



Article Operation of Different Reverse Osmosis (RO) Membrane Modules for the Treatment of High-Strength Wastewater to Enhance the Recovery of Clean Water—A Case Study in Bac Ninh, Vietnam

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Abstract: In this study, a wastewater treatment plant (WWTP) in Vietnam receiving high-strength wastewater with COD of about 30,000 mg/L and various heavy metals from industries was treated by different RO membrane modules in order to meet the stringent national discharge standard and recover wastewater for reuse. The Fenton and coagulation pre-treatments were employed based on optimal conditions, which were experimentally pre-determined. For the RO membrane system, the two-stage treatment employed a plate frame RO (PFRO) followed by spiral wound RO (SPRO) to obtain high-quality permeate, while the high-pressure PFRO (HP PFRO) module was employed for the recovery of concentrated streams from the PFRO unit. As a result, a significant COD removal efficiency of 99.62% was achieved in the SPRO module. The heavy metal concentrations (i.e., Cu, Fe, Zn, Mn, Cr) measured in the output mostly met the standards for discharge levels. A significant decrease in electrical conductivity (EC) to below 250 μ S/cm was achieved. In addition, high rates of water recovery were achieved from the RO modules (i.e., PFRO 63%, HP PFRO 9–12%, SPRO > 80–90%). The high-quality treated wastewater was thus suitable for reuse purposes. This study highlights the feasibility of RO membranes for practical treatment of high-strength wastewater and provides valuable data for the WWTP operator.

Keywords: high-strength wastewater; water reuse; RO membrane modules; water recovery rate; electrical conductivity

1. Introduction

The electronics industry, such as semiconductor and nano-electronics production, generates highly contaminated effluents. Different from some biodegradable wastewater sources (e.g., fisheries wastewater [1,2]) which can be employed for biomass production, the treatment of wastewater from the electronics industry is complicated due to various chemicals, organic solvents, and oil generally used in most production steps [3,4]. Metal-laden wastewater with a wide-range of concentrations is also released from the manufacturing of electronic devices and metal electroplating. The properties of these wastewater sources (i.e., non-biodegradability, persistence, and accumulation of toxic compounds) cause severe effects on the environment. Heavy metal toxicity, for instance, is one of the biggest concerns in the treatment of wastewater from electronics manufacturing [5,6]. Therefore, the collection, treatment, and management of high-strength wastewater sources are crucial to eliminate toxic pollutants and harmful effects before discharging the treated water into the receiving sources.

The elevated toxic properties of high-strength wastewater make conventional treatment methods, such as biological processes, unfeasible. In contrast, physicochemical



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Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). methods, such as membrane filtration, ion exchange, adsorption, electrochemical, and advanced oxidation, have been commonly employed due to their advantages in removing recalcitrant compounds [5–8]. Among these methods, membrane filtration, especially using reverse osmosis (RO) membranes, has been commonly reported as a feasible method to produce high-quality treated water [9,10]. Treated water from RO membranes can be reused for different purposes [11]. Other treatment methods need further processes to obtain reusable water output. However, physicochemical pre-treatment steps are required before using RO to avoid membrane fouling and maximize performance [12]. Advanced oxidation processes (AOPs), such as the Fenton process, can rapidly degrade persistent organic pollutants (POPs), and thus decrease the COD concentration [13,14]. On the other hand, coagulation increases the removal efficiency of organic matter and suspended solids (SS) [15]. Several studies have investigated RO membranes for the treatment of high-strength wastewater; however, a complete process, including pre-treatment steps and membrane stage with actual toxic wastewater, has not been examined closely in Vietnam.

This study investigated a high-strength wastewater treatment plant (WWTP) owned by Green Star Environment Co., Ltd. with a capacity of 200 m³/day. The WWTP is located in Dong Sai village, Phu Lang Commune, Que Vo District, Bac Ninh Province, Vietnam. The WWTP location is on a plain with stable geological features and flat topography. The WWTP is designed to receive the effluents from different industrial factories in the local area (i.e., electronics manufacturing, metal electroplating, painting processes, waste gasoline). The treatment process diagram is presented in Figure 1. In this process, the reverse osmosis (RO) membrane modules are the main treatment stage, and Fenton-based AOP and coagulation serve as the pre-treatment stages. Since the WWTP is not fully operated due to the lack of experimental data, this study helps in determining the optimal operation mode which can be adopted by the plant.

Furthermore, the membrane module (e.g., plate and frame; spiral wound) directly affects the direction of feed flow which then affects the formation of foulants and cleaning process accordingly [16,17]. Thus, different RO membrane modules should be tested to maximize the quantity of clean water produced and minimize the concentrated flow or secondary waste.

Furthermore, the stringent discharge standards according to the Vietnamese national technical regulation on industrial wastewater (i.e., QCVN 40:2011/BTNTMT, level A), are also a target that the treatment system must satisfy.

This study was conducted to investigate the treatment efficiency of the WWTP for high-strength wastewater. The removal of COD, heavy metals, and levels of electrical conductivity were observed with the aim of meeting the current national standard of wastewater discharge (i.e., QCVN 40:2011/BTNTMT, level A). To investigate the important roles of different RO modules in producing clean water and their effects on the water recovery rate (R_C) during the operation, three types of RO membranes, including plate and frame RO (PFRO), high-pressure PFRO (HP PFRO), and spiral wound RO (SPRO), were employed. The two-stage treatment of feed wastewater by PFRO and SPRO, and the recovery of the concentrated flow by HP PFRO are designed to maximize clean water production. The economic aspect is also considered, and the reuse of wastewater after treatment is proposed for further works, which partially helps to reduce the operational cost. The novel aspect of this study is the use of different RO modules to produce high-quality water output for further reuse and enhance the water recovery while meeting the stringent standards. This arrangement of different modules can be successfully utilized by local agencies and industries generating high-strength wastewater.



Highly toxic WW inlet, 200 m³/day

Figure 1. Flow diagram of the wastewater treatment plant.

2. Materials and Methods

2.1. Wastewater Samples Characterization

To choose an appropriate operation mode, the wastewater samples were collected directly from the storage tank of the WWTP and analyzed for various parameters. The wastewater is complex due to the co-existence of various pollutants and toxic compounds. High COD concentration and different heavy metals at different concentration ranges were found, as summarized in Table 1. During five months of monitoring, the concentration of contaminants fluctuated strongly, making the required the wastewater treatment more

complicated. As shown in Figure 1, a treatment process was designed that comprised three main units (i.e., a Fenton reaction tank, coagulation tank, and RO membrane system). The treated wastewater is expected to meet the discharge standard and the reuse of wastewater after treatment is also an objective. Figure 1 represents the actual treatment system that is currently under investigation and monitoring.

No.	Parameters	Unit	Concentration of Wastewater before Treatment ^(*)	Water Quality Requirement after Treatment National Standard QCVN 40:2011/BTNMT (Level A)
1	pН	-	3.5–9.5	6–9
2	COD	mg/L	23,707–37,333	75
3	Chromium (VI)	mg/L	1.6–3.2	0.05
4	Copper (Cu)	mg/L	372–532	2
5	Zinc (Zn)	mg/L	3.64–375.7	3
6	Manganese (Mn)	mg/L	11.19–36	0.5
7	Iron (Fe)	mg/L	113.6–460	1

Table 1. Characteristics of high-strength wastewater sample collected at the WWTP.

 $^{(*)}$ The values were obtained during the continuous monitoring of the wastewater treatment system for 5 months with several sampling times.

The regulated discharge standard for each specific parameter according to the QCVN 40:2011/BTNMT, level A, is also presented in Table 1 to highlight the water quality requirements after treatment, compared with the raw wastewater.

2.2. Investigation of On-Site Operation at WWTP

Before the WWTP operation, preliminary experiments at a lab-scale were conducted to determine the optimal parameters of physicochemical pre-treatment processes (i.e., Fenton and coagulation processes). Accordingly, for the Fenton experiments, four duplicate experiments were conducted to determine the optimal values of reaction time, pH, H_2O_2 dosage, and FeSO₄ dosage. For the coagulation experiments, three duplicate tests were carried out to determine the optimal pH, PAC dosage, and PAM polymer dosage. The pre-treatment helped in improving the performance of the RO membrane system in the next stage.

As a result, the optimal conditions of the Fenton process were determined as follows: a reaction time of 60 min, a pH of 3.0, a H_2O_2 dosage of 600 mg/L, and a FeSO₄ dosage of 200 mg/L. For the coagulation, a pH of 7.5, PAC dosage of 700 mg/L, and anion PAM polymer dosage of 15 mg/L are defined as optimal values.

The WWTP (Figure 1) was then operated based on these optimal conditions identified in the lab-scale experiments. Specifically, at the Fenton unit, the oxidizing agents H_2O_2 (600 mg/L) and the catalyst FeSO₄ (200 mg/L) were employed. The chemical Fenton reaction can degrade refractory organic matters or certain inorganic pollutants and help to increase wastewater biodegradability [13]. The concentrated sulphuric acid (H_2SO_4 , 98%) was used to adjust the initial pH since an acidic environment is favorable for the generation of OH[•] groups during the Fenton reaction.

For the coagulation, a coagulation tank integrated with a sedimentation system was installed to improve the separation of flocs. The PAC coagulant and PAM flocculant were used to form the large-size flocs, which were then removed from the wastewater by either flotation with dissolved air bubbles or settling down due to gravity. The calcium hydroxide solution (i.e., Ca(OH)₂, 92%) was also added to adjust the pH and improve the flocs adhesion.

For the RO membrane system, the technical specification of each RO module and operational parameters are presented in the flow diagram in Figure 2. The coarse filtration

and microfiltration (MF) module (pore size of 10 μ m, Pentair, USA) were designed as pre-filtration units before the RO modules. Three different types of RO modules (pore size of 0.0001 μ m, ROCHEM, Germany) (i.e., PFRO, HP PFRO, and SPRO) were installed at the same time to treat the feed wastewater and recover the concentrated flow. Specifically, the PFRO (Plant A) received the output flow of MF filtration, whereas HP PFRO (Plant B) was used to treat the concentrated flow. In order to enhance the quality of treated wastewater, the permeate flow from both PFRO and HP PFRO modules was passed to the SPRO module (Plant C). The feed flow, working pressure, and expected R_C were designed differently among these RO modules. During the operation, 30% hydrochloric acid (HCl), sodium metabisulphite (SMBS, Na₂S₂O₅), and other common antiscalants, were used for membrane cleaning and prevention of fouling.



Figure 2. Flow diagram and technical specifications of the RO membrane system.

Due to the complexity of the real wastewater inlet, the investigation of the WWTP operation was carried out continuously for five months (i.e., from April to August 2021). The wastewater at each stage (i.e., inlet, after Fenton reaction tank, after coagulation tank, after the RO membrane system outlet) were sampled periodically to test the concentration of COD and heavy metals for the evaluation of treatment efficiency.

2.3. Analytical Methods and Calculation

During the experiments and operation of the WWTP, the wastewater samples were stored until analysis according to the standard methods [18]. The physical and chemical parameters were then analyzed and measured under laboratory conditions following standard methods.

Specifically, pH was measured by a portable pH meter (HI8314, Hanna Instruments, Nusfalau, Romania); COD concentration was analyzed and measured by a COD heating reactor (HI839800-02, Hanna Instruments, Romania) and a COD Benchtop Photometer

(HI83314-02, Hanna Instruments, Romania). The operation of the RO membrane system was controlled automatically by a Programmable Logic Controller (PLC), and the monitoring results (i.e., flow rate, water recovery rate, electrical conductivity) were obtained directly as the output of the PLC program. Heavy metal concentrations were measured by an Inductively Coupled Plasma (ICP) Spectrometer (Optima 8000, PerkinElmer, Waltham, MA, USA). All the analyses and measurements were performed in triplicate. The results reported are average values, and the standard deviation was calculated.

All the experiments and operations were conducted at the research lab and the WWTP on-site in Bac Ninh province, Vietnam. All chemicals used in this study are analytical grade. The solutions and reagents were prepared by using deionized water.

The water recovery rate (R_C) in the permeate stream of the RO system is calculated by the ratio between the flow of permeate flow (Q_p) and feed flow (Q_f), as shown below.

$$R_c = \frac{Q_p}{Q_f} \cdot 100\% \tag{1}$$

3. Results and Discussion

3.1. Performance of WWTP in Meeting the Current Discharge Standard 3.1.1. COD Removal Efficiency

The wastewater samples were collected at different points (i.e., the inlet, Fenton outlet, coagulation outlet, and SPRO outlet) of the WWTP during the investigation period of 5 months. The removal efficiency was calculated based on the COD concentration measured in the real wastewater samples. Due to the large number of samples analyzed and results obtained in this long-term investigation, the results recorded during June 2021 were chosen and are shown in Figure 3.



Figure 3. Changes in COD concentration in different treatment stages at specific sampling times during June, 2021.

Figure 3 shows that the COD concentration at the inlet varied in a wide range of 25,031–32,379 mg/L. This is an inevitable obstacle since the WWTP always receives the real wastewater from various industrial effluents, which significantly affected the operation and treatment efficiency. As a result, the COD concentration in the outlet of each treatment step changed accordingly with the variation of the inlet. At the end of June (i.e., 28/06, 29/06, 30/06, Figure 3), when the COD concentration of the inlet was relatively stable in the range of 31,794–32,379 mg/L, a stable removal efficiency was obtained. Specifically, after treatment with the Fenton reaction tank, the COD decreased to 23,425–23,557 mg/L, corresponding to a removal percentage of 26.32–27.29%. Similar results were found in

the coagulation tank where the COD concentration dropped to 18,462–18,658 mg/L, corresponding to a removal efficiency of 20.75–21.19%. The operation of physicochemical pre-treatment units was based on the optimal parameters, which were experimentally identified in the laboratory in advance (as described in Section 2.2). This helped to optimize the treatment efficiency and minimize the operational cost. It has been reported that the Fenton process, when operated at optimal pH and dosages of Fe^{2+} and H_2O_2 along with their optimal molar ratio, can minimize the scavenging of OH[•] reactive radicals and significantly affects the COD removal [13]. For coagulation, although a slightly acidic pH (i.e., around 6.0) is frequently reported and suggested for wastewater treatment [19], in this study, a neutral pH (i.e., 7.5) was suggested to maintain at the WWTP due to the complicated effects on the wastewater components and characteristics. Additionally, the co-existence of a variety of organic and inorganic matter, such as heavy metals at very high concentrations from different wastewater sources collected, may cause different interferences between colloids and the PAC coagulant. It is also reported that the addition of PAM anion polymer at optimal dosage significantly affects the performance of coagulation due to the improvement in floc formation and prevention of the re-stabilization of colloids [15]. In this study, it is believed that PAM polymer helped to improve the overall efficiency of pre-treatment step.

Wastewater, after pre-treatment by Fenton and coagulation, was transferred to the membrane system, and the SPRO module served as the final step of the treatment process. The results show that SPRO could produce high-quality treated water since the COD concentration in the permeate flow decreased sharply to 51–75 mg/L (Figure 3), corresponding to a removal percentage of over 99%. The two-stage treatment of permeate flows, as shown in Figure 2 (i.e., the permeate from both PFRO and HP PFRO was passed to the SPRO module) resulted in very high COD removal efficiency. The effluent COD concentration meets the allowable discharge limits according to the national standard (i.e., QCVN 40:2011/BTNMT, level A, COD of 75 mg/L), as shown in Table 1. This result was repeated in the data recorded during the 5 months of investigation, indicating the stability of the treatment system. The low COD concentration remaining in the permeate flow of the SPRO system also indicates the opportunity to reuse of treated wastewater.

3.1.2. Heavy Metals Removal Efficiency

Heavy metals with a high concentration in the inlet flow are a particular feature of the WWTP since the wastewater was mostly from metal-related industries. Removal of heavy metals to meet the discharge standard is also an important target of the treatment process. At the beginning of operation (i.e., during two months from April to May 2021), the heavy metals removal was not effective since their concentrations measured in the permeates of SPRO system were still higher than the allowable levels as regulated. This may be due to the complexity of the wastewater inlet and the low adaptability of the WWTP in the early stage.

After that, the remaining measurements of heavy metal concentration met the national standard. Specifically, Figure 4 presents the concentration of five representative metals (i.e., Cu, Fe, Zn, Mn, Cr) in the permeate flow of SPRO (also the outlet of the WWTP) at different sampling times during the last three months of the investigation period (i.e., from June to August 2021). They are mostly lower than the corresponding allowable concentration for discharge (dashed lines in Figure 4). Although the concentrations of Fe²⁺ (24/06), Cu²⁺ and Zn²⁺ (29/07), and Cr⁶⁺ (26/08) were still slightly higher than the allowable limits at some specific times, the general removal efficiency of heavy metals was considerably improved. Compared with the initial concentrations of heavy metals in the inlet flow (Table 1), they decreased sharply at the final treatment stage of WWTP (i.e., SPRO membrane system).



Figure 4. Changes in heavy metals concentrations in the permeate flow of SPRO membrane at specific sampling times. (ND: Not detected due to the limit of detection of measurement methods).

The stability and efficiency of the physicochemical pre-treatment helped to obtain the high removal efficiency of pollutants in the RO membrane system. The Fenton and coagulation processes mitigated the heavy metals concentrations before the wastewater flow entered the membrane system. It has been reported that advanced oxidation could be applied successfully to treat high-strength wastewater, such as landfill leachate, in which heavy metal is one of the major contaminants [13,20]. RO membrane technology is widely known as an effective process for removing heavy metals from the effluents of different industrial sectors [5,12]. In this study, the combination of Fenton-based AOP and coagulation as pre-treatment and RO membrane technology was demonstrated to be effective in removing heavy metals.

3.1.3. Electrical Conductivity and Water Recovery

The differences in the performance of the three RO membrane modules at three plants (A, B, and C, as shown in Figure 2) was assessed by the electrical conductivity (EC, μ S/cm), the flow of permeate, and the corresponding water recovery rate (R_C). Due to the large amount of data produced automatically from the PLC program for controlling the membrane system and the operational cycle for each plant, this study presents a selection of representative data during 96 h of continuous monitoring for plant A and plant C and 72 h for plant B (Figure 5).

Basically, EC represents the occurrence of dissolved conductive material commonly existing in wastewater. EC is generally converted to total dissolved solids (TDS), and the ratio between EC and TDS varies depending on the types of water (e.g., fresh water, saline water, wastewater) [21,22]. In this study, the wastewater sources transferred to WWTP were mostly from electronic manufacturing and metal electroplating factories, which contained a high concentration of electrically charged components. Accordingly, it is important to consider this parameter. Low EC measured in the permeate flow corresponds to a low concentration of dissolved conductive pollutants, indicating good performance of the membrane process. Figure 5(a1,b1,c1) show the EC measured in the PFRO (plant A), HP PFRO (plant B), and SPRO (plant C), respectively.



Figure 5. Changes in EC (**a1**,**b1**,**c1**) and flow rate (**a2**,**b2**,**c2**) with time during the operation of the PFRO (plant A), HP PFRO (plant B), and SPRO (plant C), respectively.

For plant A (Figure 5(a1)), the EC of the feed flow increased from 25,000 to 31,800 μ S/cm during the 80 h of operation but slightly decreased thereafter due to the change of wastewater inlet. The permeate flow, however, was relatively stable with a low average EC of 1138 μ S/cm being observed during the monitoring period. This result demonstrates the high capacity of the PFRO membrane for retaining conductive components. A similar result was found for HP PFRO (plant B, Figure 5(b1)), in which the concentrated flow released from plant A was treated to recover the clean water and minimize the disposal of concentrated waste. Although very high EC often existed in the feed flow (i.e., 51,812–64,118 μ S/cm) and fluctuations occurred every 6 h during the startup cycle, the EC measured in the permeate of the HP PFRO membrane was stable with a low average value of 1292 μ S/cm. For SPRO (plant C), the feed flow was the confluence of the

permeate from PFRO and HP PFRO. Thus, the EC fluctuated according to the fluctuations of flow and wastewater characteristics during the operating hours (Figure 5(c1)). Specifically, the EC of the feed flow varied within low range of 700–2200 μ S/cm. However, the EC measured in the permeate flow of SPRO was below 250 μ S/cm during the operation. This demonstrates the importance of the SPRO membrane to producing high-quality permeate since low outlet EC corresponds to low TDS concentration. In this study, the ratio between EC and TDS in the outlet flow was chosen as 0.64, as previously reported [21]. Accordingly, the TDS concentration was about 160 mg/L. The current Vietnam standard for industrial wastewater (i.e., QCVN 40:2011/BTNTM, level A), however, does not regulate the EC and TDS. On the other hand, the domestic wastewater standard (i.e., QCVN 14:2008/BTNMT, level A) specifies the limit for this parameter at 500 mg/L. Therefore, it can be considered that the EC of the effluent from SPRO met the discharge limit.

In terms of the flow rate of permeate and R_C from each RO module, Figure 5(a2,b2,c2) present the changes in the feed and permeate flow by time, from which the corresponding R_C was determined. Different results were observed among the RO modules due to differences in the feed flow properties and working pressure at each plant (i.e., 65, 90, and 41 bar corresponding to plants A, B, and C, respectively). The working pressure was determined through a preliminary test before an operation, which was based on the guidelines of the membrane manufacturer (ROCHEM, Germany) and the actual characteristic of the wastewater feed flow.

For the PFRO module operated at 65 bar (plant A, Figure 5(a2)), the feed flow was maintained at 9600 L/h during the operation, while the permeate flow changed periodically depending on the cleaning cycle (i.e., 6000 L/h during the first 36 h, decreased to 5000 L/h from the 36th to 48th hour, increased again to 6000 L/h from the 48th hour due to chemical cleaning, and decreased to 4400 L/h after the 80th hour). The average R_C at the beginning of the operation and after cleaning was about 63%, but after 80 h, R_C slightly decreased to 47%. This result was aligned with the expected R_C (i.e., 50–75%) of the PFRO design (Figure 2).

The feed flow of the HP PFRO module (plant B, Figure 5(b2)) was designed to be 4800 L/h, although the working pressure was high (i.e., 90 bar) as this received the concentrated flow of PFRO. Compared with PFRO, a different trend in permeate flow was observed for HP PFRO in which the cleaning cycle was conducted more often (i.e., every 6 h) and resulted in the continuous recovery of permeate flow. The average permeate flow was 439 L/h, corresponding to an R_C of 9.14%. At some specific times, especially after membrane cleaning, R_C could be improved to over 12%. The design of HP PFRO was expected to recover 10–20% clean water (Figure 2). The results indicated that frequent cleaning helped to improve the R_C of the HP PFRO module.

In the case of the SPRO membrane system (Plant C, Figure 5(c2)), the operation was divided into two levels of feed flow (i.e., 4000 L/h and 8000 L/h), which were controlled and monitored through a high-pressure inverter pumping system. Accordingly, the permeate was proportional to the feed flow since there were also two levels of flow rate, as shown in Figure 5(c2). The results show that an average R_C of 80–90% could be achieved indicating a high practical operating efficiency compared with the theoretically expected R_C (i.e., 90–95%, Figure 2). The high and stable R_C values obtained at the two different levels of feed flow also demonstrate the stability of the SPRO membrane system.

3.2. Comparison with Previous Studies

Several previous studies on the treatment of high-strength wastewater were carried out with different types of wastewater samples at specific operation conditions, which are summarized in Table 2. In most studies, membrane technology (i.e., MBR, MF, UF, RO) was employed to investigate the treatment of high-strength wastewater sources, which were characterized by high organic loading and various toxic compounds. Physicochemical methods (e.g., Fenton, aeration) were also applied as pre-treatment for the membrane process. Depending on the type of wastewater investigated and targets proposed in each study, different parameters were examined and monitored. The comparison indicates that the combination of physicochemical pretreatment and RO membrane is effective in producing high quality treated water. Specifically, the organic matter determined by COD concentration could be removed effectively (i.e., > 80%) in most studies, except in the case of Huang, et al. [23] (i.e., COD removal of 77%). The COD concentration is highlighted as the main parameter for characterizing the performance of a treatment process since most studies used high-loading organic influent. For water recovery produced from an RO system, an $R_C > 50\%$ has been reported and the opportunity of wastewater reuse has also been reported and proposed. However, in some studies [14,15,24], the flocculation and Fenton process were emphasized while the RO system was not employed in the treatment process, thus, the R_C was not mentioned.

Treatment Method	Type of Wastewater	Technical Features of Operation	Performance (Removal Efficiency)	Ref.
UV + Fenton	Textile wastewater	H_2O_2 of 660 mg/L Fe ²⁺ of 20 mg/L pH of 3.0 Reaction time of 90 min	Color of 94.5% COD of 95.5% No mention of water recovery	[14]
Coagulation + Fenton	Wastewater from printing factory	Coagulation: PAC of 150 mg/; CaCO ₃ of 67.5 mg/L Fenton: H ₂ O ₂ of 100 mg/L; Fe ²⁺ of 80 mg/L; and reaction time of 45 min	COD of 81.5%, COD of the output met the regulation as national standard QCVN 40:2011/BTNMT (level B). No mention of water recovery	[15]
CW ^(*) + UF + RO	Wastewater generated from an iron and steel factory	CW served as pretreatment; operation duration of 61 days	77.3% COD; 76.9% NH ₄ ⁺ ; 95.8% turbidity; 96.9% Fe; and 92.5% Mn; R _C = 75%; EC decreased to 10–30 μS/cm	[23]
AOP + MF	Effluent from leather industry	H ₂ O ₂ and FeSO ₄ optimal dosages were pre-determined; MF membrane operated with aerated condition	97.2% COD; 97.0% BOD ₅ ; 93.0% TSS; 87.4% oil and grease; 75% TKN; 40.8% TP. No mention of water recovery	[24]
MF + RO	Tannery wastewater	Dual-stage treatment; COD of 5680 mg/L; BOD ₅ 759 mg/L	>90% BOD ₅ , COD, TOC, NH ₄ ⁺ , Cl ⁻ , SO ₄ ² ⁻ ; reuse of 50% wastewater after treatment	[25]
MBR + MF + RO	Wastewater from paper mill	Pilot scale	EC < 200 μS/cm, COD < 15 mg/L, Turbidity < 0.1 NTU; and Color < 15 PCU; R _C > 65%	[26]
Electro-Fenton + MBR	Wastewater containing Glyphoaste herbicide	Electro-Fenton in 40 min and MBR in 24 h	The glyphosate concentration decreased from 29.5 mg/L to 0.3 mg/L COD, BOD ₅ , ammonia after treatment: 32.6, 10.8, 0.76 mg/L	[27]
Fenton + Coagulation + RO with 3 different modules	Highly toxic wastewater collected from electronic and metal electroplating factories	Optimal conditions for Fenton and coagulation were experimentally determined; The membrane system included PFRO; HP PFRO; and SPRO	>99% COD, EC of permeate decreased to < 250 μ S/cm; heavy metals concentration met the national discharge standard; average R _C = 63% with PFRO; 9–12% with HP PFRO; and 80–90% with SPRO	This study

Table 2. A comparison of recent studies on the treatment of high-strength wastewater.

^(*) CW: Constructed wetland.

The results obtained in this study are comparable with the findings reported by others. Specifically, very high COD removal efficiencies (i.e., >99%) with a significant decrease

in EC to below 250 μ S/cm, corresponding to TDS of 160 mg/L in the permeate flow of the SPRO, were found in this study. Furthermore, different RO modules (i.e., PFRO, HP PFRO, SPRO) with specific working pressures were employed in this study to maximize the water recovery from the concentrated flow and produce high-quality permeates. Such investigations have not been conducted previously. The different R_C results obtained from these RO modules were used to assess their productivity compared with the expected R_C as designed. The results can help WWTP authorities to choose the appropriate design and operation mode.

The study's results also indicate that the wastewater after treatment can be reused for non-drinking purposes, such as irrigation, cleaning, and flushing. This helps to reduce the operational cost of WWTPs, which are normally high when RO membrane technology is employed. Furthermore, all factories and manufacturers from which wastewater is currently generated and transferred to WWTP, need to pay the prescribed fee to the WWTP investors. Thus, when the WWTP is operated in a stable state with high treatment efficiency, an economic benefit can also be achieved, which contributes to minimizing the cost.

4. Conclusions

This study demonstrated successful employment of a Fenton-coagulation-RO membrane process as the main treatment methods for high-strength wastewater. The testing was conducted continuously for five months. The treatment system resulted in a COD removal by the Fenton unit of 26.32–27.29%, by the coagulation unit of 20.75–21.19%, and the SPRO membrane system of > 99%. The heavy metal concentrations (i.e., Cu, Fe, Zn, Mn, Cr) in the permeate flow of the SPRO unit were under the allowable discharged levels. The treated water meets the national discharge standard of Vietnam (i.e., QCVN 40:2011/BTNMT, level A), indicating the good performance of RO membranes. The water recovery $R_{\rm C}$ values from the three RO modules were 63% (PFRO), 9–12% (HP PFRO), and 80–90% (SPRO). For the three RO modules, high performance levels in decreasing the EC were also observed, especially in the case of the effluents of SPRO in which the EC outlet was below $250 \,\mu\text{S/cm}$, corresponding to a TDS of 160 mg/L. The results achieved in this study help to build a technical database for the operation and management of WWTP. Reuse of wastewater after treatment and the benefits gained from the collection of fees for treatment of wastewater paid by industrial manufacturers can partially reduce the operation cost of the WWTP. The combination of AOP, the coagulation process, and RO membranes is thus a feasible approach to treating high-strength wastewater.

Future Perspectives

- Further investigation on the collection, classification, and mixed ratio of different highstrength real wastewater sources should be carried out to determine the appropriate operational conditions since the properties of wastewater at the inlet are important in determining the optimum operational parameters.
- Studies on pretreatment methods should be conducted to minimize the fouling and increase the performance of the RO system, which helps to achieve higher R_c
- Future investigations should be consider methods to treat the concentrated flows from RO systems since the current treatment method (i.e., incineration) has high costs.

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References

- García-Martínez, J.B.; Sanchez-Tobos, L.P.; Carvajal-Albarracín, N.A.; Barajas-Solano, A.F.; Barajas-Ferreira, C.; Kafarov, V.; Zuorro, A. The Circular Economy Approach to Improving CNP Ratio in Inland Fishery Wastewater for Increasing Algal Biomass Production. *Water* 2022, 14, 749. [CrossRef]
- García-Martíneza, J.B.; Urbina-Suarezb, N.A.; Zuorroc, A.; Barajas-Solanob, A.F.; Kafarova, V. Fisheries wastewater as a sustainable media for the production of algae-based products. *Chem. Eng.* 2019, 76, 1339–1344.
- 3. Liu, J. Aerobic Treatment of Effluents from the Electronics Industry. In *Current Developments in Biotechnology and Bioengineering;* Elsevier: Amsterdam, The Netherlands, 2017; pp. 145–160.
- Noor, I.-e.; Coenen, J.; Martin, A.; Dahl, O.; Åslin, M. Experimental investigation and techno-economic analysis of tetramethylammonium hydroxide removal from wastewater in nano-electronics manufacturing via membrane distillation. *J. Membr. Sci.* 2019, 579, 283–293. [CrossRef]
- 5. Azimi, A.; Azari, A.; Rezakazemi, M.; Ansarpour, M. Removal of heavy metals from industrial wastewaters: A review. *ChemBioEng Rev.* 2017, 4, 37–59. [CrossRef]
- 6. Sankhla, M.S.; Kumari, M.; Nandan, M.; Kumar, R.; Agrawal, P. Heavy metals contamination in water and their hazardous effect on human health-a review. *Int. J. Curr. Microbiol. App. Sci.* **2016**, *5*, 759–766. [CrossRef]
- Ahmed, M.B.; Zhou, J.L.; Ngo, H.H.; Guo, W.; Thomaidis, N.S.; Xu, J. Progress in the biological and chemical treatment technologies for emerging contaminant removal from wastewater: A critical review. *J. Hazard. Mater.* 2017, 323, 274–298. [CrossRef] [PubMed]
- 8. Ma, W.; Ding, Y.; Li, Y.; Gao, S.; Jiang, Z.; Cui, J.; Huang, C.; Fu, G. Durable, self-healing superhydrophobic nanofibrous membrane with self-cleaning ability for highly-efficient oily wastewater purification. *J. Membr. Sci.* **2021**, *634*, 119402. [CrossRef]
- 9. Aslam, M.; Charfi, A.; Lesage, G.; Heran, M.; Kim, J. Membrane bioreactors for wastewater treatment: A review of mechanical cleaning by scouring agents to control membrane fouling. *Chem. Eng. J.* 2017, 307, 897–913. [CrossRef]
- 10. Deng, L.; Guo, W.; Ngo, H.H.; Zhang, H.; Wang, J.; Li, J.; Xia, S.; Wu, Y. Biofouling and control approaches in membrane bioreactors. *Bioresour. Technol.* 2016, 221, 656–665. [CrossRef] [PubMed]
- 11. Obotey Ezugbe, E.; Rathilal, S. Membrane technologies in wastewater treatment: A review. *Membranes* **2020**, *10*, 89. [CrossRef] [PubMed]
- 12. Trishitman, D.; Cassano, A.; Basile, A.; Rastogi, N.K. Reverse osmosis for industrial wastewater treatment. In *Current Trends and Future Developments on (Bio-) Membranes;* Elsevier: Amsterdam, The Netherlands, 2020; pp. 207–228.
- 13. Deng, Y.; Zhao, R. Advanced oxidation processes (AOPs) in wastewater treatment. Curr. Pollut. Rep. 2015, 1, 167–176. [CrossRef]
- 14. Vinh, L.X.; Phung, L.T.; Hien, T.T. Textile wastewater treatment by UV/Fenton process. Sci. Technol. Dev. J. 2015, 18, 201–211.
- 15. Viet, L.H.; Binh, T.P.; Hau, M.T.; Ngan, N.V.C. Investigation of operating parameters of flocculation combined with fenton process for wastewater treatment of printing factories. *Can Tho Univ. J. Sci.* **2017**, 162–172. [CrossRef]
- 16. Baker, R.W. Membrane Technology and Applications; John Wiley & Sons: Hoboken, NJ, USA, 2012.
- 17. Kucera, J. Desalination: Water from Water; John Wiley & Sons: Hoboken, NJ, USA, 2019.
- 18. APHA. *Standard Methods for the Examination of Water and Wastewater;* American Public Health Association (APHA): Washington, DC, USA, 2005; Volume 21.
- 19. Crittenden, J.C.; Trussell, R.R.; Hand, D.W.; Howe, K.J.; Tchobanoglous, G. MWH's Water Treatment: Principles and Design; John Wiley & Sons: Hoboken, NJ, USA, 2012.
- 20. Deng, Y.; Englehardt, J.D. Treatment of landfill leachate by the Fenton process. *Water Res.* 2006, 40, 3683–3694. [CrossRef] [PubMed]
- 21. Ali, N.S.; Mo, K.; Kim, M. A case study on the relationship between conductivity and dissolved solids to evaluate the potential for reuse of reclaimed industrial wastewater. *KSCE J. Civ. Eng.* **2012**, *16*, 708–713. [CrossRef]
- 22. Rusydi, A.F. *Correlation between Conductivity and Total Dissolved Solid in Various Type of Water: A Review;* IOP conference series: Earth and environmental science; IOP Publishing: Bristol, UK, 2018; p. 012019.
- 23. Huang, X.-F.; Ling, J.; Xu, J.-C.; Feng, Y.; Li, G.-M. Advanced treatment of wastewater from an iron and steel enterprise by a constructed wetland/ultrafiltration/reverse osmosis process. *Desalination* **2011**, *269*, 41–49. [CrossRef]
- 24. Abdel-Shafy, H.I.; El-Khateeb, M.A.; Mansour, M.S. Treatment of leather industrial wastewater via combined advanced oxidation and membrane filtration. *Water Sci. Technol.* 2016, 74, 586–594. [CrossRef] [PubMed]
- Bhattacharya, P.; Roy, A.; Sarkar, S.; Ghosh, S.; Majumdar, S.; Chakraborty, S.; Mandal, S.; Mukhopadhyay, A.; Bandyopadhyay, S. Combination technology of ceramic microfiltration and reverse osmosis for tannery wastewater recovery. *Water Resour. Ind.* 2013, 3, 48–62. [CrossRef]

- 26. Zhang, Y.; Ma, C.; Ye, F.; Kong, Y.; Li, H. The treatment of wastewater of paper mill with integrated membrane process. *Desalination* **2009**, 236, 349–356. [CrossRef]
- 27. Duong, L.T.; Son, L.T. Performance evaluation of combined electro-fenton process and membrane bioreactor (MBR) for treating wastewater containing Glyphoaste herbicide. *TNU J. Sci. Technol.* **2019**, *204*, 211–217.