



Article Textile Wastewater Treatment for Water Reuse: A Case Study

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Abstract: The reduced natural waters and the large amount of wastewater produced by textile industry necessitate an effective water reuse treatment. In this study, a combined two-stage water reuse treatment was established to enhance the quality and recovery rate of reused water. The primary treatment incorporated a flocculation and sedimentation system, two sand filtration units, an ozonation unit, an ultrafiltration (UF) system, and a reverse osmosis (RO) system. The second treatment included an ozonation unit, a sand filtration unit, and UF and RO systems. The color removal rate increased with the increasing ozone dosage, and the relational expression between the ozone dosage and color removal rate was fitted. Ozonation greatly reduced the color by 92.59 and 97.27 times during the primary and second ozonation stages, respectively. RO had the highest removal rate. The combined processes showed good performance in water reuse treatment. The treated, reused water satisfied the reuse standard and surpassed the drinking water standard rates for chemical oxygen consumption (COD_{cr}), color, NH₃-N, hardness, Cl⁻, SO₄²⁻, turbidity, Fe³⁺, and Cu²⁺. The operating cost of reuse water treatment was approximately 0.44 USD·m⁻³.

Keywords: textile wastewater; water reuse; ozonation; ultrafiltration; reverse osmosis; water recovery rate

1. Introduction

The decrease in natural water resources caused by drought and population growth has incited authorities to establish and encourage wastewater reuse. Industries are not immune to this water shortage. The increase in water costs and the obligation to respect the standards of wastewater disposal in the environment have compelled manufacturers, such as those from the textile industry, to rethink their management of residual waters [1]. Residual water has thus become a water resource, especially for high water consumers. Water consumption, wastewater treatment, and effluent reuse potential have become crucial factors for sustainable production [2].

The textile industry uses large quantities of water in stages, such as pretreatment, bleaching, dyeing, and printing, which demand approximately 100–200 L of high-quality water per kilogram of textile product and consequently generate huge amounts of dye wastewater [3]. The wastewater produced by textile industry contains organic matter, toxic chemicals derived from fixing agents, detergents, dyes, and salts and therefore is the most polluting among all industrial wastes [4]. The recalcitrant substances

in the water are harmful to aquatic life because they reduce light penetration. Therefore, searching an effective method of treating and reusing wastewater from the textile industry is highly challenging.

In combined treatment systems, pH adjustment, coagulation–flotation [5], and sedimentation are usually employed as pre-treatment phases; biological processes (aerobic and anaerobic) are appropriate for organic removal [6,7]. However, textile wastewater also exhibits high color and low biodegradability [8], thus making it difficult to treat with physio-chemical and biological treatment methods [9]. Advanced oxidation processes have emerged as an effective water treatment technology for oxidation of various organic contaminants, including those not treatable by conventional techniques [10]. Zhu et al. [11] pointed out that ultrasonic oxidation technology is very efficient for the complete decolorization and mineralization of dyes. Oppong et al. [12] came to a conclusion that visible-light-driven photocatalytic activity of Gd-TiO₂-GO nanocomposites presents better visible light photocatalytic performance and degradation efficiency over pure TiO₂. Musmarra et al. [13] discovered that hydrodynamic cavitation is valid in the removal of ibuprofen. Iovino et al. [14] found that combined treatments based on UV light compared to other advanced treatment technologies are more eco-friendly for the removal of Ibuprofen from synthetic water streams. Therein the advanced oxidation processes, ozonation is one of the best options to treat wastewater containing non-biodegradable organic contaminants and to satisfy the reuse standard [15,16]. It has proved effective on water disinfection and micropollutants removal [17]. In addition to chemical oxygen consumption (COD_{cr}) and biochemical oxygen demand (BOD₅) removal, ozonation attains a high level of color removal, which is advantageous over conventional methods [3]. The potential of ozonation makes it widely used in water reuse systems [16]. On the contrary, one of its disadvantages is the high investment costs and energy consumption [18]. Furthermore, effluent salinity must be removed for textile wastewater reuse. Lin et al. [19] pointed out that typical textile wastewater may contain 6.0 wt% NaCl or 5.6 wt% Na₂SO₄. Membrane separations may help achieve the water quality for recycling [6] and allows contaminant removal and water reuse for certain applications [20]. Lin et al. [19] designed ultrafiltration (UF)-diafiltration to separate a dye/Na₂SO₄ aqueous mixture and has achieved 98% desalination efficiency and >97% dye recovery. Lara et al. [21] used a UF ceramic membrane for the removal of a reactive dye, Reactive Black 5, which was removed by up to 95.2%. Lafi et al. [22] pointed out that primary treated textile wastewater with combined UF-electrodialysis presents similar parameter values to those of normal feed water. Amar et al. [1] researched the integrating of activated sludge treatment with either nanofiltration (NF) or reverse osmosis (RO) to reclaim wastewater from denim fabric production. NF allows a high yield while maintaining the Tunisian standard of water reuse (COD_{cr} < 90 mg L^{-1}). Jager et al. [23] pointed out that the residual color in the UF permeate can be reduced from an average of 660 ADMI (color standard used by American Dye Manufacturer's Institute) units to 12 ADMI units in the RO permeate; this value is lower than that required for potable water. Although NF shows better COD_{cr} removal efficiency than RO, its salinity removal efficiency is dissatisfactory [24]. The yield of RO is lower than that of other systems [1], and its concentrate is difficult to treat due to its high concentration of organic matter, inorganic matter, ammonia nitrogen, and salt [25]. Thus, integrating membrane technique with conventional techniques or with other membrane-based techniques in a combined system is necessary to achieve technical and economic objectives [26].

There are many wastewater treatments for textile wastewater reuse, such as constructed wetland [27,28], activated carbon [29,30], ion exchange [31,32]. However, the constructed wetland has poor removal effect on color and the occupation area of it is very large. Activated carbon has a high removal rate on water-soluble dyes, but it cannot adsorb suspended solid (SS) and insoluble dyes. Besides, the activated carbon is difficult to regenerate and thus the operating cost of it is high. Although ion exchange treatment has great removal rate on some specific dissolved pollutants, it is not suitable for the treatment of a large number of multi-component textile wastewater. UF is a good pretreatment process [19]. RO has great effects on the residual color and remaining salt [23]. Ozone is one of the best treatment method in advanced oxidation methods [33], and it is a suitable alternative specially when integrated with biological treatments [10]. However, the industrial application of O_3/UV or

 $O_3/UV/H_2O_2$ is not recommended because the investment and operating costs are too high [33]. Therefore, in this research, ozonation + UF + RO combination sequence was developed to treat and reuse textile wastewater. The ozonation process mainly aimed to reduce follow-up membrane pollution. Furthermore, specific preprocesses, such as flocculation precipitation, sand filtration, self-cleaning filter, and security filter, were added before ozonation and membrane processing to reduce the operating costs. Additionally, the reuse rate of recycled water was subsequently increased through the secondary water reuse system.

The objective of this research is to find an effective method of treating and reusing wastewater from the textile industry, and analyze its elimination effect and operation cost in different stages with the best operative conditions. Although the ability of ozonation and membrane technologies for the treatment of textile wastewater is well known, most studies focused on synthetic wastewater prepared in laboratories and used laboratory-scale plants over limited periods. Comparing to the existing work, this research used detailed field studies, pollutants monitoring and electricity log processing to investigate the elimination effect and operation cost in different stages. To the best of our knowledge, this is the first time to analyze the operation cost of two-stage ozonation + UF + RO combination sequence with engineering data.

2. Materials and Methods

2.1. Setup and Operational Details of the Combined Systems

The schematics of the water reuse treatment processes are shown in Figure 1. Pretreatment and biological treatment techniques have been introduced in [3]. Therefore, the treatment technology of recycled water from textile wastewater was introduced in this study after biochemical treatment.



Figure 1. Schematics of the water reuse treatment processes.

2.1.1. Reactive Precipitation + Primary Sand Filtration + Primary Ozonation + Second Sand Filtration

As a type of strong oxidant, ozone exhibits the following significant characteristics: (1) in addition to its decolorization, deodorization, and removal of organic matters, ozone also shows outstanding sterilizing effect; (2) ozone produces no sludge; and (3) ozone can be prepared on site to avoid storage and transportation problems. However, ozone consumption must be reduced to save the reuse cost of recycled water because ozone preparation consumes considerable electricity resources. Suspended matter, colloid, color, turbidity, and organic matter in water shall be eliminated partially to reduce follow-up ozone consumption. The biochemical effluent was first adsorbed and precipitated in the reaction tank and tube settler and then filtered by the primary sand filtration tank to remove part of residual pollutants (e.g., SS and COD_{cr}). The processed water was subsequently decolorized in the primary ozone aeration tank. Effluents of the primary ozone aeration tank were sent to the clean-water reservoir after being filtered by the secondary sand filtration tank. The second sand filtration tank mainly aimed to eliminate the suspended matters, which generate in the ozone aeration, and some residues falling from the ozone aeration tank.

The hydraulic retention time of processing water in the tube settler was 6.4 h. In the sand filtration tank, wastewater was uniformly distributed to layers of filtering materials by using the water distributor. The filtering surfaces in the primary and secondary sand filters were approximately 450 m^2 and 700 m^2 , respectively. The proposed treatment technology aimed to reuse recycled water and thus required higher ozone dosage than the methods previously described [3]. The ozone dosage and oxidation time were approximately $80\text{--}100 \text{ g}\cdot\text{m}^{-3}$ and 3.20--3.29 h, respectively. The hydraulic retention time of effluent was 0.98 h. The sand filtration tank was cleaned regularly through backwash assisted with air scrubbing. The backwash water in the primary sand filtration tank and tube settler was precipitated in precipitation tank III to eliminate sludge and was then sent to the primary sand filtration tank I + precipitation tank I + sand filtration tank I and then flowed to the clean water reservoir. The sand filtration tank was cleaned for 0.5 h every 8 h. Water consumption in the primary and secondary sand filtration tanks was approximately $1500 \text{ m}^3 \cdot \text{d}^{-1}$. Polymeric aluminum (PAC) was used as the flocculants in the reaction tank and reaction tank I.

2.1.2. Primary UF System

In order to remove part of the turbidity and COD_{cr} and then protect the follow-up RO system, primary UF system was set. The procedure involved water production in the clean water reservoir \rightarrow primary self-cleaning filters \rightarrow feed pumps \rightarrow primary UF devices \rightarrow primary UF tank. The self-cleaning filters were used to purify water and protect normal operation of other equipment in the system. The filters intercepted impurities (e.g., suspended matters and particles) in the water by sieves, thus reducing turbidity and decreasing the productions of dirt and bacteria and the corrosion of the system. The filtering accuracy of the primary self-cleaning filters was approximately 100 µm. The UF membranes were composed of compressive modified polypropylene hollow fiber (PP) membrane. The operation pressure of primary UF was lower than 0.15 MPa. The UF device applied the full-automatic continuous operation mode composed of compressive dead-end filtering and gas-water backwash. Intercepted materials were eliminated through backwash or chemically enhanced backwash (CEB). The UF device performed 2 min of backwash every 1 h of the operation. Pollutants that could not be eliminated by backwash required online CEB at an interval of 24 h. Backwash water in the primary UF system entered the RO tank after being treated in the reaction tank II, precipitation tank II, and sand filtration tank II. The primary UF system provided approximately 350 m³·d⁻¹ of backwash water. PAC was also used as the flocculants in the reaction tank II.

The primary RO system was mainly composed of booster pumps, security filters, high-pressure pumps, RO membrane elements, wash-water pumps, valves, pressure pipes, and meters. The procedure included the primary UF tank \rightarrow RO booster pumps \rightarrow primary security filters \rightarrow high-pressure pumps \rightarrow primary RO devices \rightarrow primary RO tank. The security filters aimed to prevent the blockage of the RO membrane by small particulate impurities and to maintain RO's stable water yield. PP filter bags with a pore size of 5 µm were used in the security filter. Each RO device was equipped with one security filter. The operating pressure of the primary RO membrane was approximately 1.0–1.2 MPa. One backwash pump was installed in the RO system for regular backwash of the RO membrane, and the RO device was backwashed for 5 min after 3 h of operation. Some pollutants (e.g., organic matters and inorganic salt scaling) that were difficult to be washed away were cleaned by chemicals. Water produced by the primary RO system entered the RO tank and was then reused in enterprises in the industrial park. The primary RO concentrated water was processed in the secondary ozone aeration tank and then treated in the third sand filter + secondary UF + secondary RO system. The primary RO backwash water of approximately 130 m³·d⁻¹ ran through the reaction tank II + precipitation tank II + sand filtration tank II, and finally entered into the RO tank.

2.1.4. Secondary Ozone Aeration Tank

In order to increase the reuse rate of recycled water, a secondary water reuse system, which included secondary ozone aeration tank, third sand filtration tank, secondary UF system and secondary RO system, was set to treat the primary RO concentrated water. So, a secondary ozone aeration tank was set behind the primary RO system. The reaction time and ozone dosage in this secondary ozone aeration tank were 1.5 h and 100–120 g·m⁻³ of wastewater, respectively.

2.1.5. Third Sand Filtration Tank

The third sand filtration tank mainly aimed to eliminate suspended matters and some residues falling from the tank during ozone aeration to maintain the stability of follow-up procedures. Backwash water of approximately 750 m³·d⁻¹ in the third sand filtration tank was precipitated in the precipitation tank IV and then sent to the biochemical system. The filtering surface in the third sand filtration tank was 280 m².

2.1.6. Secondary UF System

Effluent from the third sand filtration tank first entered into the secondary UF raw water tank, was filtered in the secondary self-cleaning filters, and finally entered into the secondary UF films and then the secondary UF tank. The filtering accuracy of the secondary self-cleaning filters was approximately 100 μ m. Backwash water of approximately 130 m³·d⁻¹ from the secondary UF tank entered into the biochemical system.

2.1.7. Secondary RO System

Water in the secondary UF tank ran through the RO booster pumps, filtered by the secondary security filters, and successively entered the secondary RO membranes through high-pressure pumps. The filtering accuracy of filter bags in the secondary security filters was approximately 5 μ m, and the operating pressure of the secondary RO membranes was approximately 1.5–2.5 MPa. Treated water in the secondary RO system entered the secondary RO tank, whereas the concentrated water and wash water entered the biochemical system. The wash water volume in the secondary RO system was approximately 350 m³·d⁻¹.

2.2. Sewage Quality

A sampling program was conducted in a selected textile company with a production capacity of 75,000 t \cdot d⁻¹ located at Foshan, China. The sewage parameters and corresponding standards are listed in Table 1. The COD_{cr} in the effluent of biological treatment was similar to that previously described [3], but the color value was slightly higher due to the increased color in the raw water.

Table 1. Pollutant contents of influents from a textile wastewater treatment plant and corresponding standards (COD_{cr}: chemical oxygen consumption; BOD₅: biochemical oxygen demand; TP: total phosphorus; TN: total nitrogen; SS: suspended solid).

Parameter	Unit	Raw Wastewater Value	Wastewater after Biological Treatment	Discharge Standard [34]	Reuse Standard [35]	Drinking Water Standard [36]
pH	-	8.96-12.50	7.7-8.16	6–9	6.5–8.5	6.5-8.5
COD _{cr}	$mg \cdot L^{-1}$	400-1000	70.2-108	80	60	3
BOD ₅	$mg \cdot L^{-1}$	200-450	1–9	20	10	None
Color	times	400-1500	110-250	50	30	15
NH ₃ -N	$mg \cdot L^{-1}$	4–21	0.21-3.64	10	10	0.5
TP	$mg \cdot L^{-1}$	1-20	1.97-6.34	0.5	1	None
TN	$mg \cdot L^{-1}$	7–46	2.14-8.88	15	None	None
SS	$mg \cdot L^{-1}$	50-250	12.9-49	50	30	None
Turbidity	ŇTU	Uncertainty	0.42-1.33	None	5	1
Hardness	$mg \cdot L^{-1}$	Uncertainty	82—190	None	450	450
Conductivity	$\mu s \cdot cm^{-1}$	Uncertainty	5098—7410	None	None	None
Total alkalinity	$mg \cdot L^{-1}$	Uncertainty	323—696	None	350	None
Cl-	$mg \cdot L^{-1}$	Uncertainty	820—1496	None	250	250
SO_4^{2-}	$mg \cdot L^{-1}$	Uncertainty	1504-2498	None	250	250
Fe ³⁺	$mg \cdot L^{-1}$	Uncertainty	Uncertainty	None	0.3	0.3
Cu ²⁺	$mg \cdot L^{-1}$	Uncertainty	Uncertainty	None	None	1.0

2.3. Analytical Methods and Instruments

The pH was measured according to the glass-electrode method, using 420A + pH meter (Orion, Beijing, China). COD_{cr} analyses were performed by the fast digestion spectrophotometric method with DR890 chromometer (HACH, Shanghai, China). BOD₅ analyses were determined by the dilution and inoculation method with YSI5100 dissolved oxygen meter (YSI, Beijing, China). Color was measured according to the dilution times method. NH₃-N was performed by the Nessler's reagents spectrophotometric method with 722 spectrophotometer (Juchuang, Qingdao, China), while total phosphorus (TP) was performed by the ammonium molybdate spectrophotometry with 722 spectrophotometer. Total nitrogen (TN) was determined by the alkaline potassium persulfate digestion UV spectrophotometry with UV-1800 ultraviolet spectrophotometer (Aucy, Shanghai, China). SS was measured according to the gravimetric method, using electronic scales (Sanxinheng, Shenzhen, China). Turbidity, hardness, conductivity, total alkalinity and Cl⁻ were determined by turbidity meter (PTURB-202, Tiandishouhe, Beijing, China), hardness tester (CHCM-210, Changhong, Shenzhen, China), conductivity meter (GTCON-400C, GeneTest, Shanghai, China), total alkalinity meter (CHTA-211, Changhong, Shenzhen, China) and portable chlorometer (CLS-10A, Haiheng, Shanghai, China), respectively. SO_4^{2-} was performed by the gravimetric method with electronic scales. Fe³⁺ and Cu²⁺ were determined by the flame atomic absorption spectrometric method with flame atomic absorption photometer (AA320N, Shangfen, Shanghai, China).

2.4. Mathematical Model

The over standard rates were formulated using Equation (1):

$$E_i = (C_i - S_i) / S_i, \tag{1}$$

where E_i is the over standard rate for the *i*th parameter, C_i is the monitoring value of the *i*th parameter in each sample, and S_i is the standard value for the *i*th parameter in each sample. Each sample was determined in triplicate.

The percentage removal of the parameters evaluated was determined using the following equation:

$$\text{%removal} = \left(\frac{C_{i,in} - C_{i,out}}{C_{i,in}}\right) \times 100,\tag{2}$$

where $C_{i,in}$ and $C_{i,out}$ are the inlet and outlet concentration of the *i*th parameter. Each sample was determined in duplicate.

The unit operating cost of the reuse water can be calculated as

$$\cos t = \frac{TOC}{P},\tag{3}$$

where *TOC* is the total operating cost of the reuse water treatment process and *P* is the inlets of the primary RO tank and the secondary RO tank. The total operating cost *TOC* includes electricity cost of ozone and oxygen production, electricity cost of pumps, agentia cost, machine maintenance cost, PAC cost, cost of UF and RO membrane replacement, employee cost and cost of filter-bag replacement.

3. Results and Discussion

3.1. Performance of Individual Process for the Removal of Various Contaminants

Ozonation and sand filtration have been well practiced for water treatment—the former in the destruction of chemical and biological contaminants, and the latter in the removal of particulate matter [37]. Sand filtration is a good pretreatment for UF because it removes the organic foulants from the secondary effluent and significantly increases the UF water flux [38]. Therefore, wastewater was preprocessed by primary sand filte-ozone-secondary sand filter technique before membrane treatment. Moreover, PAC was added for the flocculation of wastewater and then precipitated before sand filtration to reduce ozone consumption. As shown in Figure 2, the removal rate of SS was the highest $(99.34 \pm 0.92\%)$, followed by color (74.01 \pm 8.68%), COD_{cr} (39.85 \pm 7.51%), NH₃-N (27.35 \pm 31.78%), TP (13.25 \pm 15.94%), and TN (-4.39 \pm 25.02%) after treatment by the reaction, precipitation, primary sand filtration, ozonation oxidation and secondary sand filtration. SS was mainly eliminated during sand filtration. In addition, turbidity can also be eliminated during sand filtration, and the average removal rate of turbidity in the secondary sand filtration was about $37.42 \pm 5.21\%$. The ozone dosage in the primary ozone aeration tank was approximately 90 g \cdot m⁻³ of wastewater. The average removal rates of COD_{cr}, color, NH₃-N, TP, and TN were $27.6 \pm 7.03\%$, $69.4 \pm 6.98\%$, $23.8 \pm 33.33\%$, $-18.05 \pm 19.30\%$, and $-5.63 \pm 28.54\%$, respectively, in the primary ozone aeration tank. The color removal rate was proportional to the ozone dosage (Figure 3). The relational expression between the ozone dosage x and color removal rate y was fitted with software CurveExpert 1.3 [39] (Equation (4): correlation coefficient = 0.9957).

$$y = 1.32 + 1.30 x - 1.30 \times 10^{-3} x^2 - 1.02 \times 10^{-4} x^3 + 5.46 \times 10^{-7} x^4$$
(4)

Figure 4 shows that the removal rates of NH₃-N, TP, and TN fluctuated violently during ozonation. The maximum removal rates of NH₃-N, TP, and TN were 87.99%, 28.65%, and 12.86%, respectively, whereas their minimum rates were -50.39%, -72.73%, and -64.40%, respectively. On the one hand, during the reaction between ozone and wastewater, either ozone oxidized some NH₃-N into NO₂-N [40], or the supply of ozone formed bubbles that drove NH₃-N spillover in the form of NH₃. On the other hand, ozone degraded some organic nitrides in wastewater into NH₃-N through oxidation [41]. The production of NH₃-N was accelerated when a high content of organic nitrides formed in the wastewater, thus increasing the concentration of NH₃-N in wastewater. Therefore,

the removal rate of NH₃-N was negative. The removal rate of TN fluctuated for two reasons. On the one hand, some organic nitrogen particles were oxidized into inorganic nitrogen or entered into liquid phase or spillover in the form of N₂/NH₃ [42]. On the other hand, some byproducts (e.g., NO₂) were produced during ozone preparation and were then dissolved in the wastewater [43,44]. Nevertheless, only a small proportion of NO₂ was produced during ozone oxidation [43]. The removal rates of NH₃-N and TN were mainly positive and negative, respectively. This finding indicates that the organic nitrogen in wastewater slowly oxidized into N₂/NH₃. Fluctuation of TP content in wastewater could be mainly interpreted from the following two aspects. First, soluble inorganic phosphorus that mainly exist in PO_4^{3-} combined with metal ions (e.g., Ca^{2+} , Mg^{2+} , and Fe^{3+}) and then formed sediments. Second, parts of macromolecular organic matter in wastewater were degraded into soluble phosphorus through ozonation [45].

Figure 5 shows that after all the treatments, the value of the wastewater attained 33–70.8 mg·L⁻¹ COD_{cr} (average of 52.02 mg·L⁻¹), 20–80 times color (average of 39 times), 0.15–2.02 mg·L⁻¹ NH₃-N (average of 0.86 mg·L⁻¹), 0–0.5 mg·L⁻¹ SS (average of 0.16 mg·L⁻¹), 2.73–11.9 mg·L⁻¹ TN (average of 6.00 mg·L⁻¹), and 1.47–5.68 mg·L⁻¹ TP (average of 3.08 mg·L⁻¹).



Figure 2. Removal rate of the pollutants during reactive precipitation + primary sand filtration + primary ozonation + second sand filtration.



Figure 3. Effect of ozone dosage on the color removal rate (initial color at 125 times).



Figure 4. Removal rate of the NH₃-N, TP, and TN in primary ozonation.



Figure 5. Pollutants of the effluent after treatment by the reaction, precipitation, primary sand filtration, primary ozone oxidation, and secondary sand filtration (**a**) chemical oxygen consumption (COD_{cr}) and color, (**b**) suspended solid (SS), NH₃-N, TN and TP.

UF retains suspended particles and macromolecules. The major constituents of effluent organic matter are biopolymers, mainly soluble microbial products [46]. Therefore, UF can eliminate COD_{cr} and TP to some extent but performs poorly in removing color, ammonia nitrogen, and TN [24] (Figure 6a,b). UF could effectively reduce turbidity but hardly influence hardness, total alkalinity, Cl⁻, SO_4^{2-} , and conductivity (Figure 6c,d). After UF treatment, the respective removal rates of COD_{cr} , TP, and turbidity were $17.34 \pm 5.86\%$, $4.03 \pm 3.13\%$, and $52.18 \pm 18.07\%$ (Figure 7), which were lower than previously described [24]. This finding reflected that the contents of suspended matter and macromolecular organics in wastewater might be relatively low after sand filtration + ozonation + sand filtration.

The primary RO pressure was controlled between 1.0–1.2 MPa to protect stable water yield. Pressure gradually increased upon the blockage of the RO membrane by dirt. Water yield and desalinization rate of primary RO treatment slightly changed with the increase in inlet pressure (Table 2). The average removal rates of COD_{cr} , color, TP, CI^- , SO_4^{2-} , and conductivity in the primary RO were 99.17 \pm 0.88%, 99.11 \pm 1.60%, 98.09 \pm 1.55%, 92.02 \pm 1.87%, 99.17 \pm 0.81%, and 92.94 \pm 2.49%, respectively. The average removal rates of total alkalinity, NH₃-N, TN, hardness, and turbidity were 87.74 \pm 2.64%, 91.39 \pm 2.45%, 85.07 \pm 6.69%, 82.83 \pm 5.19%, and 76.98 \pm 5.46%, respectively (Figure 8). The removal rate of conductivity was lower than that previously described [47], which might have occurred because an old RO membrane was used in the primary RO system. This RO membrane had been used for 2 years. Contents of different pollutants in effluent of primary RO treatment are

shown in Table 3. COD_{cr} , BOD_5 , SS, color, Fe^{3+} , and Cu^{2+} were generally undetectable in RO effluent. All water quality indexes of RO effluent satisfied the reuse standards of recycled water. COD_{cr} , color, NH₃-N, hardness, Cl^- , SO_4^{2-} , turbidity, and contents of Fe^{3+} and Cu^{2+} all satisfied the standards of domestic drinking water.



Figure 6. Cont.



Figure 6. Pollutant changes in the primary ultrafiltration (UF) (**a**) COD_{cr} , color, (**b**) NH_3 -N, TN, and TP, (**c**) conductivity, SO_4^{2-} , Cl^- , and (**d**) hardness, total alkalinity, turbidity.



Figure 7. Removal rate of TP, turbidity, and COD_{cr} in the primary UF process.

Table 2. Operating conditions and results from the test carried by the primary reverse osmosis (RO).



Figure 8. Removal rate of pollutants in the primary RO process.

	Unit	Average	Max	Min
pН	-	7.90	8.15	7.74
COD _{cr}	$mg \cdot L^{-1}$	0.37	1.65	0.00
Color	times	0.33	< 5.00	0.00
NH ₃ -N	$mg \cdot L^{-1}$	0.07	0.15	0.01
TN	$mg \cdot L^{-1}$	0.91	2.26	0.10
TP	$mg \cdot L^{-1}$	0.06	0.29	0.00
Hardness	$mg \cdot L^{-1}$	22.04	50.78	8.10
Total alkalinity	$mg \cdot L^{-1}$	64.19	104.90	31.24
Cl-	$mg \cdot L^{-1}$	92.09	148.90	46.60
SO_4^{2-}	$mg \cdot L^{-1}$	15.23	70.62	0.19
Conductivity	$\mu s \cdot cm^{-1}$	415.84	712.69	127.94
Turbidity	NTU	0.06	0.14	0.01
BOD ₅	$mg \cdot L^{-1}$	Not detectable	Not detectable	Not detectable
SS	$mg \cdot L^{-1}$	Not detectable	Not detectable	Not detectable
Fe ³⁺	$mg \cdot L^{-1}$	Not detectable	Not detectable	Not detectable
Cu ²⁺	$mg \cdot L^{-1}$	Not detectable	Not detectable	Not detectable

Table 3. Pollutants of the effluent after the primary RO treatment.

To further increase the reuse rate of recycled water, we continuously purified the RO-concentrated water. Given the high contents of pollutants (COD_{cr} and color) in the RO-concentrated water, ozone dosage was adjusted to approximately 118 g·m⁻³ of wastewater to relieve membrane blockage in the future. Contents of pollutants fluctuated slightly before and after ozone was added, indicating that ozone could hardly affect hardness, total alkalinity, Cl⁻, SO₄²⁻, and conductivity (Figure 9). The removal rates of COD_{cr} and color were higher than those in the primary ozone aeration tank. The average removal rates of COD_{cr} and color in the secondary ozone aeration tank were $35.72 \pm 6.26\%$ and $75.36 \pm 7.39\%$, respectively (Figure 10). The removal rates of COD and color in the primary and secondary ozone aeration tanks were all lower than those of electrocoagulation-O₃ process, during which the ozone dose was 280 g·m^{-3} and the color removal rate was close to 100% [48].





Figure 9. Cont.



Figure 9. Pollutants change in the second ozonation process (**a**) conductivity and turbidity, (**b**) hardness, total alkalinity, Cl^- , SO_4^{2-} .



Figure 10. Removal rate of the COD_{cr} and color in the second ozonation process.

RO concentration water was further treated by sand filtration, UF, and RO after ozonation. Given that the conductivity of RO concentrated water was very high (average of 28,236 μ s·cm⁻¹, Figure 10), inlet pressure of the secondary RO system was 1.5 MPa. The average removal rates of pollutants are shown in Figure 11. The average removal rates of COD_{cr}, color, NH₃-N, TN, TP, hardness, total alkalinity, Cl⁻, SO₄²⁻, conductivity, and turbidity were 99.41 ± 0.63%, 99.01 ± 1.65%, 93.52 ± 2.08%, 89.11 ± 4.95%, 99.00 ± 0.79%, 98.89 ± 1.18%, 94.77 ± 2.64%, 96.34 ± 1.29%, 99.48 ± 0.41%, 97.99 ± 1.31%, and 90.17 ± 4.41%, respectively. Contents of different pollutants in effluent of the second RO treatment are listed in Table 4. Although conductivity, total alkalinity, hardness, and contents of Cl⁻ and SO₄²⁻ in inflow were relatively high, the water quality indexes of the secondary RO effluent were no poorer than those of the primary RO effluent due to the use of

a new RO membrane and high inlet pressure. Water quality indexes of the secondary RO effluent all satisfied the reuse standards of recycled water. Color, COD_{cr} , hardness, and contents of Cl^- and SO_4^{2-} all satisfied the standards of domestic drinking water.



Figure 11. Average removal rates of pollutants for the RO concentrated water by the third sand filtration + second UF + second RO.

	Unit	Average	Max	Min
pH	-	7.90	8.15	7.74
COD _{cr}	$mg \cdot L^{-1}$	0.44	1.75	Not detectable
Color	times	0.32	5.00	Not detectable
NH ₃ -N	$mg \cdot L^{-1}$	0.06	0.19	Not detectable
TN	$mg \cdot L^{-1}$	1.57	6.75	0.12
TP	$mg \cdot L^{-1}$	0.08	0.27	Not detectable
Hardness	$mg \cdot L^{-1}$	3.45	18.22	0.16
Total alkalinity	$mg \cdot L^{-1}$	67.31	154.95	7.26
Cl ⁻	$mg \cdot L^{-1}$	107.75	246.54	31.30
SO_4^{2-}	$mg \cdot L^{-1}$	25.03	66.04	0.5
Conductivity	$\mu s \cdot cm^{-1}$	288.60	941.95	89.64
Turbidity	NTU	0.09	0.26	Not detectable
BOD ₅	$mg \cdot L^{-1}$	Not detectable	Not detectable	Not detectable
SS	$mg \cdot L^{-1}$	Not detectable	Not detectable	Not detectable
Fe ³⁺	$mg \cdot L^{-1}$	Not detectable	Not detectable	Not detectable
Cu ²⁺	$mg \cdot L^{-1}$	Not detectable	Not detectable	Not detectable

Table 4. Pollutants of the effluent after the second RO treatment.

3.2. Overall Efficiency

The combined processes showed good performance in water reuse treatment. After treatment, the overall standard rates of the COD_{cr} , color, NH_3 -N, hardness, Cl^- , SO_4^{2-} , turbidity, Fe^{3+} , and Cu^{2+} were -0.88, -0.98, -0.86, -0.95, -0.63, -0.94, -1 and -1, respectively, for the output water of the primary RO system and -0.85, -0.98, -0.88, -0.99, -0.57, -0.90, -0.91, -1, and -1, respectively, for the output water of the second RO system compared with the drinking water standard. The overall removal rate of COD_{cr} , color, NH_3 -N, TN, TP, hardness, total alkalinity, Cl^- , SO_4^{2-} , conductivity, and turbidity reached 99.57%, 99.79%, 94.65%, 84.53%, 98.36%, 82.54%, 87.68%, 92.05%, 99.17%, 93.03%, and 93.18%, respectively, for the primary water reuse system and 99.62%, 99.75%, 94.75%, 88.26%, 99.12%, 98.90%, 94.83%, 96.40%, 99.48%, 97.95%, and 88.64%, respectively, for the second water reuse system (Table 5). The RO system achieved the highest removal rate of pollutants. However, color was decreased dramatically during ozonation (92.59 and 97.27 times). The removal rate of the conductivity with new RO membranes was higher than those during UF-electrodialysis process [22] and UF ceramic membrane [21], and close to that during UF-diafiltration process [19].

	Treatment		Reaction + Sand Filtration	Ozonation	Sand Filtration	UF	RO	Total
	COD _{cr}	$(mg \cdot L^{-1})$	9.26 (10.7%)	21.33 (27.6%)	3.91 (7.00%)	9.03 (17.34%)	42.64 (99.17%)	86.18 (99.57%)
	Color	(times)	22.26 (14.3%)	92.59 (69.4%)	0.41 (1.00%)		38.77 (99.11%)	155.34 (99.79%)
	SS	$(mg \cdot L^{-1})$		24.2 (99.34%)			Not detectable	24.39 (100%)
	NH ₃ -N	$(mg \cdot L^{-1})$		0.5 (27.35%)			0.77 (91.39%)	1.27 (94.65%)
	TN	$(mg \cdot L^{-1})$		-0.1 (-4.39%)			4.49 (85.07%)	4.99 (84.53%)
Primary water	TP	$(mg \cdot L^{-1})$		0.5 (13.25%)		0.12 (4.03%)	2.90 (98.09%)	3.50 (98.36%)
reuse system	Hardness	$mg \cdot L^{-1}$)		Uncertainty			103.91 (82.83%)	104.23 (82.54%)
	Total alkalinity	$(mg \cdot L^{-1})$		Uncertainty			456.67 (87.74%)	456.71 (87.68%)
	Cl-	$(mg \cdot L^{-1})$		Uncertainty			1063 (92.02%)	1067 (92.05%)
	SO_4^{2-}	$(mg \cdot L^{-1})$		Uncertainty			1816 (99.17%)	1829 (99.17%)
	Conductivity	$(\mu s \cdot cm^{-1})$		Uncertainty			5491 (92.94%)	5550 (93.03%)
_	Turbidity	NTU	Uncert	tainty	0.33 (37.42%)	0.28 (52.18%)	0.21 (76.98%)	0.82 (93.18%)
	COD _{cr}	$(mg \cdot L^{-1})$	None	41.15 (35.72%)		73.94 (99.41%)		115.09 (99.62%)
	Color	(times)	None	97.27 (75.36%)		32.58 (99.02%)		129.85 (99.75%)
	NH ₃ -N	$(mg \cdot L^{-1})$	None			0.83 (93.68%)		1.01 (94.75%)
Second water reuse system	TN	$(mg \cdot L^{-1})$	None			12.16 (88.57%)		11.80 (88.26%)
	TP	$(mg \cdot L^{-1})$	None			7.97 (99.06%)		8.50 (99.12%)
	Hardness	$(mg \cdot L^{-1})$	None			308.22 (98.89%)		309.87 (98.90%)
	Total alkalinity	$(mg \cdot L^{-1})$	None			1212.21 (94.74%)		1234.45 (94.83%)
	Cl ⁻	$(mg \cdot L^{-1})$	None			2838 (96.34%)		2888 (96.40%)
	SO_4^{2-}	$(mg \cdot L^{-1})$	None			4743 (99.48%)		4775 (99.48%)
	Turbidity	NTU	None			0.68 (88.61%)		0.69 (88.64%)
	Conductivity	$(\mu s \cdot cm^{-1})$	None			13,845 (97.96%)		13,813 (97.95%)

Table 5. Summary of results of combined process as indicated by average drop and removal rate.

Note: '--' means that the removal rate is very small.

3.3. Energy and Chemicals Costs

Data on energy and chemicals costs for the water reuse treatment at a flow rate of $7.5 \times 10^4 \text{ m}^3 \cdot \text{d}^{-1}$ (working time 24 h day⁻¹) are summarized in this section. Water yields in the primary RO system and secondary RO system were 50,250 and 14,850 m³ · d⁻¹, respectively. The reuse rate of recycled water in the whole system was 86.8%, higher than that during forward osmosis (FO)-RO system by 31.8% [49].

For the primary ozone reaction tank, ~90 g $O_3 \cdot m^{-3}$ wastewater was consumed on average, whereas approximately 118 g $O_3 \cdot m^{-3}$ wastewater was consumed for the second ozone reaction tank. The electricity consumption of the ozone and oxygen production was 12 kWh·kg⁻¹ O_3 power [3]. Thus, an operating cost of approximately 0.11 USD·m⁻³ wastewater (approximately 8100 USD·d⁻¹ for 7.5×10^4 m³·d⁻¹ wastewater) was calculated for a power price of 0.1 USD·kWh⁻¹ for the primary zone reaction tank and $0.14 \text{ USD} \cdot \text{m}^{-3}$ wastewater (approximately 3505 USD·d⁻¹ for 24,750 m³·d⁻¹ wastewater) for the second ozone reaction. Powers of inlet pumps in the primary UF and primary RO system were 340 kW and 2155 kW, respectively. The total power consumption of primary self-cleaning filter, sand filter, UF, and RO backwash pump was approximately 17 kW. Then, the power consumption of the pumps in the primary water reuse system was $60,288 \text{ kWh} \cdot \text{d}^{-1}$, and the electricity cost of the pumps in the primary water reuse system was 6029 USD \cdot d⁻¹. The total electricity cost in the primary water reuse system was 14,129 USD \cdot d⁻¹. The power consumptions of inlet pumps in the secondary UF and secondary RO system were 110 and 770 kW, respectively. The total power consumption of secondary self-cleaning filter, sand filter, UF, and RO backwash pump was approximately 11 kW. Then, the power consumption of the pumps in the second water reuse system was $21,384 \text{ kWh} \cdot \text{d}^{-1}$, and the electricity cost of the pumps in the second water reuse system was $2138 \text{ USD} \cdot d^{-1}$. The total electricity cost in the second water reuse system was $5643 \text{ USD} \cdot d^{-1}$.

The PAC dosage was approximately 80 g·m⁻³ of wastewater and thus the PAC cost was approximately 644 USD·d⁻¹. The number of RO membrane and UF membrane were 5280 and 1224, respectively, and the prices of them were 500 and 2167 USD, respectively. So the costs of UF and RO membranes replacement were 53.04×10^4 and 88×10^4 USD·a⁻¹. The number of workers in the sewage plant were 50, and the annual salary was 25,000 USD·person⁻¹. So the employee cost was 125×10^4 USD·a⁻¹. The Machine maintenance cost was approximately 21.91×10^4 USD·a⁻¹. The cost of the agentia and the filter-bag were 11.42×10^4 and 3.96×10^4 USD·a⁻¹, respectively.

The total operating cost of treating textile wastewater is displayed in Table 6, reaching $0.44 \text{ USD} \cdot \text{m}^{-3}$ reuse water, wherein the cost of the ozone production was $0.18 \text{ USD} \cdot \text{m}^{-3}$ (40%), that of sand filtration, UF and RO system was $0.19 \text{ USD} \cdot \text{m}^{-3}$ (43%), and that of machine maintenance, employee cost, agentia, and PAC was $0.08 \text{ USD} \cdot \text{m}^{-3}$ (17%) (Figure 12). In the filtering system, RO claimed the highest cost ($0.14 \text{ USD} \cdot \text{m}^{-3}$ reuse water), followed by UF ($0.04 \text{ USD} \cdot \text{m}^{-3}$). In RO system, the electricity cost and membrane cost were $0.11 \text{ and } 0.04 \text{ USD} \cdot \text{m}^{-3}$, respectively. The operating cost of secondary reuse system was approximately $0.086 \text{ USD} \cdot \text{m}^{-3}$ higher than that in the primary system, which was caused by the increased ozone dosage and operating pressure of RO. Based on the above analysis, the electricity cost for ozonation and RO treatment accounted for 64.84% of the total cost. Therefore, the key to reducing the operating cost of the proposed sequential system in this research was lower than that of FO-RO system in literature [49] by $0.06 \text{ USD} \cdot \text{m}^{-3}$ reuse water. It was also much lower than that of electrocoagulation-O₃ process, which was $5.80 \text{ USD} \cdot \text{m}^{-3}$ treated wastewater [48].

	Unit	Data
Electric rate (USD·kWh ⁻¹)		0.1
Primary water reuse system	Reclaimed water $(m^3 \cdot d^{-1})$ Power consumption for ozone and oxygen production (kWh·d ⁻¹) Power consumption of the pumps (kWh·d ⁻¹) Electricity cost (USD·d ⁻¹)	
	Polymeric aluminum (PAC) (kg·d ^{-1}) PAC cost (USD·d ^{-1})	6098 644
Second water reuse system	Reclaimed water $(m^3 \cdot d^{-1})$ Power consumption for ozone production (kWh·d ⁻¹) Power consumption of the pumps (kWh·d ⁻¹) Electricity cost (USD·d ⁻¹)	14,850 35,046 21,384 5643
Agentia	Anti-scalant (kg·d ⁻¹) Reducing agents (kg·d ⁻¹) Sterilizing agent (kg·d ⁻¹)	114 114 6.65
Agentia cost $(10^4 \text{ USD} \cdot a^{-1})$ Machine maintenance cost $(10^4 \text{ USD} \cdot a^{-1})$ Ultrafiltration (UF) membrane $(10^4 \text{ USD} \cdot a^{-1})$ RO membrane $(10^4 \text{ USD} \cdot a^{-1})$ Filter-bag of the precision filter $(10^4 \text{ USD} \cdot a^{-1})$ Employees cost $(10^4 \text{ USD} \cdot a^{-1})$ Unit water reuse treatment cost $(\text{USD} \cdot \text{m}^{-3})$		

Table 6. Operating cost of the combined process.

Notes: (1) UF membrane was replaced every 5 years, RO membrane was replaced every 3 years, and security filter bag was replaced every 10 years. (2) The cost for filter-bag of the precision filter was $0.0017 \text{ USD} \cdot \text{m}^{-3}$ produced water. (3) Working days in one year were 365 days.



Figure 12. Cost breakdown analysis of water reuse treatment.

4. Conclusions

A combined two-stage water reuse treatment was developed to obtain quality water for reuse in industrial processes. The elimination effect and operation cost of the combined two-stage water reuse treatment were investigated.

SS is mainly eliminated in the sand filtration process. Ozone can thoroughly eliminate color. The removal rates of the primary and secondary ozone aeration tanks were $69.4 \pm 6.98\%$ and $75.36 \pm 7.39\%$, respectively. The removal rate of color is proportional to ozone dosage. UF mainly eliminates COD_{cr}, TP, and turbidity. RO treatment shows the highest removal rate. The removal rates

of COD_{cr} , color, and SO_4^{2-} by RO treatment were higher than 99%. The removal rate of the secondary reuse system of recycled water is higher than that of the primary system due to the high ozone dosage and usage of a new RO membrane.

The proposed sequential system is extremely effective. Effluents of the primary and secondary recycled water processing systems satisfy the reuse standards of recycled water. COD_{cr} , color, NH₃-N, hardness, Cl⁻, SO₄²⁻, turbidity, Fe³⁺, and Cu²⁺ satisfy the standards of domestic drinking water. The removal rate of the conductivity with new RO membrane was higher than those during UF-electrodialysis process and UF ceramic membrane, and close to that during UF-diafiltration process. The reuse rate of recycled water in the whole system was 86.8%, higher than that during FO-RO system by 31.8%.

The total operating cost of treating textile wastewater reached 0.44 USD m⁻³ reuse water, wherein the cost of sand filtration, UF and RO system was maximum (approximately 0.19 USD·m⁻³), followed by that of the ozone production (approximately 0.18 USD·m⁻³). In the filtration system, RO claimed the highest cost (0.14 USD·m⁻³ reuse water), followed by UF (0.04 USD·m⁻³). In RO system, the electricity cost and membrane cost were 0.11 and 0.04 USD·m⁻³, respectively. The operating cost of secondary reuse system was approximately 0.086 USD·m⁻³ higher than that of the primary system. Electricity cost for ozonation and RO treatment accounted for 64.84% of the total cost.

The operating cost of the proposed sequential system in this research was lower than that of FO-RO system by $0.06 \text{ USD} \cdot \text{m}^{-3}$ reuse water. Although the color removal rate of the electrocoagulation-O₃ process was close to 100%, the operating cost of it was much higher than that in this research, 5.80 USD·m⁻³ treated wastewater.

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Abbreviations

E_i	over standard rate for <i>i</i> th parameter
C _i	monitoring value of <i>i</i> th parameter, (mg·L ⁻¹ , times, μ s·cm ⁻¹ , NTU)
S _i	standard value for <i>i</i> th parameter, (mg·L ^{-1} , times, μ s·cm ^{-1} , NTU)
$C_{i,in}, C_{i,out}$	inlet and outlet concentration, (mg·L ^{-1} , times, μ s·cm ^{-1} , NTU)
$\cos t$	unit operating cost of the reuse water, (USD \cdot m ^{-3})
TOC	total operating cost, (USD· d^{-1})
Р	output of reuse water, $(m^3 \cdot d^{-1})$

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