

1 **Enhancing seafood processing wastewater treatment efficiency of an anaerobic membrane**
2 **bioreactor (AnMBR) system: a comparison of post-treatment methods**

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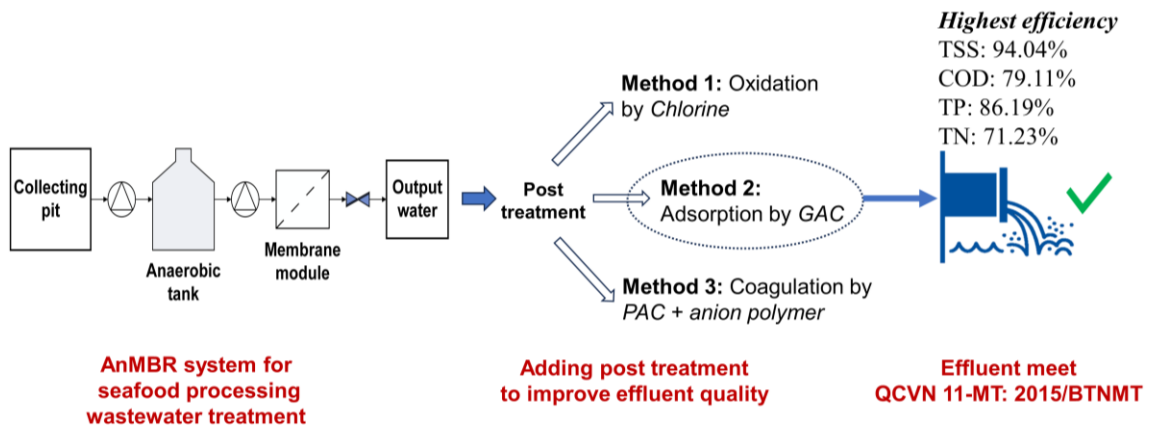
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27 **Graphic abstract**

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ACCEPTED MANUSCRIPT

30 **ABSTRACT**

31 An Anaerobic membrane bioreactor (AnMBR) was applied for treating wastewater of a seafood
32 processing factory in Vietnam. However, COD, TP, and TN values of the AnMBR effluent were
33 250.43 mg/l, 19.9612.115 mg/l, and 62.65 mg/l, respectively, and were about two times higher than
34 the technical regulation for seafood processing wastewater in Vietnam. AnMBR needs to be
35 combined with other post-treatment processes to ensure that treated water can meet the technical
36 regulation. Therefore, current research has tested various advanced post-treatment methods for
37 AnMBR effluent including chlorine, poly aluminum chloride (PAC) combined with anionic polymer,
38 and granular activated carbon (GAC). Different concentrations of the above chemicals and reaction
39 times were tested to select the appropriate post-treatment method. The results showed that chlorine
40 had the lowest treatment efficiency for all four parameters (TSS, COD, TP, and TN). GAC was
41 more effective than PAC in treatment of TN and TSS. Besides, GAC with a concentration of 5000
42 mg/L and a reaction time of 60 minutes brought the highest TSS, COD, TP, and TN treatment
43 efficiency with 94.04, 79.11; 86.19, and 71.23%. This implies that GAC is the suitable method for
44 the post-treatment of AnMBR seafood processing effluent.

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

47 **1. Introduction**

48 Over the past 20 years, the seafood industry of Vietnam has always maintained a growth rate of 10%-
49 20%. However, the seafood processing industry is also one of the industries that cause serious
50 pollution to the environment, especially wastewater. Wastewater generated from seafood processing
51 has high concentrations of COD, BOD₅, suspended solids, total nitrogen, and phosphorus. It can range
52 from 800-3000 mg COD/L, in which the high proportion of biodegradable organic compounds
53 represents the BOD₅/COD ratio ranging from 0.6-0.9 (Ngoc *et al.*, 2022). Therefore, biological
54 treatment methods are often chosen to treat this type of wastewater. Currently, most wastewater
55 treatment systems in seafood processing factories use activated sludge tanks or up-flow anaerobic
56 sludge blanket (UASB) tanks combined with activated sludge tanks to achieve high organics and
57 nutrient removal (Massé *et al.*, 2006).

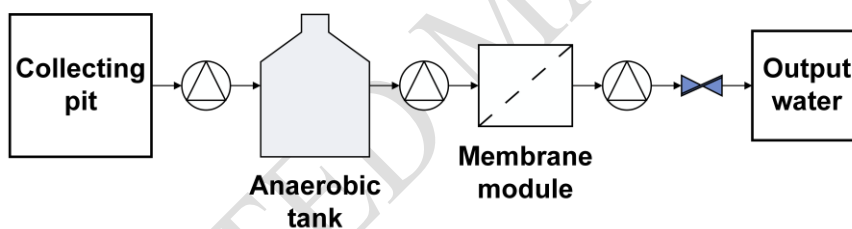
58 In recent years, anaerobic membrane bioreactor (AnMBR) technology has been receiving more and
59 more attention from scientists around the world. AnMBR has several outstanding advantages such
60 as low sludge production capacity, lower energy demand compared to aerobic processes, and the
61 ability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

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

72 **2. Materials and methods**

73 2.1. Configuration of the pilot-scale anaerobic membrane bioreactor (AnMBR) system

74 The AnMBR system was installed and operated at a seafood processing factory, in Ba Ria Vung Tau,
75 Vietnam. AnMBR system with a capacity of 0.5 m³/day, dimensions Length × Width × Height = 600
76 × 600 × 1500 (mm) is made of stainless steel. Here, anaerobic microorganisms will conveniently
77 consume organic substances in the water. Next, the water is pumped to the MBR tank to continue
78 removing chemicals, solids as well as disease-causing bacteria. The membrane used in the AnMBR
79 was from Microdyn-Nadir, Germany with a molecule weight of 150,000 Da, flux LMH (L/m²/h)/bar
80 of 153.06/85.03 and a rejection size of 5K-Dextran and 5000 Da pore size, HRT was 10 h. To
81 investigate different post-treatment techniques, effluent after the AnMBR system was collected and
82 stored at 4°C, then followed a tank that contains a 3-phase 240VAC induction motor with a maximum
83 frequency of 50Hz and a maximum speed of 1400 revolutions per minute (RPM). A transmission
84 with a 20:1 gear ratio was attached to the engine. Chemical coagulants were added alternately to the
85 tank, first mixed well poly aluminum chloride, then Anionic Polymer.



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88
89 **Figure 1.** Anaerobic membrane bioreactor (AnMBR) system for seafood processing wastewater
90 treatment

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

99 2.2 Evaluating post-treatments methods

100 2.2.1 Post-treatment with PAC

101 The post-treatment was carried out with jar testing to find out the effect of the post-treatment
102 chemicals on the output water from the process. In this experiment, the most efficient dose of the
103 different chemicals, which are Chlorine, Poly Aluminium Chloride with Anionic Polymer as
104 flocculant would be tested. The jar tester used in the experiment was from Aqualytic – Germany with
105 6 positions for stirring, time, and speed (RPM) adjustable and turning off automatically. The chemical
106 used in the experiment was poly aluminum chloride (PAC) - $[Al_2(OH)_nCl_{6-n}]_m$ as a coagulant and
107 anionic polymer $CONH_2[CH_2-CH-]_n$. Firstly, the coagulant was added and stirred at 30 RPM for 15
108 minutes then rest for 5 minutes as the chemical reaction and sedimentation with the respective dosage
109 of 500 mg/L, 550 mg/L, 600 mg/L, 650 mg/L, and 700 mg/L. After stirring with the coagulant, 30
110 mg/L of anionic polymer was added to each jar as flocculation. The motor would run at 30 RPM for
111 15 minutes and then rest. After that, the output wastewater was analyzed to choose the optimal dosage
112 for the coagulant that would be used for finding the optimal Anionic polymer dosage. There were 5
113 different anionic polymer dosages from 10 mg/L to 50 mg/L, which was: 10 mg/L, 20 mg/L, 30 mg/L,
114 40 mg/L, and 50 mg/L.

115 2.2.2 Post-treatment with Chlorine

116 The chlorine used in the experiment was Chlorine 70%, there were 5 different dosages 1 mg/L, 2
117 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
118 70 RPM for 20 minutes for mixing the chemical.

119 2.2.3 Post-treatment with GAC

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

124 3. Results and Discussion

125 *3.1. Seafood processing wastewater treatment efficiency of AnMBR system*

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

134 **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

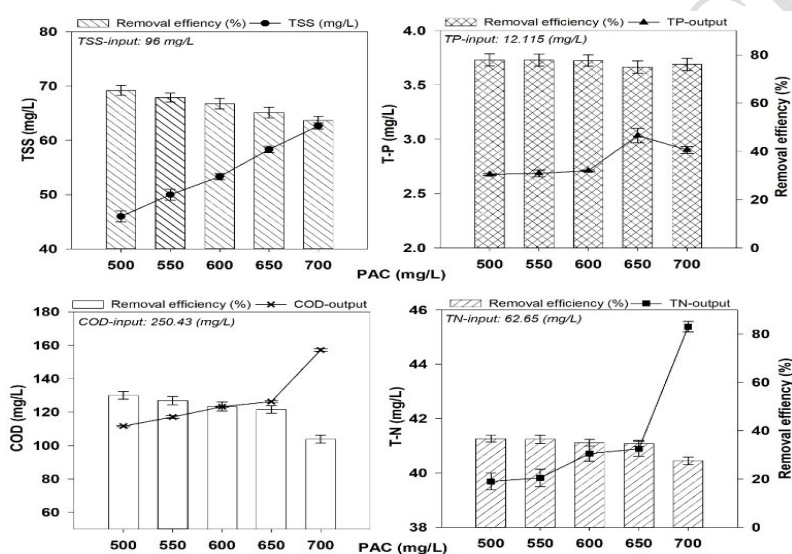
135 **QCVN 11-MT:2015/BTNMT Vietnamese national technical regulation on the effluent of aquatic*
136 *Products Processing industry*

137 *3.2 Performance of post-treatment methods*

138 *3.2.1 Performance of PAC*

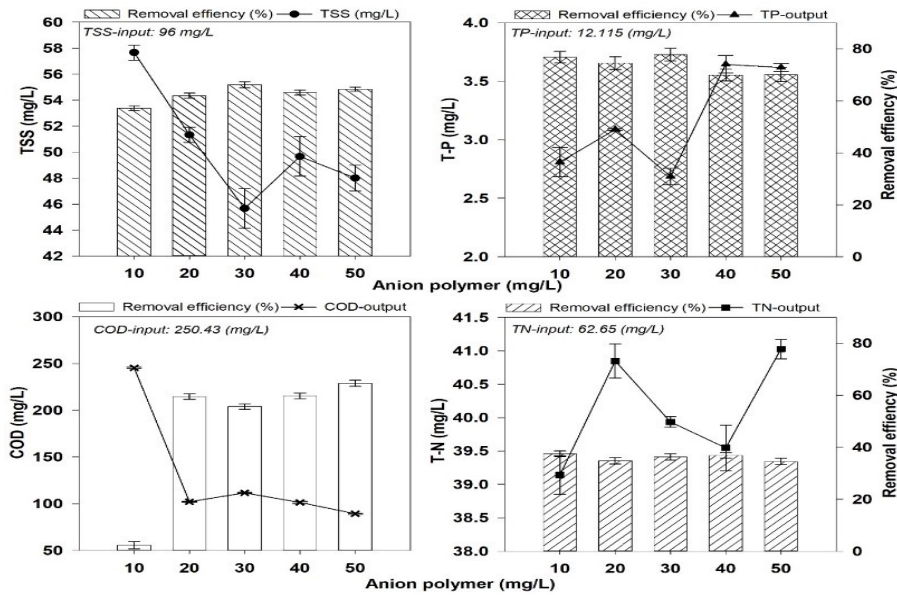
139 PAC is a chemical widely used in water treatment. When combined with anionic polymers, it will
140 form suspended substances, reducing TSS, turbidity, and COD content in water (Zarei Mahmudabadi
141 *et al.*, 2018). As shown in Figure 2, compared to the output value of the AnMBR tank, with a PAC
142 value of 500 mg/L, the COD content has decreased from 250.43 mg/L to 111.492 mg/L, the value
143 TSS decreased from 90 to 46 mg/L. However, as the PAC content continued to increase, the TN,
144 COD, and TSS values also increased. TP value did not change much when increasing PAC content.

145 The reason is that increasing the PAC content too high will slow down the precipitation due to
 146 creating a repulsive environment (Nti *et al.*, 2021), affecting the reaction process and the quality of
 147 the water to be analyzed. Thus, the PAC content of 500 mg/L was suitable for use in further research.
 148 Adding anionic polymer will increase the settling efficiency of suspended solids and substances.
 149 From Figure 3, it can be seen that the TSS and COD values gradually decreased with increasing
 150 anionic polymer content. TN and TP values were slightly affected by polymer content. From the two
 151 tests, it can be seen that the optimal dosage of flocculants and coagulants, specifically poly aluminum
 152 chloride should be 500 mg/L and anionic polymer with 30 mg/L.

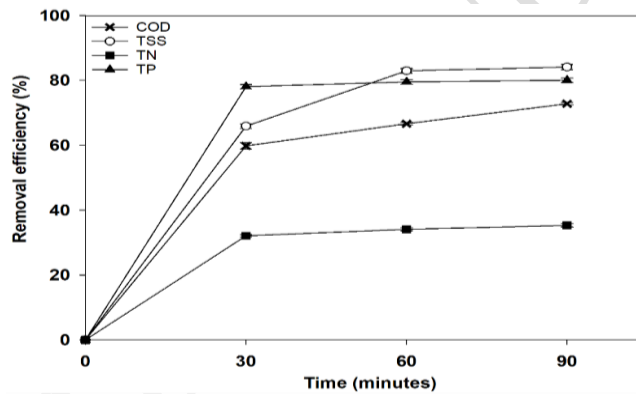


153
 154 **Figure 2.** Post-treatment of AnMBR effluent by PAC only.

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



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163
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Figure 3. Post-treatment of AnMBR effluent by PAC+ anion polymer.

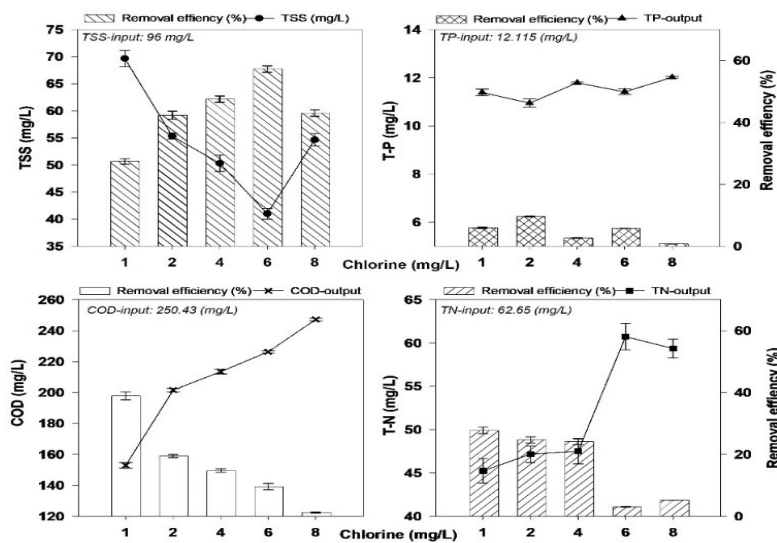


165
166
Figure 4. Performance of PAC + anion polymer under different reaction times.

167 *3.2.2. Performance of Chlorine*

168 It is noticed that the TSS of the samples gradually decreases when increasing the chlorine content in
 169 wastewater from 1 to 6 mg/L. TN and COD values tend to increase with increasing chlorine content.
 170 TP reached the lowest value of 10.78 mg/L when the chlorine content was 2 mg/L. Then it gradually
 171 increased with increasing chlorine, reaching a value of 11.98 mg/L when the chlorine content was 5
 172 mg/L, as can be seen in Figure 5. Furthermore, the results of correlation analysis between chlorine
 173 concentration and wastewater parameters show that there was no significant correlation between
 174 chlorine concentration with TSS and TP of water ($p > 0.01$), when there was a correlation between

175 chlorine and COD and TN. This can be explained by the ability of chlorine to oxidize organic and
176 inorganic compounds (ammonia, nitrite, etc.) in water (Aber *et al.* 2011), (Mazhar M. A. *et al.* 2020).

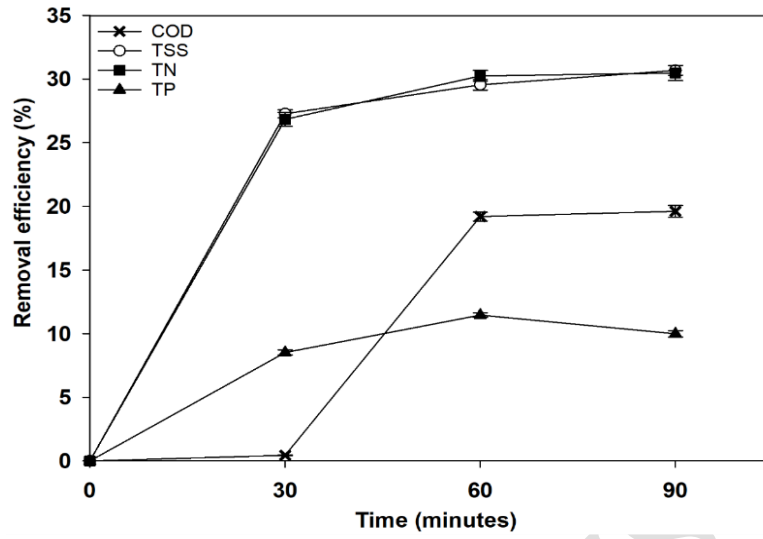


177
178 **Figure 5.** Post-treatment of AnMBR effluent by chlorine.

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

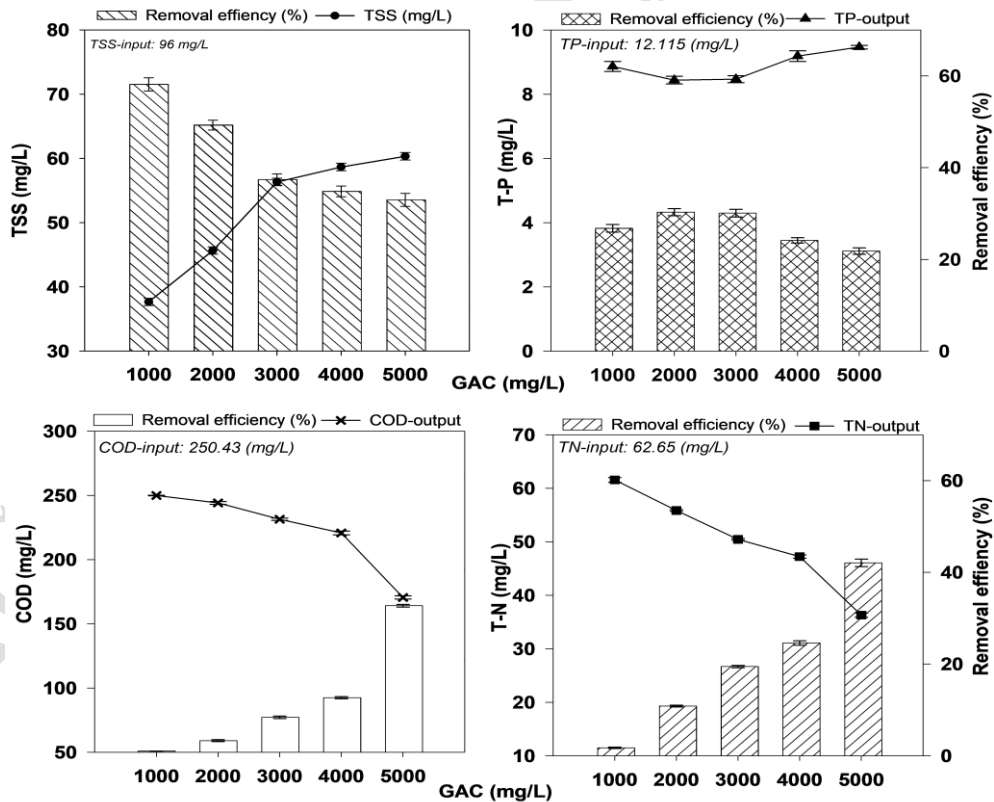
186 Time is also an important factor affecting the effectiveness of wastewater treatment. In theory, the
187 longer the processing time, the higher the processing efficiency. With the most optimal dose of 2
188 mg/L, the effect of a longer time reaction was most visible on TSS as the chlorine reacts with the
189 wastewater, and all the suspended solids in the liquids settle down to the bottom of the tank. There
190 were slight effects on the TN and COD as the longer the reaction time, the better quality the
191 wastewater got as those values decreased lightly. There were no effects on the TP, however, as the
192 longer settling time, the indices fluctuate. The results show that 60 minutes was the most suitable
193 time for the processing process (Figure 6). However, the treatment efficiency was relatively low, and

194 the treated water could not meet the national discharge standards. In fact, chlorine is mainly used to
 195 disinfect water before discharging it into the environment (Mazhar *et al.*, 2020).



196
 197 **Figure 6.** Performance of chlorine under different time reaction

198 **3.2.3 Performance of GAC**



199
 200 **Figure 7.** Post-treatment of AnMBR effluent by GAC

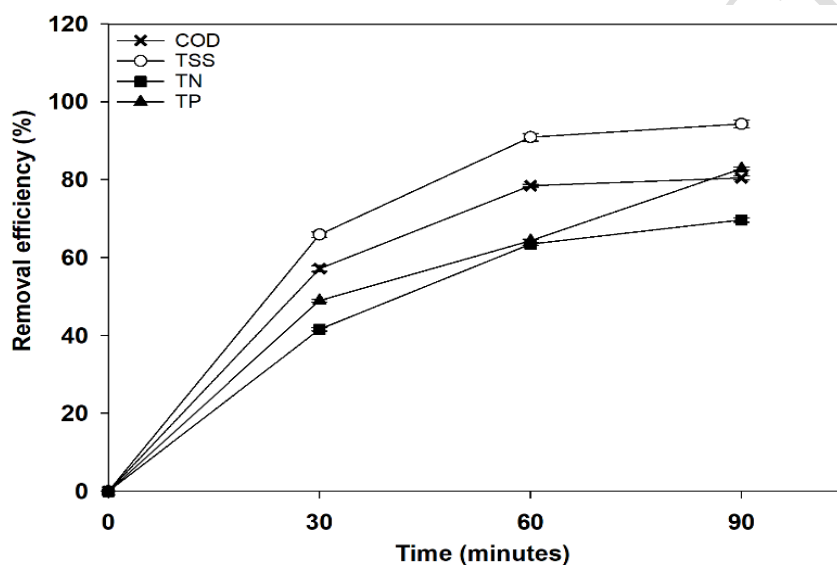
201 Through Figure 7, it can be seen that the COD adsorption and total buffer activity were very good.
 202 When increasing the active concentration, more COD and TN were adsorbed. COD and TN contents

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

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

223 The results of analyzing the significant correlation between parameters also show that GAC
224 concentration was closely related to the TSS, TP, TN and COD performance ($p < 0.01$). In general,
225 based on the output values of 4 water quality indicators, the appropriate GAC concentration was 5000
226 mg/L to treat seafood processing wastewater. For better improvement as activated carbon, the post-
227 treatment process should be done differently. The activated carbon should be in a reactor that can
228 separate the suspended carbon so the output water TSS result is not affected. Another option that

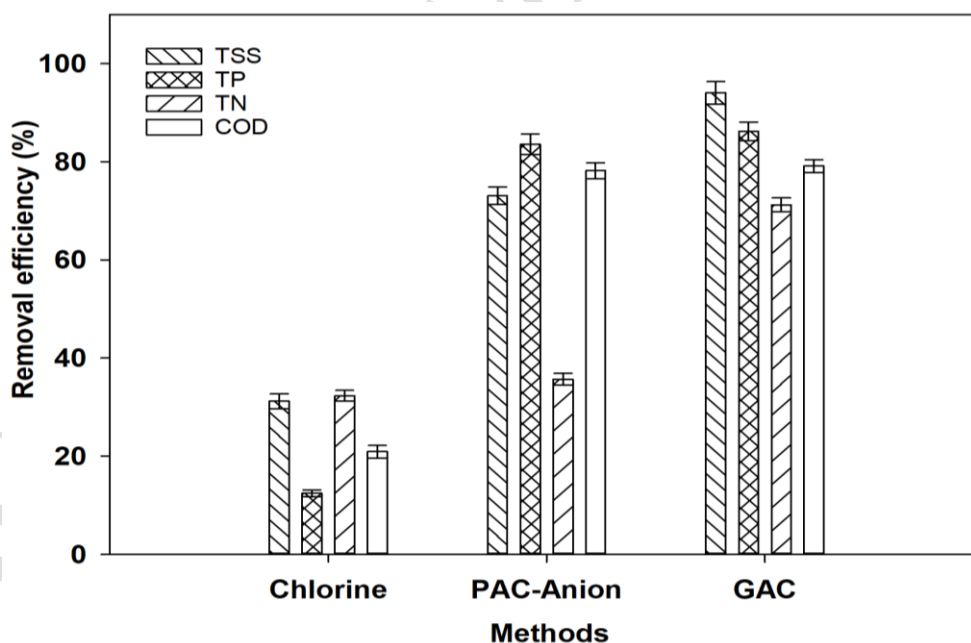
229 should be considered is building a filter column with a pressure pump instead of a reactor. The
230 activated carbon inside could be in granular form (Rogers et al., 2018).
231 The optimal dose of GAC used for this experiment is 5000 mg/L. Looking at the results, there was a
232 huge drop in TSS and TP in increasing reaction time for the Activated Carbon reacts and settled down
233 to the bottom of the beaker. As a result, to avoid the huge amount of TSS of the wastewater, better
234 resting time for the liquid can be considered as well and this method can help to reduce the TP.
235 Furthermore, the activated carbon reacted better leading to the reduction in COD and TN but not as
236 significant as the chemical impacts on TSS and TP.



237
238 **Figure 8.** Performance of GAC under different reaction times.

239 It can be seen in Figure 9 that post-treatment using GAC achieved the highest treatment efficiency
240 among the three substances used. The treatment efficiency of TSS, COD, TP, and TN were 94.04,
241 79.11; 86.19; and 71.23% respectively. In general, MBRs are known to be very effective in removing
242 hydrophobic and biodegradable contaminants, but may not be effective in removing hydrophilic
243 compounds (Liu *et al.*, 2020). This may be the reason why the AnMBR process could completely
244 remove COD or TN and requires a combination of advanced treatment. And the test results with
245 different pretreatment methods all showed the ability to improve the treatment efficiency of the
246 AnMBR process despite the differences in efficiency. While chlorine had the lowest treatment
247 efficiency for all four parameters, PAC and GAC both showed significantly higher treatment

248 efficiency, and GAC achieved the highest treatment efficiency for all four parameters monitored. On
 249 the other hand, metal-based coagulants such as PAC are often better at treating hydrophobic or high
 250 molecular weight compounds than they are at treating hydrophilic or low molecular weight substances
 251 (Park *et al.*, 2020). PAC also has some disadvantages such as the potential to increase the
 252 concentration of dissolved solids and low efficiency in treating nitrogen compounds. In contrast,
 253 adsorption mechanisms unrelated to the hydrophobic properties of the compound such as surface
 254 complexation, hydrogen bonding or ion exchange can play an important role in the treatment of trace
 255 organic compounds by GAC (Dickenson and Drewes, 2010) and thus help achieve high treatment
 256 efficiency even for hydrophilic compounds . GAC also has excellent and diverse treatment ability for
 257 N compounds due to its structure containing oxygen functional groups such as carboxyl, anhydride,
 258 lactone, phenol, carbonyl, and quinone and the efficiency increases with higher oxygen concentration
 259 in the functional groups (Almarri *et al.*, 2009). These characteristics have helped GAC achieve higher
 260 TSS, TN and COD treatment efficiency than PAC.



261
 262 **Figure 9.** The efficiency of the pilot with laboratory experiments

263 Furthermore, the values of the above parameters were significantly lower than the standard QCVN
 264 11-MT: 2015/BTNMT. Thus, GAC is completely suitable for use in the post-treatment process to
 265 combine with AnMBR in seafood processing wastewater treatment.

266 4. Conclusion

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

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