Wastewater of Seaweed industry contains macronutrients, alkalis, moderately biodegradable organics, and has a large volume, which it is treated using a biological process (activated sludge) followed by an adsorption process. The results of this treatment are quite effective in reducing the concentration of pollutants to reach the quality standard. However, the reuse of seaweed industrial wastewater into drinking water or process water is still not effective when using a combination of these processes.

On the other hand, biological treatment such as aerobic fixed film biofilter (AF2B) and nano adsorption has high effectiveness in reducing biodegradable organic matter (BOD, COD), bacteria, and other metals. Therefore, the reuse of seaweed industrial wastewater into drinking water can be done by combining the two types of processes. The aim of the research was to study the ability of the AF2B – Granular adsorption – nano adsorption process in reducing pollutants in seaweed industrial wastewater. The experiment is conducted by flowing wastewater continuously into a series of process an AF2B reactor, GAC, and CNTs column. By making changes to the factors that affect the performance of each process, and then measuring the pollutant concentration at the input and output of each process, so the performance of each process can be known. Data is analysed to obtain optimum condition in each process that used for design process in wastewater treatment plant. The results showed that of the AF2B/GAC process was able to reduce contaminants such as TSS, BOD, COD, NH3N and Chlorine by 98 %, 99 %, 97.3 %, 97.8 %, and 100 % respectively. Furthermore, in the CNTs process, all pollutants not detected until they meet drinking water quality standards

Keywords: aerobic biofilter, granular adsorption, nano adsorption, seaweed, performance, pollutants, wastewater

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AN ANALYSIS OF PERFORMANCE OF COMBINED AEROBIC BIOFILTER – GRANULAR ADSORPTION – NANO ADSORPTION PROCESS IN SEAWEED INDUSTRIAL WASTEWATER TREATMENT

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1. Introduction

The seaweed processing industry is one of the food industries, where in the seaweed processing industry into ATCC (alkaline treated cotton chips) products, which wastewater are generated from the washing, soaking, and cooking processes. The average volume of wastewater generated from this industry is large (300–500 $m^3 \cdot d^{-1}$) with characteristics: pH (11–13), BOD (300–500 $mg \cdot L^{-1}$), COD (700–1,300 $mg \cdot L^{-1}$), TSS (150–300 $mg \cdot L^{-1}$), Ammonia, NH_3N (10–20 $mg \cdot L^{-1}$), and Chlorine (5–15 $mg \cdot L^{-1}$). For this reason, the operational costs of the wastewater treatment plant (WWTP) are high operating costs. In addition, the large volume of wastewater (200–300 $M^3 \cdot$day) and the low amount of biodegradable organic matter have the potential to be reused as process or drinking water. Several studies have shown that wastewater from the seaweed processing industry is still processed on a laboratory scale with the target of achieving quality standards and there has been no research in pilot plant scale that aims to utilize wastewater from the seaweed processing industry for process water or drinking water. Several types of treatment have been conducted, including: a combination of activated sludge and stabilization processes, up-flow anaerobic sludge blanked (UASB) [1, 2], moving bed bioreactor (MBBRs) that combined with ozonation [3], Integrated Fixed Film Activated Sludge [4], Ozonation and granular activated carbon filtration adsorption process [5]. Meanwhile, the combination of processes that have been developed to obtain process water and drinking water are biofilter-ultrafiltration, biofilter-nano adsorption, and biofilter-ozonisation. The combinations of processes had developed in the last few decades, including combination of biofilter-ultrafiltration [6, 7], biofilter-nano adsorption and biofilter-ozonation [8]. However, the combination of the aerobic biofilter-granular adsorption-nano adsorption process in seaweed wastewater treatment to drinking water or process water has not been studied, especially those that operate continuously and on a pilot plan scale. For this reason, a combination of processes is needed to increase the effectiveness of the treatment so that the treated water can be reused as process water or drinking water.

2. Literature review and problem statement

The aerated fixed film biofilter (AF2B) reactor can reduce pollutants in wastewater hospital such as BOD, ammonia, COD, and phenol where one of the factors that affect the reactor's ability is the starter volume and air flowrate [9]. The aerated fixed film biofilter (AF2B) reactor that operates in batches can reduce pollutants in hospital wastewater such as BOD, ammonia by 92 % and 76 %, respectively [10]. Meanwhile, in the same experiment using an AF2B reactor and a bacterial consortium that operates in batch, it can reduce COD and phenol pollutants by 86 % and 88 %, respectively [11]. However, for the AF2B reactor which treats seaweed industry wastewater and operates continuously, no study has yet been carried out. Meanwhile, reactors like AF2B such as the MBBR reactor have not yet found a study on their performance. The combination of the PAC-MBR process was better than MBBR-GAC in reducing the concentration of pollutants, especially phototrophs and diclofenac contained in pharmaceutical wastewater [12]. While the MBBR system is more efficient than the integrated fixed film activated carbon system (IFAS) in taking BOD and TSS, where the MBBR system can take BOD of 98.1–99.4 % [13]. The combination of the biological process-ozonisation also has a good ability to reduce the concentration of pollutants in hospital wastewater and resin industry, especially pathogenic compounds, which organic carbon removals from 55 to 100 % [14] and organic nitrogen removals from 41 to 77 % were obtained after biological treatment [15]. The combination of biological processes and ozonation has good ability to reduce pollutants, where organic carbon removals from 55 to 100 % and organic nitrogen removals from 41 to 77 %, respectively. The effect of the contact time was studied at a constant ozone dose of 13.0+/1.2 $mg{\cdot}L^{-1}{\cdot}min^{-1}$ and contact times ranging from 30 to 180 min [14]. While AF2B/O3 reactor could reduce BOD5, faecal coli, phenol and Pb were 97.92 %, 99.23 %, 100 %, 100 %, respectively [15]. The combination of ozonation-biological activated carbon (BAC) filtration was effective in reducing trace organic chemicals (Tr–O–Cs) in wastewater. While the combination of ultrafiltration and adsorption processes can reduce POME content by more than 90 % for all parameters compared to decantation which only reduces 40–80 % [16]. Meanwhile, the combination of ultrafiltration and adsorption processes combined with membrane nanofiltration (NF) can take all pollutants [17]. Thus, the ultrafiltration or ozonation process combined with biological processes can increase the effectiveness of wastewater treatment in reducing pollutants. However, previous studies have not shown a combination of biological-adsorption or ultrafiltration-ozonisation processes to treat seaweed processing industrial wastewater, where the wastewater has distinctive characteristics. Besides that, combining it with the use of nano adsorption technology is also still not found.

On the other hand, in recent decades, nanoparticle technology had developed in wastewater treatment, both in forms and types of use [18]. One type of nano particles that used activated carbon nano in the form of tubes or membranes [19]. This is because activated carbon nano has a large surface, good volumetric potential, microporous structure, extra shelf-life time, less mechanical stress, high adsorption capacity, high surface reactivity, no secondary pollution, strong chelating capabilities and they are easy to recover and reuse [20]. The most extensively studied nanomaterials, zero-valent metal nanoparticles (Ag, Fe, and Zn), metal oxide nanoparticles (TiO_2, ZnO, and iron oxides), and carbon nanotubes (CNTs), and nanocomposites were highlighted [21]. The nanoparticles can be used as effective adsorbents for removing various heavy metals from contaminated water. The ease of synthesis, economic feasibility, and easy surface modifications is a few vital features that have led to the development of this novel technology. So far, lower concentrations of heavy metals have been removed with high selectivity and adsorption capacity. Work must be done on scaling up the laboratory-scale experiments to a larger scale, improving the biocompatibility of nanoparticles, making it eco-friendly, and making the entire process cost-effective [21]. However, an in-depth study of the ability of nano adsorption in treating seaweed industrial wastewater has not yet been found so that it can provide an opportunity to conduct a study on the use of nano adsorption in seaweed industrial wastewater.

Previous studies have shown that nanomaterials are feasible for use in removal metals such as cadmium, lead, and chromium dissolved in water, especially in the future requires a water treatment process that has high capabilities and is cost efficient [22]. Future research works on developing a cost-effective way of nanocomposite production and toxicity testing of nanomaterials in wastewater applications are recommended. Further studies on the efficiency of the nano adsorbents on a pilot or industrial scale are highly needed to test the practicality of the nano adsorbents for selected heavy metals removal from real wastewater [23]. Due to lower filtration pressures the hybrid processes feature lower energy consumption and produce fewer problematic concentrates consisting of organics and multivalent ions which can precipitated. They also feature advantages in indirect potable reuse applications such as managed aquifer recharge since the salt content of the product water is closer to natural conditions. The optimum combination of nanofiltration and activated carbon depends on local boundary conditions such as size of plant, raw water characteristics and plant location. In the light of growing water scarcity and increasing concerns about organic micropollutants activated carbon treatment in combination with nanofiltration has the potential to applied in an increasing number of cases [24]. The applications of column nano tubes (CNTs) with based composite membranes in water treatment is comprehensively review, including seawater or brine desalination, oil-water separation, removal of heavy metal ions and emerging pollutants as well as membrane separation coupled with assistant techniques [25]. Thus, these problems in study are how to find the ability of combination aerobic fixed film biofilter-granular adsorption-nano adsorption process in treating wastewater from the seaweed processing industry. So, the result of this study is expected as a reference in implementation or practice in the field.

3. The aim and objectives of the study

The aim of study is to analysis the ability of combination aerobic fixed film biofilter-granular adsorption-nano adsorption process in treating seaweed wastewater to water with drinking water standard.

To achieve the aim so the objective of the research is:

– to measure pollutant removal in aerobic biofilter processes by AF2B;

– to measure pollutant removal in the granular adsorption process by GAC;

– to measure pollutant removal in nano adsorption processes by CNTs.

4. Materials and methods

4. 1. The studied materials and equipment used in the experiment

The wastewater used in this study is wastewater discharged from the washing and soaking process in the seaweed processing industry, where the wastewater before being fed into the AF2B reactor is pre-treated in the neutralization-coagulation-flocculation process with the addition of H_2SO_4 98 % (v/v), PAC 50 % (w/v), and polymer so that the feed has a pH: 6.5–7.0, TSS: 250 mg·L^{-1}, BOD: 380 mg·L^{-1}, COD: 480 mg·L^{-1}, NH3-N: 28 mg·L^{-1}, and Chlorine: 12.5 mg·L^{-1}. Furthermore, wastewater is fed into the influent tank of 2,000 L to be flowed into the treatment system as feed in the combination process AF2B-GAC-CNTs which operates continuously as Fig. 1.

The influent tank was made from fiberglass material with a diameter of 83 cm, and height of 111 cm and working volume of 1,500 L. The influent tank is equipped with a ball valve that served to regulate the flow rate entering the AF2B reactor.

AF2B. AF2B reactor is a biological reactor that made a fiberglass material with dimensions as shown in Table 1. The AF2B reactor is divided into 3 equal parts, where each part contains biofilter that is made of plastic with a bee nest shape. The material of the biofilter is plastic with a density of 0.125 g·cm^{-3}, volume of 5,160 cm^3 and specific surface area of 150–240 m^2·m^{-3}. pH controller is fitted on the inlet of the AF2B while the pump recycle is fitted on the sedimentation section to recycle a part of the sludge into each biofilter. The diffuser is placed at the bottom of the biofilter connected with a blower to supply the air for the perfection of the mixing process and the growth of microorganisms. Circulation flow in the reactor is maintained by making baffles in each biofilter chamber.

The research preparation, especially the preparation of the AF2B reactor which there are 3 stages of preparation: seeding stage, acclimatization stage and implementation stage. Seeding stage was conducted to grow aerobic microorganisms. In this study, seeding was conducted in AF2B reactor using wastewater from the seaweed processing industry and added bacterial consortium culture (BCC) was mixed culture of several types of bacteria such as *Nitrobacter sp, Nitrosomonas sp, Bacillus sp1, and Bacillus sp2*, where the bacteria are obtained from the selection and isolation of potential bacteria in seaweed wastewater. The bacterial require certain nutrients such as carbon (C), nitrogen (N) and phosphorus (P) for growth. In addition to carbon, trace amounts of glucose, urea nitrogen and phosphate (SP-36) were given after every two days. The C:N:P nutrient ratio of approximately 350:7:1 was maintained. The seeding stage formed a biofilm covering the top layer of biofilter, which caused a strong odor and showed sizable and stable COD reduction. To maintain aerobic conditions in the reactor and maximum bacterial growth, the air supply from the blower through the diffuser is carried out continuously at an adjusted flowrate. In the acclimatization process, the result of seeding (starter) is added with wastewater (substrate) every two days followed by the addition of 10 % by volume of nutrients until the total volume reaches 500 L. In this process the air supply for aeration is maintained. Seeding stopped after the COD reduction was stable and had reached high efficiency. Generally, organic compounds in the wastewater can be decomposed by microorganisms. The decomposition process can be faster and more effective in breaking down pollutants present in wastewater along with the growth of microorganisms. Likewise, the acclimatization process also shows an increasing microbial growth which is indicated by the thickening of the biofilm on the surface of the biofilter in the form of a honeycomb. The acclimatization process was stopped, and the running process was started when the COD concentration decreased, pH: 6.0–7.5, and the temperature was stable at 29–31 °C [26]. Next follow the implementation stage.

GAC. GAC is an adsorption column that made of PVC (Poly Vinyl Chloride) with dimensions as shown in Table 1, where the adsorption column contains granular activated carbon (GAC) weighing 5,000 g. GAC supplied from Aquametric (Bc-830 a virgin activated carbon, granular form, 0.7 to 1.0 mm). One PVC column was used as adsorption units with a dimension of 60 cm (height)×7.62 cm (inside diameter). The column was filled with adsorbent material to a height of 30 cm. Perforated Plexiglas disks (hole diameter: 1.0–1.5 mm) were placed at the inlet and outlet of the columns and allowed uniform distribution (shower) of the influent into the adsorbent bed. These perforated disks were horizontally installed and allowed the delimiting of the inlet and outlet zones.

Nano Adsorption (NA). Nano adsorption used in this experiment is Column Nano Tube (CNTs) which CNTs are nano-adsorbent membranes in the form of tubes which are arranged horizontally. The material of membrane has hydrophilic charge which can promote run times between cleaning and reduce clogging rates. An experimental unit of the pilot type was used. It was a tangential filtration unit operating in batch mode with permeate recirculation in the feed tank. The experimental unit comprised a recirculation pump hydro-cell (with a speed regulator) capable of delivering a flow rate of a maximum hydrostatic pressure 606.2 kPa. The system CNTs provided a manometer and a concentrate outlet needle valve for securing the transmembrane pressure. Table 1 shows the characteristics of AF2B, GAC, and CNTs system [27].

Table 1

Characteristics of used in this research

Components	AF2B	GAC	CNTs
Dimension:			
– Length (cm)	60	–	50
– Width (cm)	30	–	–
– Height (cm)	30	60	–
– Diameter (cm)	–	76.2	10
– Effective volume (L)	50	–	–
Biofilter, Activated carbon			
– Surface area (m^2·m^{-3})	150–240	–	–
– Volume (cm^2)	5,160	–	–
– Size (mesh; nm)	–	200–100	100–1,000
– Type	Bee nest	Granular	Membrane
Operation Pressure (kPa)	100.1	100.1–151.7	606.2–1,200
Material	Acrylic	Acrylic	Carbon membrane
Amount	1	1	3

Table 1 shows that the equipment used for this experiment is on a pilot plan scale, where the volume of the AF2B reactor is 50 L, containing a biofilter shaped bee nest with a surface area of 150–240 $m^2 \cdot m^{-3}$. So that microorganisms have a greater opportunity to degrade pollutants. On the other hand, the very high operating pressure on the CNTs column at 606.2–1,200 can cause the adsorption process on the nano adsorbent to run well.

4. 2. Operational technique

As showed in Fig. 1, which the wastewater that has been pre-treated as much as 2,000 L is stored in the feed tank, then flowed into the AF2B reactor using a peristaltic pump (Type: Watson Marlow 050.7131.10U) at a rate of 0.167 $L \cdot min^{-1}$ (HRT: 5 hours). The flowrate is regulated by the valve opening and is read in a rotameter flowmeter 0.5" (Type: DFG-10, DARHOR). Wastewater is flowed into the AF2B reactor continuously so that the wastewater is in contact with the biofilter and forms a circulating flow. The end process of the AF2B of 10 % of flowrate was recycled. The sampling was taken from the effluent of the AF2B at every 15 minutes until steady state to determine the performance of the AF2B reactor. Furthermore, the effluent of the AF2B reactor is flowed to the top of the GAC column so that the process of adsorption of pollutants occurs by granular activated carbon (adsorbent). The sampling was taken from the effluent of the GAC at every 5 minute to determine the performance of the GAC column. The effluent of the GAC column is flowed to NA column (CNTs) using jet pump (Type: SurgefloDP-160) which pressure operations on 606.2 kPa–1,200 kPa so that pressure difference between input-output CNTs of 515 kPa. The sampling was taken from the effluent of the CNTs (permeate) at every time to determine the performance of the CNTs column, while the concentrate is recycled to the GAC column.

The AF2B-GAC-NA system is a pilot plant scale system because the volume of wastewater treated is quite large, namely 2,000 L, so that the operating conditions and data obtained from this experiment can be used as a reference for the real operating conditions in the field. For example, the operating pressure on a column of CNTs is influenced by the size of the nano membrane and the number of columns of CNTs, where the smaller the size of the nano membrane and the more columns of CNTs, the greater the input operating pressure in the column of CNTs.

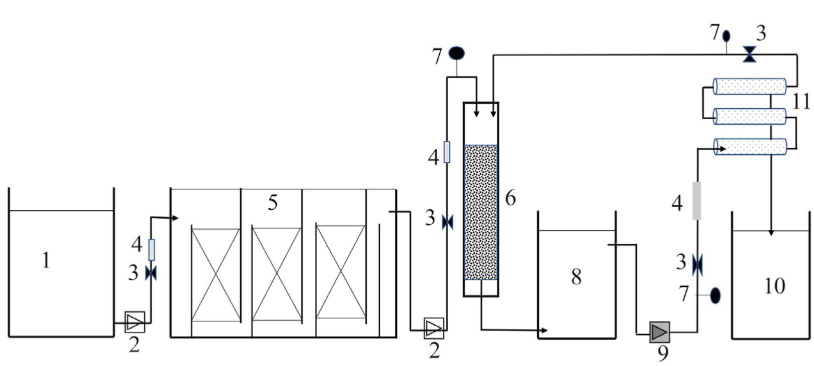

Fig. 1. The process flow chart of AF2B-GAC-NA system: 1 – Feed tank; 2 – Pump; 3 – Valve; 4 – Water Manometer; 5 – AF2B Reactor; 6 – GAC Column; 7 – Pressure Indicator; 8 – Storage Tank; 9 – Jet pump; 10 – Water tank (permeate tank); 11 – CNTs

4. 3. Technique for determining of the properties of sample

The samples were analyzed by using APHA standard procedures to investigate the pH, BOD, COD, TSS, and TDS. The pH was measured using a Fisher Scientific type pH meter (Model XCL25) previously calibrated with pH 6, 7, 8 standard solutions. BOD was measured using modified Winkler's method, where the dissolved oxygen concentration was measured before and after incubation in a Winkler bottle for 5 days at 20 °C. Then the sample was titrated using $Na_2S_2O_5$ and corrected with a blank solution. To determine the concentration of COD, samples were immediately stored at 4 °C and then analyzed using a UV-type spectrophotometer 0811 M136 (Varian brand, Canada Inc) according to the APHA standard method, with a calibration curve including standards of 10, 100, 400, 700 and 1000 $mg \cdot L^{-1}$.

The determination of total suspended solids (TSS), according to the analytical method (MA115-S.S. 1.2), was carried out by filtering a sample portion (normally 100 ml) through a previously dried and weighed Whatman 934 AH glass microfiber filter (pore size1.5 µm) under vacuum. When the filtration was completed, the residue was dried at 103–105 °C overnight or at least 3 hours. The weight of suspended solids is obtained by calculating the difference of the filter weights before and after drying. To determine pH and TDS using an electrical conductivity method in the form of measuring instrument pH-TDS EC ORP Temp 5 in 1 Meter EZ-9910. While microbial growth was measured using the MLSS parameter, where MLSS was measured using the gravimetric method.

5. Results of examining the indicator of the sample

5. 1. Pollutant removal in the AF2B reactor

Fig. 2 shows the experimental data on the AF2B reactor, where the data is the result of sampling every 15 minutes after the hydraulic residence time is reached (HRT: 5 hours). Fig. 2 shows that at HRT: 5 hours, all pollutants concentration in the effluent decreased, and the maximum decrease occurred at 45 minutes after discharge from the reactor, i. e.: TSS (90 %), BOD (90 %), COD (88 %), NH3-N (81 %), and Chlorine (82 %). After 45 minutes, the effluent flow at steady state so that it can be said that the AF2B process experienced the optimum conditions for HRT: 5 hours at 45 minutes. Fig. 2 shows that BOD, COD, and TSS decreased faster than other pollutants in the 30^{th} to 45^{th} minutes. Meanwhile, chlorine decreased the slowest compared to other pollutants. Chlorine is a disinfectant compound, so it has the potential as an inhibitor for the bacterial consortium contained in the AF2B reactor. Sampling was stopped because the changes in pollutant degradation by the consortium bacteria had reached a constant condition.

Time 0–15 minutes is the time when the wastewater begins to discharge of the AF2B reactor after staying for 5 hours in the reactor, and no sampling is done. Meanwhile, after 75 minutes, no sampling was carried out because the conditions were steady state, and the measurement results did not change significantly.

Based on the results of mathematical analysis shows that the concentration of each pollutants (Y) is influenced by decomposition time (X) by following the non linier regression (polynomial equation) of order 3, as follows:

Y_{TSS}=0.0833X^3+0.5714X^2–12.655X+67.6 with R^2: 0.9775;

Y_{BOD}=–0.4167X^3+7.857X^2–47.44X+127 with R^2: 0.9933;

Y_{COD}=–0.4167X^3+7.3571X^2–41.94X+133 with R^2: 0.9889;

$Y_{NH3\text{-}N}$=–0.3333X^3+5.8571X^2–33.524X+112.4 with R^2: 0.9954;

$Y_{Chlorine}$=–0.3333X^3+4.7143X^2–22.381X+58.6 with R^2: 0.9986.

An R^2 value>0.96 indicating that the pollutant concentration and the decomposition time are closely related.

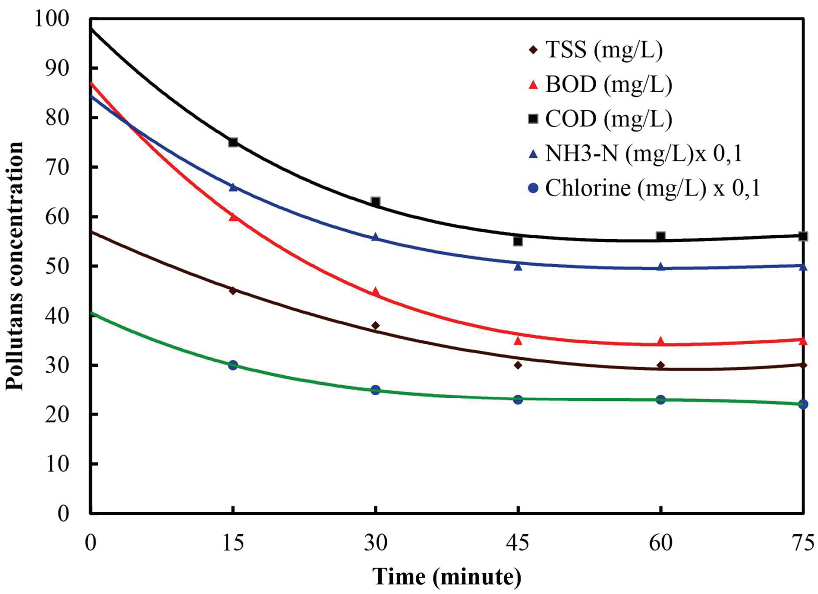

Fig. 2. Pollutant's removal in aerated fixed film biofilter reactor with hydraulic residence time of 5 hours

5. 2. Pollutant removal in the GAC

The effluent from the AF2B reactor with the same characteristics is pumped continuously at a flowrate of 0,167 L·min^{-1} into an adsorption column containing granular activated carbon which is placed in a bed, so that the adsorption process of pollutants occurs by the activated carbon (adsorbent). Sampling is done when the first effluent out (5th minute), and the next 5 minutes continuously.

Fig. 3 shows that the concentration of pollutants decreased in the 5th minute of the effluents leaving the adsorption column, and continuously decreased until the 20th minute. This shows that at minute 5th, pollutant adsorption has not (slightly) occurred so that the influent is the same as the effluent concentration. This means that the adsorption process in the first 5 minutes takes place on the adsorbent surface. While in the 10th to the 20th minute the maximum pollutant adsorption process occurs. After the 20th minute, the ability of the adsorbent to adsorb decreased due to the achievement of a saturation state on the surface of the adsorbent. All pollutants except chlorine experienced a very significant removal in the 5th to 15th minute.

At the end of the adsorption process, the concentration of each pollutant is TSS (5 mg·L^{-1}), BOD (2 mg·L^{-1}), COD (12 mg·L^{-1}), NH3-N (0.6 mg·L^{-1}), and Chlorine (not detected). This shows that the adsorption process using granular activated carbon is very effective in reducing the concentration of pollutants, where the absorbent size of 100–200 mesh has an impact on increasing the Van der Wall force between the adsorbate and adsorbent. The performance of the adsorption process is influenced by several factors, including: ratio of adsorbate-adsorbent, adsorbent weight, type of adsorbent, and the adsorption process [5].

Based on the results of mathematical analysis shows that the removal of pollutants (Y) in the granular adsorption column is influenced by time (X) by following the polynomial equation of order 3–5, as follows:

Y_{TSS}=–7E-05X^4+0.009X^3–0.2494X^2+ +0.5648X+29.889 with R^2: 0.9959;

Y_{BOD}=0.0063X^3–0.2054X^2–0.1257X+ +35.96 with R^2: 0.9784;

Y_{COD}=–7E-05X^5+0.0239X^4–0.064X^3+ +0.1617X^2–0.4567X+60 with R^2: 1;

$Y_{NH3\text{-}N}$=–5E-06X^5+0.0002X^4+ +0.0067X^3–0.305X^2–0.9367X+43 with R^2: 1;

$Y_{Chlorine}$=0.0008X^3–0.026X^2+0.046X+ +2.9762 with R^2: 0.9548.

An R^2 value>0.97 indicating that the pollutant concentration and the adsorption time are closely related.

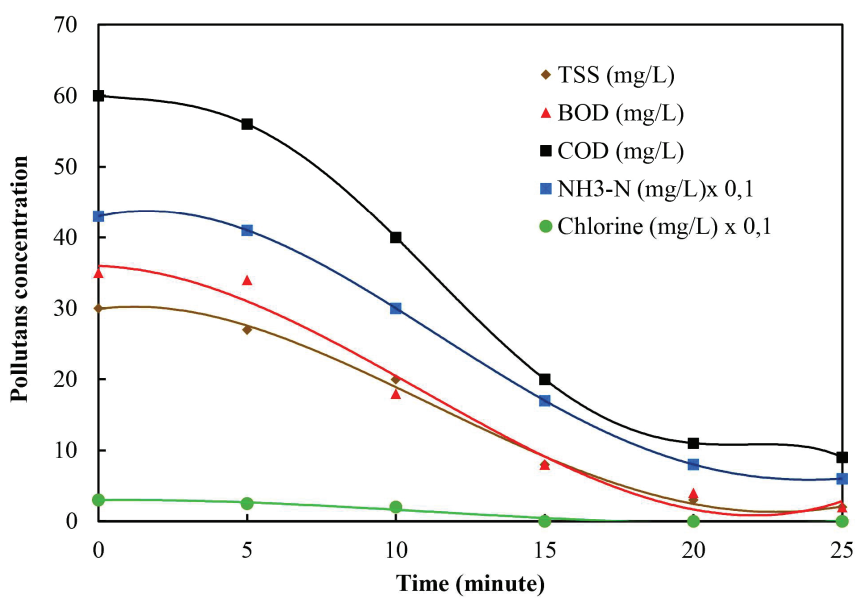

Fig. 3. Pollutant's removal in granular activated carbon column

5. 3. Pollutant removal in the CNTs

The effluent from the adsorption column containing contaminants with the same characteristics is collected in a temporary storage tank to flow into the nano adsorbent column (CNTs) which are arranged in series with particle sizes of 1,000–500–100 nm. Wastewater is pumped using a jet pump at a pressure of 606.2 kPa. While the pressure difference in and out of CNTs is maintained at 5–15 kPa. The magnitude of the pump pressure, the concentration of total dissolved solids (TDS), pH, and the kinds of the adsorbent affect the formation of fouling in the adsorption process. To obtain continuous flow, it is necessary to adjust the pump pressure were using multiwalled carbon nanotubes (MWCNTs) will provide a large pressure so that a fluid (wastewater) can flow, although with very large pressure it can affect the effectiveness of adsorption.

The results of the analysis of the samples that came out of the nano adsorption process (CNTs) showed that all pollutants (TSS, TDS, BOD, COD, NH3-N, and Chlorine) were not detected. Thus, the nano adsorption process can remove 100 % of pollutants. Meanwhile, the system performance of the combined AF2B-GAC-CNTs process in removal pollutants from wastewater from the seaweed processing industry can be seen in Table 2.

Table 2

Concentration of pollutants at the inlet-outlet of the process unit (AF2B-GAC-CNTs)

Parameters	AF2B		GAC		CNTs		Drinking water standard
	in	out	in	out	in	out	
TSS ($mg \cdot L^{-1}$)	290	30	30	5	1.7	ND	NA
TDS ($\mu g \cdot L^{-1}$)	NM	NM	2.800	495	490	ND	1,000
BOD ($mg \cdot L^{-1}$)	350	35	35	2	2	ND	NA
COD ($mg \cdot L^{-1}$)	440	56	60	12	10	ND	NA
NH3-N ($mg \cdot L^{-1}$)	27	5	4.3	0.6	0.7	ND	1.5
Chlorine ($mg \cdot L^{-1}$)	13	2.3	2.0	ND	ND	ND	250

Note: ND – not detected; NM – not measured; NA – not available

The use of the TDS parameter in the CNTs column is due to the difficulty of measuring TSS. Several parameters such as TSS, BOD, COD, NH3-N, and Chlorine were not detected using existing tools and methods, while TDS was not detected due to lack of equipment accuracy. The drinking water standard that applies is the drinking water standard in Indonesia.

6. Discussion of the research of aerobic biofilter-granular adsorption-nano adsorption in removal pollutants

Fig. 2 shows that the AF2B reactor containing a honeycomb-shaped biofilter as a medium for microorganism growth has a good ability to degrade pollutants contained in wastewater especially in degrading biodegradable organic materials (BOD, COD). This condition is due to the honeycomb-shaped biofilter has a large surface area so that the contact and decomposition of pollutant by microbes into non-pollutant compounds is getting bigger [28]. The results of previous studies stated that the AF2B reactor has an effective ability to reduce the concentration of pollutants in hospital wastewater, i. e.: BOD (92 %), NH3-N (72 %), COD (88 %), and Phenol (88 %) [10, 11]. The research on acrylonitrile wastewater shows that optimum conditions were obtained at the lowest flow rate of 1.25 $L \cdot hr^{-1}$ for all the parameters and the efficiency decreased with the average values of 83.32 % for BOD, 73.31 % for COD and 71.24 % for the phosphate [1]. While AnF2B as a modified form of AF2B which operates under anaerobic conditions for cassava wastewater treatment shows good performance which can COD removal of 98 % and produce biogas of 4.8 $L \cdot day^{-1}$ at HRT: 6 days and OLR: 1.72 $g \cdot day^{-1}$ [29].

Thus, the AF2B reactor as a type of membrane bioreactor (MBR) which operates continuously with HRT for 5 hours has a very good performance to reduce pollutants compared to the fixed film reactor (FFR). This is because microorganisms (a consortium of bacteria) have excellent ability to degrade contaminants into non-polluting materials, and the growth media in the form of a honeycomb with a large surface area is a good medium for contact between microorganisms and the substrate (pollutants) [13].

Fig. 3 shows that adsorption process by granular adsorbent carbon to pollutants is excellent. Adsorption of pollutant in the wastewater is caused by capillary force with GAC pores that is higher than pollutant with the solute itself, hence organic molecules are attracted inside the pores of GAC. However, when the saturation point is reached, COD removal using GAC is no longer effective. For 5,000 g of GAC used, after 20 min, the GAC has reached its saturation point and the adsorption rate becomes slower. Inconsistency shown can be the result of weak Van der Walls force between the pollutant and activated carbon. In the 20th minute, the concentration of each pollutant decreased by TSS (93 %), BOD (99 %), COD (98 %), NH3-N (98 %), and Chlorine (100 %), respectively.

Biological granular activated carbon (BGAC) better than that of GAC, which BGAC and GAC able to DOC removal of 96 % and 54 %, respectively [30]. While the combination of ozonation and adsorption can remove COD of 377.12 $mg \cdot L^{-1}$ and TSS of 26 $mg \cdot L^{-1}$ [31]. Thus, the adsorption process using granular activated carbon has good performance to reduce the concentration of suspended solids, organic matter, and disinfectant (chlorine) to exceed the quality standard.

There are two types of CNTs, namely single walled carbon nanotubes (SWCNTs) and multiwalled carbon nanotubes (MWCNTs) where SWCNTs are formed from units of rolled graphene sheets whereas MWCNTs are formed from many rolled graphene sheets [27, 32].

The experimental results show that there is a very significant decrease in pollutant concentration, where all parameters are not detected. Table 2 shows that the effluents of the nano adsorption process using CNTs have a quality that far exceeds the drinking water standard, where several parameters are not detected (BOD, COD, NH3-N, and COD). The adsorption ability of CNTs in reducing pollutant depends on the functional groups of the surface of the adsorbent and adsorbate, where the acidity of the surface prefers adsorption on polar compounds or in other words, the surface of non-functional CNTs has higher adsorption on non-polar compounds [25, 33].

On the other side, the optimum combination of nanofiltration and activated carbon depends on local boundary conditions such as size of plant, raw water characteristics and plant location. In the light of growing water scarcity and increasing concerns about organic micropollutants activated carbon treatment in combination with nanofiltration has the potential to be applied in an increasing number of cases [24, 34].

This study has limitations in terms of waste characteristics which are generally expressed as organic load. The characteristic of wastewater in this study has a BOD of 300–500 mg·L^{-1}. Where in real conditions in the field, wastewater can have a BOD concentration of more or less than the BOD for this research. It can result in very significant deviations in the ability of the reactor to removal pollutants. Another limitation of this research is the processing capacity or volume of treated wastewater, which for real conditions in the field has the capacity and volume of treatment it could be 1,000 times. However, this limitation can be solved by referring to the hydraulic residence time, where the hydraulic residence time has a direct correlation with the volume and flowrate of the waste. Therefore, in terms of processing capacity, the possibility of deviations is less significant when applied in the field. Thus, for real applications in the field, the results of this study have less significant deviations when using the parameters of 5 hours hydraulic residence time, adsorbate-adsorbent ratio, and nano-adsorbate volume ratio.

This research shows that the combination of the aerobic biofilter – GAC–CNTs process provides a very good ability to reduce pollutants from wastewater seaweed industrial into treated water that meets drinking water standards. Thus, the research provides hope and answers to the problems faced by several seaweed processing industries and provides hope for the provision of drinking water in the future. However, to get more valid research results, further comprehensive research needs to be carried out.

7. Conclusions

1. The aerobic biofilter process can removal of pollutants by 81–90 %, where the pollutant removal pattern follows the polynomial mathematical equation on the order of 3. The removal of pollutants in the aerobic biofilter process is affected by the decomposition time, where the pattern of removal follows a polynomial mathematical equation of order 3 with a removal of 81–90 %.

2. The granular adsorption process can removal of pollutants by 80–100 %, where the pollutant removal pattern follows the polynomial mathematical equation on the order of 3–5. The removal of pollutants in granular adsorption process is affected by the adsorption time, which the pattern of removal follows a polynomial mathematical equation of order 3–5 with a removal of 80–100 %.

3. The nano adsorption process has excellent performance in pollutant removal with pollutant removal of 100 %.

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