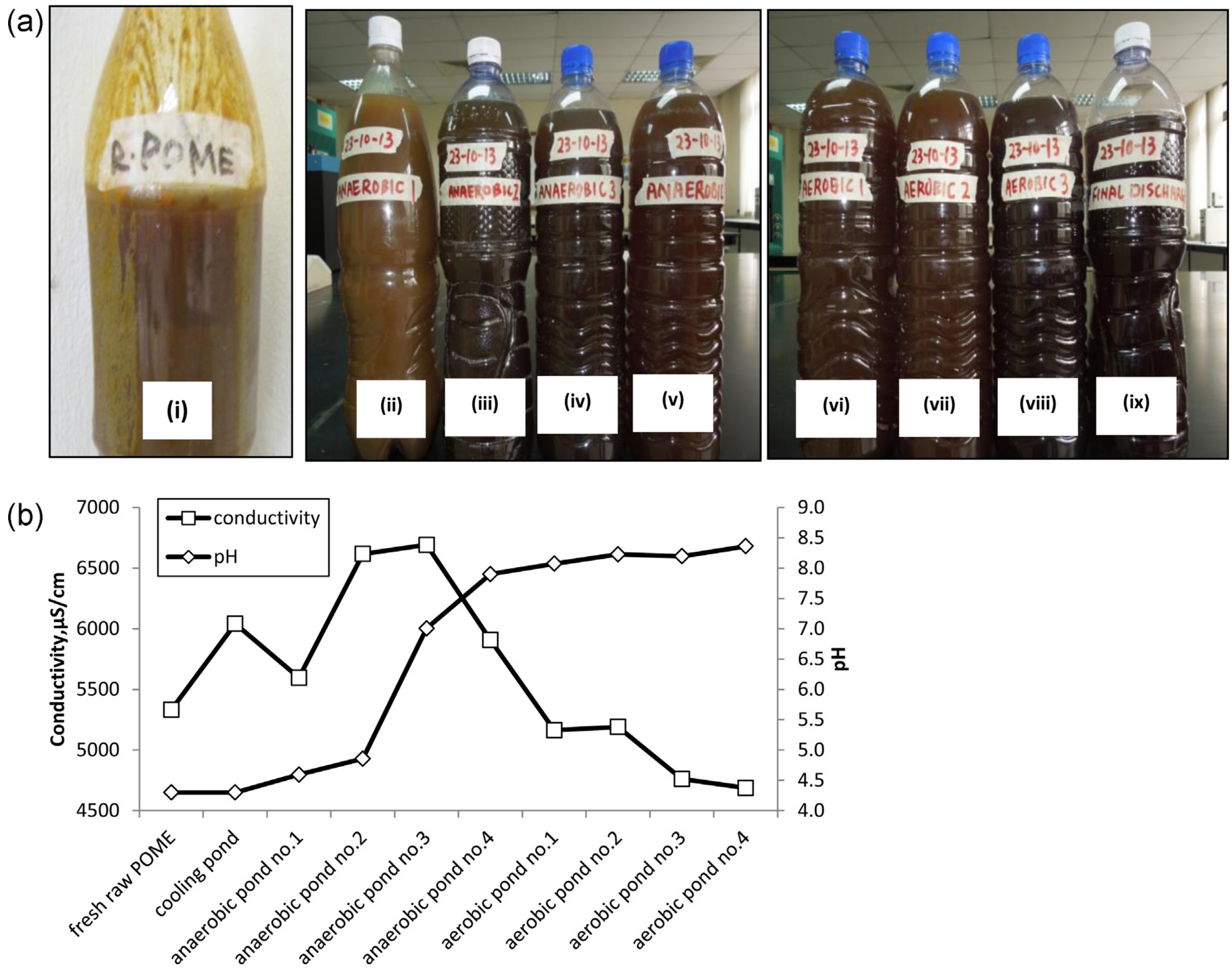

Fig. 2. (a) Evolution of colour at palm oil mill wastewater treatment plant: (i) raw POME (ii) anaerobic pond no. 1 (iii) anaerobic pond no. 2 (iv) anaerobic pond no. 3 (v) anaerobic pond no. 4 (vi) aerobic pond no. 1 (vii) aerobic pond no. 2 (viii) aerobic pond no. 3 (ix) aerobic pond no. 4/final discharge. (b) Conductivity and pH evolution at palm oil mill wastewater treatment plant.

and methanol during methanogenic stage [28]. After anaerobic pond 2, the pH increases sharply, which may be due to the higher H^+ consumption at anaerobic pond no.3 than anaerobic pond no. 2. At anaerobic pond no.3 to aerobic pond no.4 the wastewater had become alkaline, with pH values in the range 7.0–8.4. After anaerobic pond no. 4, the pH was around 8.0–8.5 at the aerobic pond.

At the anaerobic treatment ponds, the conductivity shows an increasing trend while at the aerobic pond, the conductivity is continuously decreasing. The increase in conductivity might be due

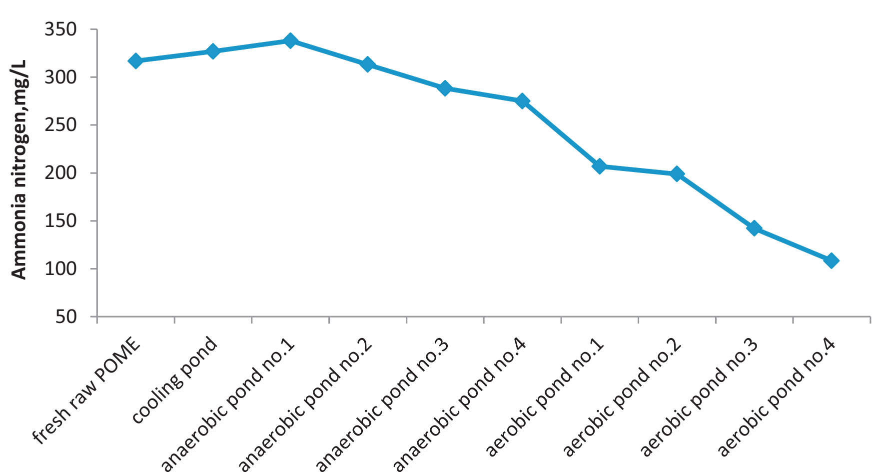

Fig. 3. NH_3-N evolution at palm oil mill wastewater treatment plant.

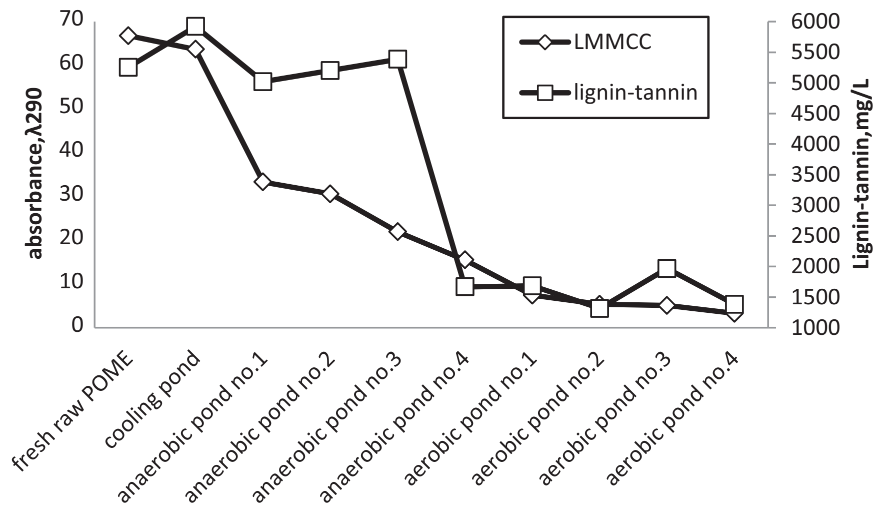

Fig. 4. LMMCC and lignin–tannin evolution at palm oil mill wastewater treatment plant.

to the formation of inorganic nitrogen compounds (NH_4^+) during anaerobic digestion [27] and the release of phosphate, potassium and magnesium [29]. On the other hand, the decrease of conductivity is due to the phosphate, potassium and magnesium uptake by the biomass [29] and volatilization of NH_3 into the atmosphere [30,31]. The trace mineral that is trapped in the biomass [32] may settle during aerobic treatment.

Fig. 3 shows the removal of ammonia nitrogen at the wastewater treatment plant. At high concentrations, inorganic nitrogenous compounds (NH_4^+, NH_3, NO_2^-, HNO_2, NO_3^-) are toxic towards aquatic life and cause excessive oxygen demand in the receiving water [33]. From Fig. 2, it can be seen that the ammonia nitrogen loss was about 13%, which might be due to volatilization during anaerobic treatment, with a further 61% removed after aerobic treatment.

Fig. 4 shows the evolution of LMMCC and lignin–tannin removal efficiency during conventional anaerobic-aerobic ponding system treatment. From the analysis of statistical variances (ANOVA) for wastewater from four aerobic ponds (Fig. 4); the LMMCC ($F = 0.1059 < 4.0662$) and lignin–tannin ($F = 3.968 < 4.0662$), were insignificantly reduced [34]. The pattern of LMMCC and lignin–tannin is similar. At anaerobic pond no. 1–3, it was observed that the concentration of lignin–tannin was significantly higher than LMMCC. This is due to the high solubility of LMMCC and hence it is easily digested by the microorganisms, as indicated in Fig. 4. By observation, the colour started to become dark yellow after anaerobic pond no. 4. In oil palm processing, as there is no chemical addition, the existing colour from this effluent appears to be from plant constituents such as lignin and phenolics as well as re-polymerization of colouring compounds after anaerobic treatment [2].

Fig. 5 shows the removal of total COD and TSS at the wastewater treatment plant. The cooling pond shows higher Total COD and TSS than the fresh raw POME. This may be due to the oil residue floating in the supernatant phase [14]. In this study, total COD and TSS after treatment by four anaerobic ponds achieve 93.9% and 88.7% of removal, which indicates the high efficiency of the anaerobic treatment used. The high efficiency of COD and TSS removal at anaerobic treatment was due to the sedimentation of particulate matters into the bottom surface of the pond [35]. Although the pollutants' removal for the conventional ponding system is high, it takes a long time to be achieved (45–60 days). Therefore, several studies have been dedicated to shortening the hydraulic retention time during anaerobic digestion of POME [36,37]. As expected, Fig. 6 shows that POME are negatively charged and the POME tend to be more negative in aerobic pond due to the presence of highly concentrated hydroxide ion (Fig. 2b). High negatively charge colloid leads to high repulsion between the POME colloids. Therefore, coagulation/flocculation can be applied to destabilize the colloid.

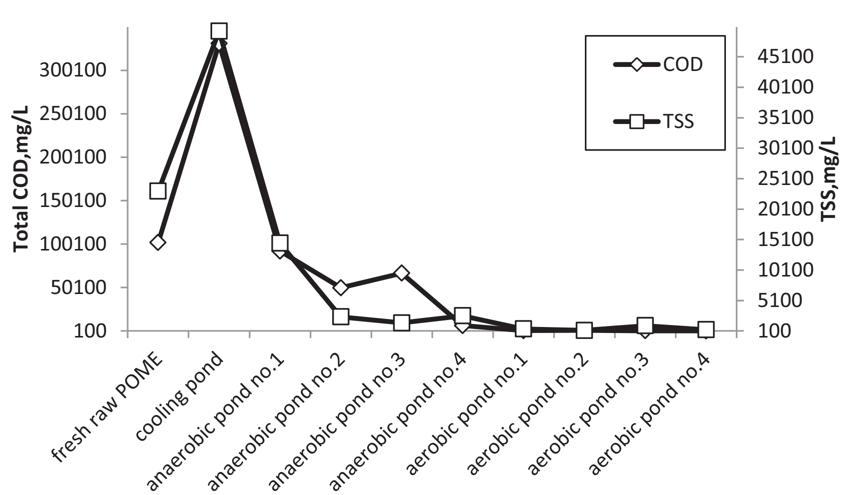

Fig. 5. Total COD and TSS evolution at palm oil mill wastewater treatment plant.

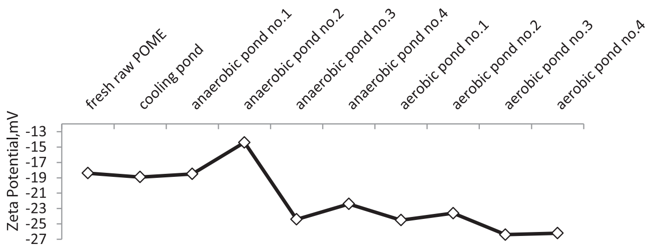

Fig. 6. Zeta Potential evolution at palm oil mill wastewater treatment plant.

3.2. Coagulation/flocculation of AnPOME

3.2.1. Effect of various types of polymer

Fig. 7 shows the effect of various types of cationic/anionic polyacrylamide (QF25610 (cationic), QF24807 (cationic), QF23912 (cationic), AN1500 (anionic) and AN1800 (anionic)) on the removal of LMMCC, lignin–tannin, ammonia nitrogen and COD during coagulation of AnPOME with calcium lactate at 1 h sedimentation time. The "Control" indicates the application of calcium lactate without any polyacrylamide. According to previous studies, the mechanism for calcium could be sweep coagulation [38], adsorption [39] and precipitation [40].

The LMMCC removal for control (calcium lactate only), QF25610, QF24807, QF23912, AN1500 and AN1800 is 21%, 53%, 25%, 44%, 26% and 28%, respectively (Fig. 7).

The highest LMMCC removal by two types of cationic PAM, i.e. QF25610 and QF23912; might be due to the increasing in the charge neutralization capacity [41] compared to calcium lactate alone. Hence, the flocs produced were more compact and more resistant to shear degradation [19]. Little LMMCC removal for both anionic PAMs may be due to the easily breakable flocs which have resulted from a bridging mechanism [41]. In another study, it was reported that the application of anionic PAM for pulp and paper mill effluent treatment also produces small and loose flocs [42].

The lignin–tannin removal for calcium lactate alone, QF25610, QF24807, QF23912, AN1500 and AN1800 is 44%, 56%, 54%, 59%, 51% and 49%, respectively (Fig. 7). The lignin–tannin removal is similar for cationic and anionic PAMs.

The NH_3-N removal for calcium lactate alone, QF25610, QF24807, QF23912, AN1500 and AN1800 is 46%, 52%, 52%, 53%, 52% and 47%, respectively (Fig. 7). The NH_3-N are attracted to the negatively charged flocs and then, the NH_3-N are removed as the flocs settling down [43].

The COD removal for calcium lactate alone, QF25610, QF24807, QF23912, AN1500 and AN1800 is 71%, 69%, 69%, 76%, 76% and 64%, respectively. The COD removal was considerably high for the control due to the presence of highly concentration of suspended solids that are easily coagulated. Only QF23912 and AN1500 showed a slight increase of COD removal compared with the calcium lactate treatment alone. This finding is similar to that observed by Wong et al. [44] who reported that the COD removal efficiency during treatment of pulp and paper mill effluent was not influenced by the molecular mass or charge density of the polymer [44].

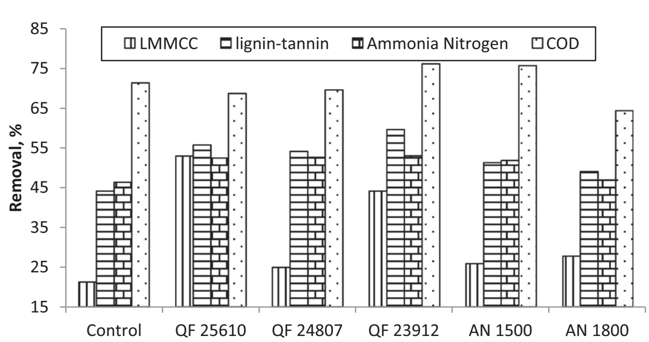

Fig. 7. Effect of polyacrylamides on the LMMCC, lignin–tannin, NH_3-N and COD removal during coagulation of AnPOME with 0.5 g/L calcium lactate (Sedimentation time: 1 h).

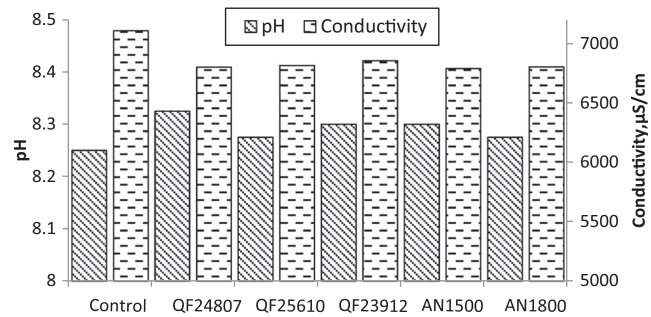

Fig. 8. Effect of polyacrylamides on the pH and conductivity during coagulation of AnPOME with 0.5 g/L calcium lactate (Sedimentation time: 1 h).

The ranking of the polymers in order of best performance was identified based on overall removal. The overall removal is the average value from the summation of LMMCC, lignin–tannin, NH_3-N and COD removal. The overall removal for calcium lactate alone is 45.5%. From the average, the most effective polymer can be arranged as QF23912 (58%) > QF25610 (57%) > AN1500 (51%) > QF24807 (50%) > AN1800 (47%). The low efficiency of AN1800 for the removal of LMMCC, lignin–tannin, NH_3-N and COD might be due to the slow adsorption rate of AN1800 towards particles [45].

The importance of polymers application compare to calcium lactate alone (control) was evaluated by determining the %removal increments. The % removal increment is defined as:

$$\frac{\%\,\text{Removal due to polymer} - \%\,\text{Removal due to control}}{\%\,\text{Removal due to control}} \times 100$$

Since the overall removal for calcium lactate alone is 45.5%, the increment for QF23912, QF25610, AN1500, QF24807, and AN1800 is 27.5%, 25.3%, 12.1%, 9.9% and 3.3%, respectively. Based on these calculations and by assuming that the significant increment should be above than 5%; all the polymers tested are believed to enhance the coagulation process except AN1800. However, the economic aspect needs to be further studied. Based on these findings, the effect of sedimentation time was studied using QF23912.

Fig. 8 shows the effect of polyacrylamides on pH and conductivity during coagulation of AnPOME with calcium lactate. The addition of calcium lactate alone or with polymer shows slight pH increment, i.e. from 8.1 to 8.25.

The pH range is suitable for the polymers during flocculation process since they function best at pH >6.0 [19]. Fig. 8 also shows that the conductivity of treated effluent decreasing after the addition of polymer. This is due to the ability of PAM to adsorb minerals [46] and inorganic nitrogenous compound [47] during flocculation.

The zeta potential values of untreated AnPOME, and supernatant from treated AnPOME (for calcium lactate alone (Control), QF25610, QF24807, QF23912, AN1500 and AN1800) were shown in Fig. 9. Higher negative charge at Control and QF25610 might be due to the residual lactate ion that is not flocculated after formation of calcium/QF25610-AnPOME complexes precipitate [43]. A shift of zeta potential to lesser negatively charge solution after cationic PAM (QF24807, QF23912) was added into the destabilized AnPOME might be due to the presence of non-reacted cationic polymer plus the residual lactate ion in the solution [46]. The higher negative charge with an addition of anionic PAM (AN1500, AN1800) is due to the presence of unreacted anionic polymer as well as lactate ion [39].

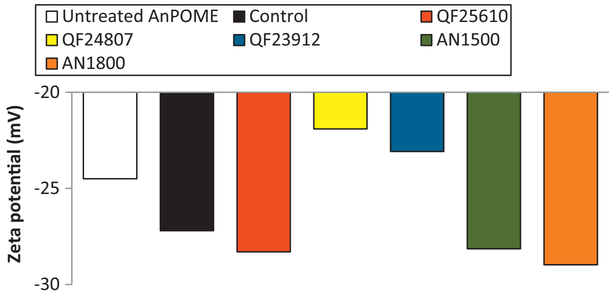

Fig. 9. Effect of polyacrylamides on the zeta potential during coagulation of AnPOME with 0.5 g/L calcium lactate (Sedimentation time: 2 h).

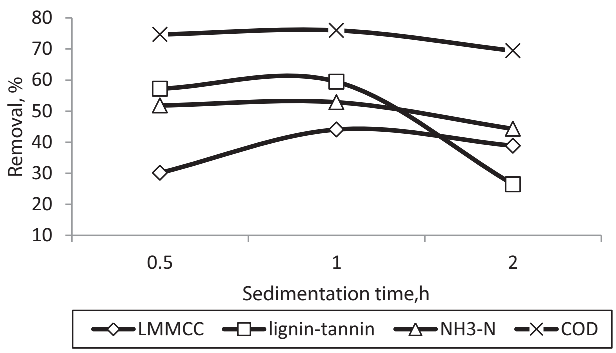

Fig. 10. Effect of sedimentation time on the LMMCC, lignin–tannin, NH_3-N and COD removal during coagulation of AnPOME with 0.5 g/L calcium lactate and QF23912.

3.3. *Effect of sedimentation time for QF23912*

Fig. 10 shows the effect of sedimentation time on the LMMCC, lignin–tannin, ammonia nitrogen and COD removal during coagulation of AnPOME with calcium lactate and QF23912. The removal of COD and NH_3-N shows similar trend probably due to the fact that the NH_3-N is adsorbed strongly to the flocs formed which has excess negatively charge surface (Fig. 9). However, the lignin–tannin removal was drop drastically might be due to the weak bonding between flocs and lignin–tannin particles. It was also observed that LMMCC take longer time to be coagulated than other pollutants might be due to very fine particles size. Generally, after 2 h sedimentation, the pollutant removal becoming lesser than 1 h sedimentation (except for LMMCC), probably due to flocs breakage [45,48].

4. Conclusion

In this study, the performance of lignin–tannin, low molecular mass coloured compounds (LMMCC), COD, ammonia nitrogen (NH_3-N), and total suspended solid (TSS) removals during conventional ponding system treatment were evaluated. The most effective polymer is QF23912 that can remove 58% of overall pollutants. After the conventional anaerobic digestion system, there is no significant change in lignin–tannin concentration and thus the coagulation/flocculation was investigated to enhance the lignin–tannin removal. It can be concluded that the addition of calcium lactate-cationic polyacrylamide can improve AnPOME treatment.

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