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Enhancing seafood processing wastewater treatment efficiency of an Anaerobic Membrane Bioreactor (AnMBR) system: a comparison of post-treatment methods

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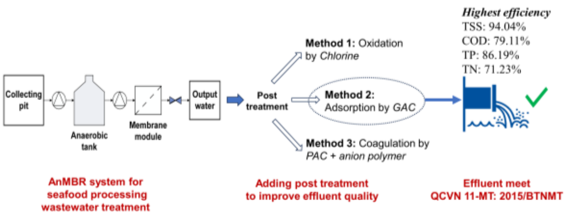
Graphical abstract



Abstract

An Anaerobic membrane bioreactor (AnMBR) was applied for treating wastewater of a seafood processing factory in Vietnam. However, Chemical Oxygen Demand (COD), Total Phosphorus (TP), and Total Nitrogen (TN) values of the AnMBR effluent were 250.43 mg/l, 19.96 mg/l, and 62.65 mg/l, respectively, and were about two times higher than the technical regulation for seafood processing wastewater in Vietnam. AnMBR needs to be combined with other post-treatment processes to ensure that treated water can meet the technical regulation. Therefore, current research has tested various advanced post-treatment methods for AnMBR effluent including chlorine, poly aluminum chloride (PAC) combined with anionic polymer, and granular activated carbon (GAC). Different concentrations of the above chemicals and reaction times were tested to select the appropriate post-treatment method. The results showed that chlorine had the lowest treatment efficiency for all four parameters (Total suspended solids – TSS, COD, TP, and TN). GAC was more effective than PAC in treatment of TN and TSS. Besides, GAC with a concentration of 5000 mg/L and a reaction time of 60 minutes brought the highest TSS, COD, TP, and TN treatment efficiency with 94.04, 79.11; 86.19, and 71.23%. This implies that GAC is the suitable method for the post-treatment of AnMBR seafood processing effluent.

Keywords: Post treatment, seafood processing wastewater, anaerobic membrane bioreactor, chlorine, polyaluminium chloride, granular activated carbon.

1. Introduction

Over the past 20 years, the seafood industry of Vietnam has always maintained a growth rate of 10%-20%. However, the seafood processing industry is also one of the industries that cause serious pollution to the environment, especially wastewater. Wastewater generated from seafood processing has high concentrations of COD, BOD_5, suspended solids, total nitrogen, and phosphorus. It can range from 800-3000 mg COD/L, in which the high proportion of biodegradable organic compounds represents the BOD_5/COD ratio ranging from 0.6-0.9 (Ngoc *et al.* 2022). Therefore, biological treatment methods are often chosen to treat this type of wastewater. Currently, most wastewater treatment systems in seafood processing factories use activated sludge tanks or up-flow anaerobic sludge blanket (UASB) tanks combined with activated sludge tanks to achieve high organics and nutrient removal (Massé *et al.* 2006).

In recent years, anaerobic membrane bioreactor (AnMBR) technology has been receiving more and more attention from scientists around the world. AnMBR has several outstanding advantages such as low sludge production capacity, lower energy demand compared to aerobic processes, and the ability to generate methane biogas and the ability to handle large organic loads (Tomar *et al.* 2023), so it is very suitable for the seafood processing industry (Kanafin *et al.* 2021; Li *et al.* 2023). However, under unfavorable conditions, such as low hydraulic retention time (HRT), psychrophilic temperature, etc., the efficiency of organic matter treatment is low (Medina *et al.* 2023), and some indicators do not meet the discharge standards output. Hence, the effective post-treatment of AnMBR effluents is needed to satisfy the discharge and the recycling standards. This is important as increasing interest in wastewater recycling due to the lack of fresh water.

In this study, post-treatment by chlorine, poly aluminum chloride (PAC) combined with anionic polymer, and granular activated carbon (GAC) were studied for AnMBR effluent from seafood processing wastewater. The COD, TN, TP, and TSS removal efficiencies were then evaluated to determine the appropriate effective post-treatment method for seafood processing wastewater.

2. Materials and methods

2.1. Configuration of the pilot-scale anaerobic membrane bioreactor (AnMBR) system

The AnMBR system was installed and operated at a seafood processing factory, in Ba Ria Vung Tau, Vietnam. AnMBR system with a capacity of 0.5 m^3/day, dimensions Length × Width × Height = 600 × 600 × 1500 (mm) is made of stainless steel. Here, anaerobic microorganisms will conveniently consume organic substances in the water. Next, the water is pumped to the MBR tank to continue removing chemicals, solids as well as disease-causing bacteria. The membrane used in the AnMBR was from Microdyn-Nadir, Germany with a molecule weight of 150,000 Da, flux LMH (L/m^2/h)/bar of 153.06/85.03 and a rejection size of 5K-Dextran and 5000 Da pore size, HRT was 10 h. To investigate different post-treatment techniques, effluent after the AnMBR system was collected and stored at 4ºC, then followed a tank that contains a 3-phase 240VAC induction motor with a maximum frequency of 50Hz and a maximum speed of 1400 revolutions per minute (RPM). A transmission with a 20:1 gear ratio was attached to the engine. Chemical coagulants were added alternately to the tank, first mixed well poly aluminum chloride, then Anionic Polymer (Figure 1).

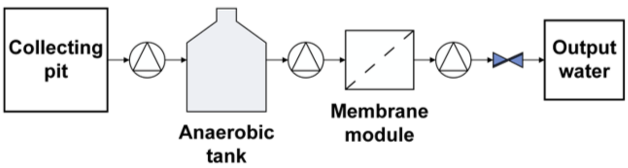


Figure 1. Anaerobic membrane bioreactor (AnMBR) system for seafood processing wastewater treatment

All chemicals with analytical grade were purchased from Bien Hoa Chemicals, Vietnam. Method of analyzing water quality parameters: pH value was measured using a handheld multi-parameter meter (HQ40d, Hach, USA). COD parameters were measured by UV-VIS spectrophotometer, according to the SMEWW 5220-D method. The TSS index was determined according to the gravimetric method TCVN 6625:2000 (filtered with 0.45 µm filter paper, dried to constant weight at temperatures 105ºC and 550ºC). Total nitrogen was determined according to the SMEWW 4500-C method, and total phosphorus was determined according to the spectrophotometric method using ammonium molybdate (TCVN 6202: 2008).

2.2. Evaluating post-treatments methods

2.2.1. Post-treatment with PAC

The post-treatment was carried out with jar testing to find out the effect of the post-treatment chemicals on the output water from the process. In this experiment, the most efficient dose of the different chemicals, which are Chlorine, Poly Aluminium Chloride with Anionic Polymer as flocculant would be tested. The jar tester used in the experiment was from Aqualytic – Germany with 6 positions for stirring, time, and speed (RPM) adjustable and turning off automatically. The chemical used in the experiment was poly aluminum chloride (PAC) - $[Al_2(OH)nCl_6\text{-}n]m$ as a coagulant and anionic polymer $CONH_2[CH_2\text{-}CH\text{-}]n$. Firstly, the coagulant was added and stirred at 30 RPM for 15 minutes then rest for 5 minutes as the chemical reaction and sedimentation with the respective dosage of 500 mg/L, 550 mg/L, 600 mg/L, 650 mg/L, and 700 mg/L. After stirring with the coagulant, 30 mg/L of anionic polymer was added to each jar as flocculation. The motor would run at 30 RPM for 15 minutes and then rest. After that, the output wastewater was analyzed to choose the optimal dosage for the coagulant that would be used for finding the optimal Anionic polymer dosage. There were 5 different anionic polymer dosages from 10 mg/L to 50 mg/L, which was: 10 mg/L, 20 mg/L, 30 mg/L, 40 mg/L, and 50 mg/L.

2.2.2. Post-treatment with chlorine

The chlorine used in the experiment was Chlorine 70%, there were 5 different dosages 1 mg/L, 2 mg/L, 4 mg/L, 6 mg/L, 8 mg/L, and 10 mg/L. After adding the Chlorine, the jar testers were run at 70 RPM for 20 minutes for mixing the chemical.

2.2.3. Post-treatment with GAC

Activated carbon used is under power form, which has a large contact area and is easily mixed under water. The doses for GAC varied from 0.2 to 5 g/L, which specificaly 1000 mg/L, 2000 mg/L, 3000 mg/L, 4000 mg/L, and 5000 mg/L. After selecting the doses, the jar test was run at 100 RPM for 30 minutes and then rested for 30 minutes as the substance precipitation to avoid errors in analyzing.

3. Results and discussion

3.1. Seafood processing wastewater treatment efficiency of AnMBR system

The analysis results in Table 1 show that the AnMBR system has effectively contributed to the seafood processing wastewater treatment process with the highest treatment efficiency with T-P reaching 79.77%. However, the values of the main parameters of the treated water were higher than the requirements according to Vietnamese standards and cannot be discharged into water bodies used for domestic purposes. Specifically, the output values of COD and TN were still quite high at 250 mg/L and 62.65 mg/L, respectively, which were 3.5 times and 2 times higher than that of the Vietnamese discharge standard. Therefore, adding post-treatment steps after AnMBR was necessary to ensure that the treated water meets standards and can be reused.

3.2. Performance of post-treatment methods

3.2.1. Performance of PAC

PAC is a chemical widely used in water treatment. When combined with anionic polymers, it will form suspended substances, reducing TSS, turbidity, and COD content in water (Zarei Mahmudabadi *et al.* 2018). As shown in Figure 2, compared to the output value of the AnMBR tank, with a PAC value of 500 mg/L, the COD content has decreased from 250.43 mg/L to 111.492 mg/L, the value TSS decreased from 90 to 46 mg/L. However, as the PAC content continued to increase, the TN, COD, and TSS values also increased. TP value did not change much when increasing PAC content. The reason is that increasing the PAC content too high will slow down the precipitation due to creating a repulsive environment (Nti *et al.* 2021), affecting the reaction process and the quality of the water to be analyzed. Thus, the PAC content of 500 mg/L was suitable for use in further research.

Table 1. Removal efficiency of AnMBR system with seafood processing wastewater

Parameters	Raw seafood processing wastewater	After AnMBR	Vietnam standard
TSS (mg/L)	268	96	50
COD (mg/L)	1100.673	250.43	75
Total nitrogen - TN (mg/L)	228.8	62.65	30
Total phosphorus - TP (mg/L)	39.895	12.115	10

**QCVN 11-MT:2015/BTNMT Vietnamese national technical regulation on the effluent of aquatic Products Processing industry*

Adding anionic polymer will increase the settling efficiency of suspended solids and substances. From Figure 3, it can be seen that the TSS and COD values gradually decreased with increasing anionic polymer content. TN and TP values were slightly affected by polymer content. From the two tests, it can be seen that the optimal dosage of flocculants and coagulants, specifically poly aluminum chloride should be 500 mg/L and anionic polymer with 30 mg/L.

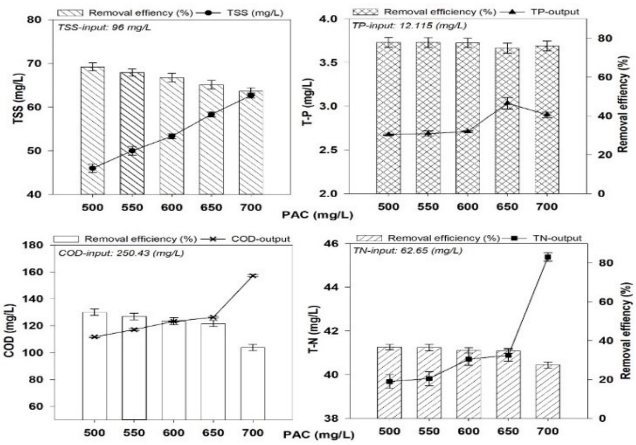


Figure 2. Post-treatment of AnMBR effluent by PAC only.

The experiment dose was 500 mg/L of PAC with 30 mg/L of anionic polymer as flocculant. The most impacted index when chemicals have longer reaction time was TSS as the suspended solids in the liquid settled down with the help of the coagulant and flocculant. On the other hand, the longer reaction time led to an increase in TN and COD, TN was slightly affected, and the COD rose significantly. This happened similarly when increasing the dosage of PAC and anionic polymer. The TP decreased as the chemicals reacted however the index was not significant, which could be shown in Figure 4.

3.2.2. Performance of chlorine

It is noticed that the TSS of the samples gradually decreases when increasing the chlorine content in wastewater from 1 to 6 mg/L. TN and COD values tend to increase with increasing chlorine content. TP reached the lowest value of 10.78 mg/L when the chlorine content was 2 mg/L. Then it gradually increased with increasing chlorine, reaching a value of 11.98 mg/L when the chlorine content was 5 mg/L, as can be seen in Figure 5. Furthermore, the results of correlation analysis between chlorine concentration and wastewater parameters show that there was no significant correlation between chlorine concentration with TSS and TP of water ($p > 0.01$), when there was a correlation between chlorine and COD and TN. This can be explained by the ability of chlorine to oxidize organic and inorganic compounds (ammonia, nitrite, etc.) in water (Aber *et al.* 2011), (Mazhar M. A. *et al.* 2020).

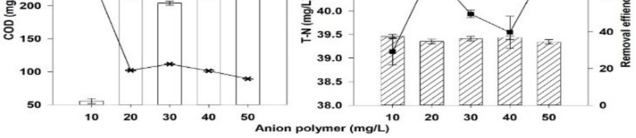


Figure 3. Post-treatment of AnMBR effluent by PAC+ anion polymer.

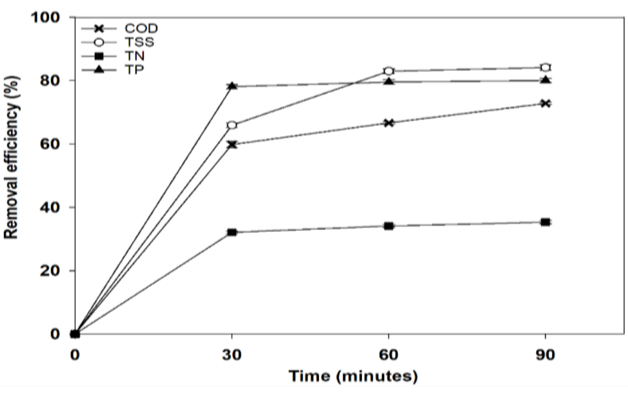


Figure 4. Performance of PAC + anion polymer under different reaction times.

However, the removal efficiency was low and the use of high chlorine concentrations also increases the possibility of forming chlorine-containing by-products, which are highly toxic and can affect human health. Chlorine also can be used in combination with UV as an advanced oxidation method to treat wastewater rich in organic compounds that are difficult to biodegrade. Cost, system complexity, and turbidity of water are factors to consider when using this method (Yeom *et al.* 2021). Thus, it can be seen that chlorine has almost little effect on the effectiveness of the treatment of substances, choosing a treatment chlorine content of 2 mg/L.

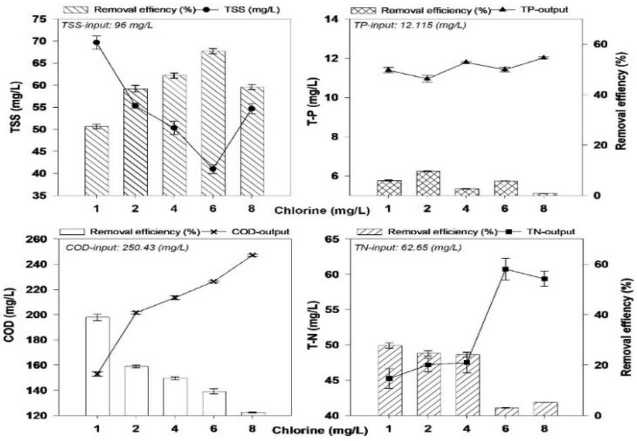


Figure 5. Post-treatment of AnMBR effluent by chlorine.

Time is also an important factor affecting the effectiveness of wastewater treatment. In theory, the longer the processing time, the higher the processing efficiency. With the most optimal dose of 2 mg/L, the effect of a longer time reaction was most visible on TSS as the chlorine reacts with the wastewater, and all the suspended solids in the liquids settle down to the bottom of the tank. There were slight effects on the TN and COD as the longer the reaction time, the better quality the wastewater got as those values decreased lightly. There were no effects on the TP , however, as the longer settling time, the indices fluctuate. The results show that 60 minutes was the most suitable time for the processing process (Figure 6). However, the treatment efficiency was relatively low, and the treated water could not meet the national discharge standards. In fact, chlorine is mainly used to disinfect water before discharging it into the environment (Mazhar *et al.* 2020).

3.2.3. Performance of GAC

Through Figure 7, it can be seen that the COD adsorption and total buffer activity were very good. When increasing the active concentration, more COD and TN were adsorbed. COD and TN contents decreased to 36.75 and 170.49 mg/L at a GAC value of 5000 mg/L. Granular activated carbon has been used routinely to remove organic pollutants from wastewater for a long time (Aber *et al.* 2011). The main mechanism of GAC's pollution treatment is based on the mechanism of adsorption and diffusion of pollutants inside the GAC structure, in which there are two main mechanisms: diffusion of pollutants inside the porous structure of GAC and diffusion of pollutants on the surface of GAC (Ocampo-Pérez *et al.* 2013). The ability of GAC to treat pollutants can vary greatly depending on the molecular size of the pollutant. However, this also gives GAC a very diverse treatment capacity. Studies on many different wastewater subjects show that GAC can treat many types of pollutants including trace organic pollutants or residual organic matter, persistent xenobiotics, soluble organic compounds (DOC), volatile fatty acids (VFAs), some low molecular weight compounds such as nitrogen containing compounds (N-compounds), heavy metals and other difficult-to-decompose organic substances (Almarri *et al.* 2009; Guillossou *et al.* 2020; Nguyen *et al.* 2014). Therefore, GAC was completely capable of effectively treating wastewater with complex components such as seafood processing wastewater, helping to reduce COD and TN in this study.

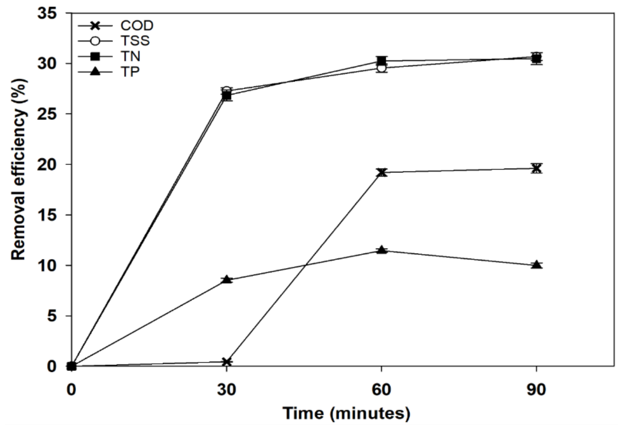


Figure 6. Performance of chlorine under different time reaction

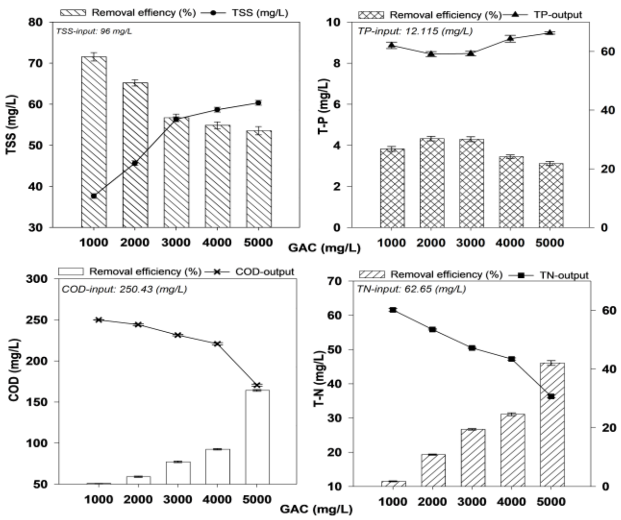


Figure 7. Post-treatment of AnMBR effluent by GAC

The total phosphorus did not change much when changing the GAC content. It can be seen that phosphorus has value independent of GAC content. The output TSS of the AnMBR system was 90 mg/L. The value was reduced to 37 mg/L when the GAC was 1000 mg/L. However, TSS tended to gradually increase with increasing GAC content. It is possible to do so, increasing the GAC content will increase the amount of excess activity in the water leading to an increase in TSS. TSS had a value of 60.3 mg/L at a GAC of 5000 mg/L.

The results of analyzing the significant correlation between parameters also show that GAC concentration was closely related to the TSS, TP, TN and COD performance ($p < 0.01$).

In general, based on the output values of 4 water quality indicators, the appropriate GAC concentration was 5000 mg/L to treat seafood processing wastewater. For better improvement as activated carbon, the post-treatment process should be done differently. The activated carbon should be in a reactor that can separate the suspended carbon so the output water TSS result is not affected. Another option that should be considered is building a filter column with a pressure pump instead of a reactor. The activated carbon inside could be in granular form (Rogers *et al.* 2018).

The optimal dose of GAC used for this experiment is 5000 mg/L. Looking at the results, there was a huge drop in TSS and TP in increasing reaction time for the Activated Carbon reacts and settled down to the bottom of the beaker. As a result, to avoid the huge amount of TSS of the wastewater, better resting time for the liquid can be considered as well and this method can help to reduce the TP. Furthermore, the activated carbon reacted better leading to the reduction in COD and TN but not as significant as the chemical impacts on TSS and TP (Figure 8).

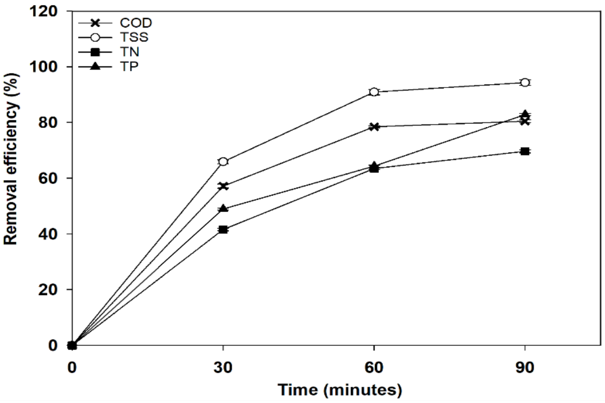


Figure 8. Performance of GAC underdifferent reaction times.

It can be seen in Figure 9 that post-treatment using GAC achieved the highest treatment efficiency among the three substances used. The treatment efficiency of TSS, COD, TP, and TN were 94.04, 79.11; 86.19; and 71.23% respectively. In general, MBRs are known to be very effective in removing hydrophobic and biodegradable contaminants, but may not be effective in removing hydrophilic compounds (Liu *et al.* 2020). This may be the reason why the AnMBR process could completely remove COD or TN and requires a combination of advanced treatment. And the test results with different pretreatment methods all showed the ability to improve the treatment efficiency of the AnMBR process despite the differences in efficiency. While chlorine had the lowest treatment efficiency for all four parameters, PAC and GAC both showed significantly higher treatment efficiency, and GAC achieved the highest treatment efficiency for all four parameters monitored. On the other hand, metal-based coagulants such as PAC are often better at treating hydrophobic or high molecular weight compounds than they are at treating hydrophilic or low molecular weight substances (Park *et al.* 2020). PAC also has some disadvantages such as the potential to increase the concentration of dissolved solids and low efficiency in treating nitrogen compounds. In contrast, adsorption mechanisms unrelated to the hydrophobic properties of the compound such as surface complexation, hydrogen bonding or ion exchange can play an important role in the treatment of trace organic compounds by GAC (Dickenson and Drewes, 2010) and thus help achieve high treatment efficiency even for hydrophilic compounds . GAC also has excellent and diverse treatment ability for nitrogen compounds due to its structure containing oxygen functional groups such as carboxyl, anhydride, lactone, phenol, carbonyl, and quinone and the efficiency increases with higher oxygen concentration in the functional groups (Almarri *et al.* 2009). These characteristics have helped GAC achieve higher TSS, TN and COD treatment efficiency than PAC.

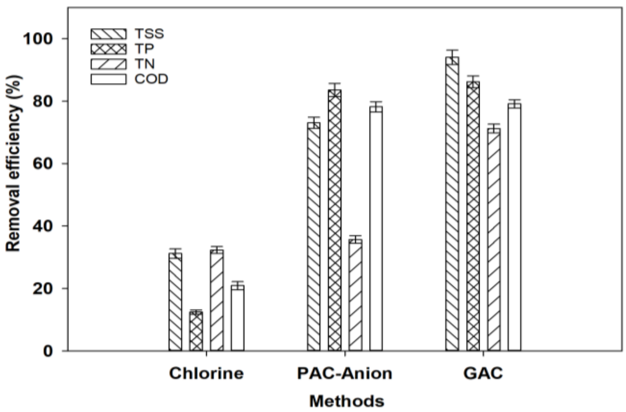


Figure 9. The efficiency of the pilot with laboratory experiments

Furthermore, the values of the above parameters were significantly lower than the standard QCVN 11-MT: 2015/BTNMT. Thus, GAC is completely suitable for use in the post-treatment process to combine with AnMBR in seafood processing wastewater treatment.

4. Conclusion

From the research results, it was found that AnMBR anaerobic membrane technology combined with the post-treatment process has brought high treatment efficiency for seafood processing wastewater. Among the three chemicals tested for post-treatment: chlorine, PAC, and GAC, GAC with a concentration of 5000 mg/L provided the highest treatment efficiency with a treatment time of 60 minutes. Chlorine needs longer treatment time, PAC processes in 30 minutes but the treatment efficiency is lower than GAC. It can be seen that this is a promising new technology with many improvements researched and implemented to suit the needs of environmental conditions in Vietnam. Combining wastewater treatment techniques brings flexibility and adaptability to many different situations, and the technology promises to be widely applied and popularized.

Acknowledgments

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