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Characteristics, Environmental Impact, and Treatment of Reverse Osmosis Concentrate Generated from Municipal and Industrial Wastewater: A Review and Futuristic Outlook

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Keywords:
Complete recirculation; Integrated technology; Partial recirculation; Reverse osmosis concentrate; Zero liquid discharge.

Highlights:

- *ROC generated from municipal and industrial wastewater is reviewed.*
- *Integrated technologies with complete and partial recirculation are overviewed.*
- *Integrated technologies are strategic solutions for improving the quality of ROC.*
- *Integrated technologies are strategic solutions for minimizing the quantity of ROC.*
- *Advanced oxidation processes are suggested for treating ROC for future research.*

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Abstract: Reusing wastewater from municipal and industrial resources has been a worldwide strategic solution to water scarcity. Reverse osmosis (RO), a well-established technology, is widely applied for wastewater treatment, producing high-quality reuse wastewater effluent. However, one of the major drawbacks of using RO technology is the volume of concentrate (known as ROC) associated with higher concentrations of constituents in wastewater feed. This drawback makes the sustainable management of ROC in terms of quality and quantity the major limitation of RO application. To address this drawback, the present review highlights and discusses the characteristics and environmental impact of ROC from municipal and industrial wastewaters, facilitating easy selection of the best applicable integrated technologies based on the concept of zero liquid discharge (ZLD) for minimizing the ROC volume produced. To achieve this objective, this paper provides an overview of various types of integrated technologies with two modes of operation (complete and partial recirculation). This paper offers critical insights into the ZLD concept and highlights future research trends by suggesting various pretreatment options for ROC. These suggestions will improve the overall recovery of water feed and minimize water pollution to meet the environmental standards for final disposal.

الخصائص والتأثير البيئي ومعالجة المياه المرفوضة بفعل التناضح العكسي من المياه العادمة البلدية والصناعية: مراجعة ونظرة مستقبلية

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الخلاصة
تعد عملية إعادة استخدام المياه العادمة الناجمة عن المصادر البلدية والصناعية هي احدى الحلول الاستراتيجية في جميع انحاء العالم عندما يتم التعامل مع قضية شحة المياه. اذ يلعب التناضح العكسي، وهو تقنية رائدة عالمياً، دوراً مهماً في معالجة المياه العادمة من حيث قابليته على انتاج نوعية مياه مطلقة عالية الجودة. وبالرغم من هذه الفائدة، الا تقنية التناضح العكسي تواجه عائق كبير من حيث حجم المياه المهدورة والتي تسمى المياه المركزة (المرفوضة بفعل أداء الفصل الغشائي) والتركيز العالي للملوثات المتواجدة فيها. هذا العائق يجعل الإدارة المستدامة للمياه المرفوضة من حيث النوعية والكمية من أكبر التحديات في تطبيق تقنية التناضح العكسي من الناحية العملية. ومن اجل معالجة هذا العائق او التحدي، يسلط بحث المراجعة الحالي الضوء ويناقش الخصائص والتأثيرات البيئية للمياه المرفوضة من معالجة المياه العادمة (البلدية والصناعية)، لتسهيل عملية اختيار أفضل التقنيات المدمجة القابلة للتطبيق بالاعتماد على مفهوم التصريف الصفري للسوائل وذلك لتقليل حجم هذه المياه. ومن اجل تحقيق هذا الهدف، فان بحث المراجعة يقدم رؤى نقدية حول هذا المفهوم ويسلط الضوء على الاتجاهات المستقبلية للبحوث عن طريق اقتراح خيارات المعالجة التمهيدية لها. هذه المقترحات سوف تعمل على تحسين عملية استرداد المياه بشكل كبير وتقليل التلوث المتواجد فيها لتتوافق مع المعايير البيئية اللازمة للتصريف النهائي لها.

الكلمات الدالة: التدوير الكامل، التقنيات المدمجة، التدوير الجزئي، المياه المرفوضة، التصريف الصفري للسوائل.

1.INTRODUCTION

Several significant factors have been severely limiting the natural resources of fresh water, such as growing populations, increasing urbanization, developing industrialization, environmental pollution, and climate change [1–4]. Municipal and industrial wastewater are considered worldwide as a strategic solution to conserve limited freshwater resources [5–10]. However, these wastewaters contain considerable amounts of pollutants [11–13]. Their discharge into water bodies without the proper treatment would negatively affect the aquatic environment and human health [14–16]. To overcome this issue, wastewater treatment plants (WWTPs) are used for treating these wastewaters using several processes and technologies [17–19]. However, conventional processes and technologies in these WWTPs cannot produce treated wastewater effluents that meet water quality regulations before discharging into natural water bodies [20, 21]. To achieve this objective, membrane technology has been increasingly implemented recently to treat secondary effluents of municipal and industrial wastewater, producing a high-quality treated wastewater effluent that satisfies the water quality criteria of WHO guidelines [22–25]. This technology uses a semi-permeable membrane that separates the feed of treated wastewater (coming from the final stage of wastewater treatment) into two streams. The first stream is termed permeate, which passes through the membrane, while the second stream is termed concentrate, retained by the membrane [26, 27]. This membrane technology based on pressure-driven processes is typically classified into microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) [28]. The three types of processes (MF, UF, and NF) are generally applied as a pretreatment step to decrease fouling before applying the RO process. The RO process is widely applied for treating municipal and industrial wastewater for its highly efficient performance in rejecting a wide range of pollutants (organic, inorganic, heavy metals, monovalent ions, and microorganisms) [23, 24, 29–33]. Despite these advantages of applying the RO membranes, the RO membrane separation process produces a concentrate as a by-product, i.e., ROC [34, 35]. During this process, on one side, the volume of ROC produced is approximately 10–60% of the input feed [20, 36]. Disposing of these massive quantities of ROC into the environment causes a considerable loss of water resources. On the other side, this volume of ROC produced contains almost all the original constituents of wastewater feed at elevated concentrations (nearly 4–7 times more than the feed concentration) [37, 38]. Thus, the management of ROC in terms of quality and quantity remains the major limitation of applying the RO process [36, 39, 40]. Applying the concept of zero liquid discharge (ZLD) or near-ZLD in ROC is a strategic management used to recover most of the water from ROC as a product, i.e., permeate, for reuse purposes and treat the pollutants in ROC [41–43]. Therefore, to the best of the authors' knowledge, the present paper is the first comprehensive review to provide a complete profile of ROC generated from municipal and industrial wastewater treatment plants in terms of characteristics, environmental impact, and strategies for treating it (to meet environmental standards for the requirements of final disposal) and minimizing the inevitable volume produced based on the concept of ZLD or n-ZLD. Other types of ROCs generated from brackish and seawater desalination are beyond the scope of the present review.

2.CHARACTERISTICS OF ROC

The characteristics of ROC generally depend on several parameters, including the nature of feed used, type of membrane applied (i.e., RO membrane), type of pretreatment, and chemical materials used in this pretreatment [23, 34]. The present paper reviews the characteristics of two types of ROCs generated from municipal and industrial wastewater treatment plants.

2.1.ROC from Municipal Wastewater Treatment

The main characteristics of ROC produced from secondary-treated municipal wastewater effluents are similar to the original constituents of municipal wastewater feed (i.e., RO influent). These characteristics can be summarized in Table 1. Also, heavy metals, such as Cu, Cr, Ni, Fe, and Mo, are present in the ROC [23, 34, 44].

Table 1 Characteristics of ROC Obtained from Biologically Treated Secondary Effluent.

Parameter	Range	Unit	Reference
pH	6.7-8.7	-	[20–23, 39, 45]
TOC	5.6-47	mgL^{-1}	[20–23]
DOC	12-95	mgL^{-1}	[20, 22, 23, 39, 45]
COD	55-470	mgL^{-1}	[20, 22, 23, 39]
BOD_5	1.2-5	mgL^{-1}	[20]
TDS	39.9-16140	mgL^{-1}	[22, 23, 39, 45]
Conductivity	1.7-23000	μScm^{-1}	[20, 22, 23, 39, 45]
Alkalinity (as $CaCO_3$)	242-914	mgL^{-1}	[20, 22, 39]
Color	51.2-278	Pt.Co	[23, 39]
A_{254}	0.25-1.3	cm^{-1}	[20, 23, 39]
Mg^{2+}	7-236	mgL^{-1}	[20, 22, 45]
Na^{+}	203-1637	mgL^{-1}	[20, 22, 45]
K^{+}	22.6-135	mgL^{-1}	[20, 22, 45]
Ca^{2+}	5-306	mgL^{-1}	[22]
Fe^{2+}	0.1-0.3	mgL^{-1}	[22]
Mn^{2+}	54-230.5	mgL^{-1}	[22]
Cl^{-}	1.4-8060	mgL^{-1}	[20, 22, 39, 45]
NO_2^{-}	1.3-8.3	mgL^{-1}	[22, 45]
NO_3^{-}	23-296	mgL^{-1}	[20, 22, 45]
SO_4^{2-}	159.1-1759	mgL^{-1}	[20, 22, 45]
HCO_3^{-}	543-2056	mgL^{-1}	[20]
PO_4^{3-}	1-39	mgL^{-1}	[20, 22, 45]

TOC: total organic carbon, DOC: dissolved organic carbon, COD: chemical oxygen demand, BOD_5: 5-day biological oxygen demand, TDS: total dissolved solids, A_{254}: ultraviolet absorbance at the wavelength of 254nm.

2.2.ROC from Industrial Wastewater Treatment

The characteristics of secondary-treated industrial wastewater effluents in terms of composition vary significantly depending on the source of feed from various industrial activities and the specific treatment involved [44, 46]. Thus, the characteristics of ROC from these industrial wastewater effluents vary accordingly. Emerging contaminants (ECs), emerging pollutants (EPs), or micropollutants (MPs) are synthetic or natural chemicals in their origin. Most of these chemicals are organic in nature, present in traces ranging from parts per trillion (ppt) to parts per billion (ppb), and not commonly monitored in the environment [5, 25, 47]. There have been a wide range of these ECs based on the industrial category, such as industrial class (PFOA, PFOS, PBDEs, PCBs, and PAHs, Bisphenol A), pharmaceutical class (Diclofenac, Carbamazepine, Amoxicillin, Estrone (E1)), pesticide class (Diazinon, Lindane, Dieldrin), and disinfection by-products class (NDMA) [36]. These ECs are not easily removed in conventional wastewater treatment processes due to their being highly resistant, i.e., recalcitrant and refractory, to biodegradation, i.e., microorganisms [25, 47–49]. Thus, they can pass through these conventional processes into the RO process, increasing their concentrations in the ROC [36].

3.ENVIRONMENTAL IMPACT OF ROC

Discharging untreated ROC obtained from municipal and industrial wastewater treatment plants into the environment (natural water bodies and soil) raises environmental concerns. On one side, discharging various pollutants of ROC from the secondary effluent of municipal wastewater treatment at concentrations exceeding the standard limits into the soil, i.e., agriculture farms, for irrigation purposes, contaminates the soil with these pollutants. Among these pollutants, as can be seen from Table 1, for example, the presence of excessive nutrients in ROC can cause eutrophication, increasing the growth of algae, i.e., algal bloom, and other plants, leading to oxygen depletion, resulting in the death of aquatic organisms [6, 23]. The presence of nutrients in ROC can degrade the soil structure, where soil quality plays a vital role in supporting ecosystem functions and promoting plants. The degradation of soil structure can affect water storage and decrease permeability. Thus, the health of crop production can be significantly affected [6, 50]. The presence of chloride ions (Cl^{-}) in ROC can easily move through the transpiration streams of crops and then accumulate in their leaves, causing toxic effects on plants such as leaf burn (i.e., drying of leaf tissue and severe foliage damage) and impaired growth [51]. Also, it can cause corrosion of the pipe system during the treatment of ROC [52]. The presence of magnesium ions (Mg^{2+}) in ROC is associated with soil aggregation and friability [51]. The presence of excess dissolved salts, i.e., salinity, in the ROC decreases soil productivity and affects plants by causing osmotic stress, i.e., a rapid change in water movement across their cell membrane [6, 51, 53, 54]. The presence of high concentrations of sodium ions (Na^{+}) in ROC increases the alkalinity of soil and, as a result, causes soil dispersion and swelling. Thus, the soil permeability is significantly affected, making less water available to crops [51, 55]. The exceeded normal pH range in ROC for irrigation can affect the production of plants, i.e., nutritional imbalance

[6, 51]. The presence of heavy metals, such as Cd, Cu, Ni, Cr, and Zn, in ROC may cause toxic effects in agricultural soil and plants. The food quality significantly deteriorates, thus posing hazards to the health and safety of humans and animals [6, 51, 56]. On the other side, discharging various emerging contaminants (ECs) of ROC from industrial wastewater effluents into natural water bodies and soil has several negative impacts. These emerging contaminants can, in their nature, persist in water bodies for long periods and be toxic to aquatic life, disrupting the natural balance of the ecosystem. They can also accumulate in the food chain, causing health hazards for humans and animals [6, 47, 49, 57, 58]. To address these concerns about the relationship between ROC and its application in soil, plants, and water; it is important to implement effective treatment methods to treat ROC to an acceptable level of target contaminants before discharging into the environment, discussed in the coming section.

4.TREATMENT OF ROC

Common disposal practices for ROC are surface water, deep well injection, evaporation ponds, land application, and dilution. Disposal into surface water includes rivers, bays, tidal lakes, brackish canals, and oceans. Deep well injection is used to transfer ROC to well depths, depending on the geological conditions at the site of ROC generation. Evaporation ponds or lagoons are applied for small volumes of ROC in specific weather conditions (warm and dry climates). The natural evaporation of water content in ROC occurs over time. Thus, the remaining ROC becomes more concentrated. However, this method requires available lands with suitable measures to prevent leakage into groundwater. The land application of ROC is used for agricultural purposes since it contains some beneficial nutrients. However, environmental considerations must be taken to prevent soil or groundwater contamination. Dilution is used to dilute the ROC with large volumes of municipal or industrial water to decrease its concentration before disposal. However, sufficient water resources for dilution are required. Based on these common disposal practices of ROC, the environmental management of ROC with respect to quality and quantity remains a significant issue [36, 41, 59, 60]. To address this issue, the characteristics of ROC play a significant role in deciding the type of efficient treatment to be selected [37]. Integrated treatment technologies are the best strategic solutions for improving the ROC quality and minimizing the ROC quantity [36]. These integrated technologies based on the role of ZLD or n-ZLD have been developed to reduce (recover) the volume of ROC disposal by 95–98% [41]. These integrated treatment technologies consist of pretreatment in conjugation with a membrane separation process for treating ROC in two modes of operation (complete and partial recirculation). Generally, in the literature, there has been little research on applying these integrated technologies with these modes of operation for the treatment of ROC. Some examples of the most efficient integrated treatment processes are reviewed below.

4.1.Complete Recirculation Mode of Operation

Comerton et al. [61] developed an integrated system for treating membrane bioreactor (MBR) effluent from municipal wastewater, as shown in Fig. 1. The MBR effluent from its storage tank was divided into two streams. The first one was treated with the photolysis process (ultraviolet, UV irradiation) as an advanced oxidation process (AOP), and the second one was treated with chemical oxidation using monochloramine (NH_2Cl) as a conventional oxidant. The treated MBR effluent collected from both streams was transferred to a mixing tank and then delivered to the crossflow membrane filtration unit. The ROC was returned to the mixing tank under the complete recirculation mode and mixed with treated MBR effluent. Both were used as influent feed to the crossflow filtration unit. This system showed a high-quality permeate that meets the regulations for water reuse for non-portable applications. Also, in this system, the complete recirculation mode of operation played a significant role in decreasing the quantity of ROC produced.

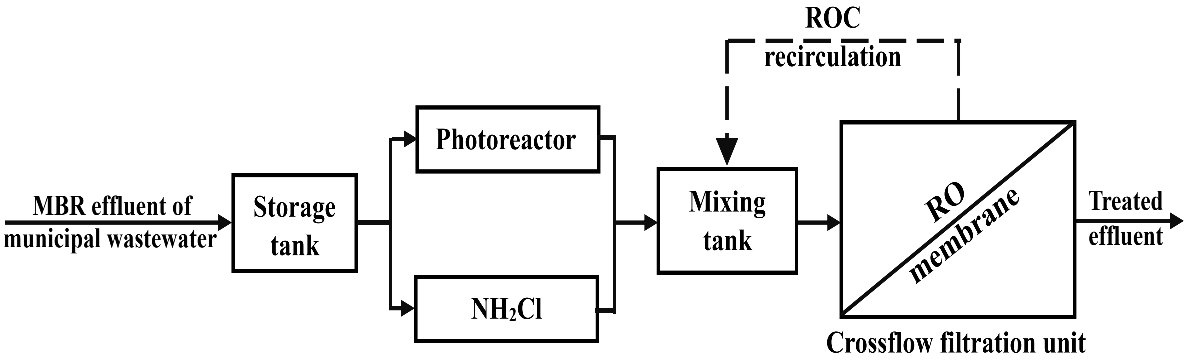

Fig. 1 A Schematic Diagram of an Integrated Treatment System, Adapted from [61].

Another example of complete recirculation of concentrate was proposed by Secondes et al. [62], who applied a hybrid system for treating three types of emerging contaminants (ECs) in synthetic industrial wastewater, including diclofenac, carbamazepine, and amoxicillin. A schematic diagram of this system is shown in Fig. 2. Three different processes–activated carbon adsorption, ultrasound irradiation, and ultrafiltration–were simultaneously used in a hybrid treatment system. Various doses of powdered activated carbon as adsorbents were added to the synthetic industrial wastewater of ECs to form a suspension solution. Adsorption by these adsorbents was dominant in removing most ECs, which was significantly enhanced by ultrasonic irradiation. The suspension solution was delivered to the crossflow filtration unit. The concentrate was completely recycled and mixed with the suspension solution. Both were used as influent feed to the filtration unit. This hybrid treatment system could remove nearly 99.5% of the ECs due to the synergistic effects of the three processes and enhance the volume recovery rate of treated wastewater as permeate.

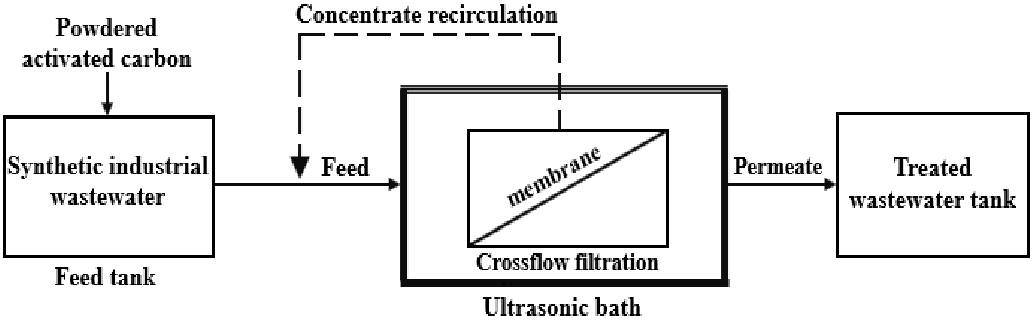

Fig. 2 A Schematic Diagram of a Hybrid Treatment System, Adapted from [62].

4.2.Partial Recirculation Mode of Operation

Joss et al. [63] proposed an integrated system for treating membrane bioreactor (MBR) effluent from municipal wastewater, as shown in Fig. 3. This effluent was chemically conditioned using monochloramine (NH_2Cl) as a conventional oxidant and CO_2 to decrease biofouling and inorganic scaling of ROC, respectively, and then fed to the crossflow filtration unit. On one side, the ROC produced was divided into two streams. The first stream of ROC was partially recycled and then mixed with chemically conditioned MBR effluent. Both were fed as influent to the filtration unit. The second stream of ROC was divided into parts. In the first part, the rate for feeding to the ozonation unit before recycling it to the biological unit, i.e., MBR, was nearly 90%. While, in the last part, the rate of disposal was 10%. On the other hand, the rate of permeate produced was 90%. This system used double-partial recirculation of ROC as a mode of operation, thus increasing the quantity of permeate production. Also, it showed a high permeate quality suitable for many reuse purposes. Malamis et al. [64] used a combined system for treating membrane bioreactor (MBR) permeate of municipal wastewater, as shown in Fig. 4. This MBR permeate was fed from its storage tank to the crossflow filtration unit. The ROC was divided into two streams. The first stream was partially recycled and mixed with MBR permeate from municipal wastewater to be used together as influent feed for the crossflow filtration unit. The second stream was treated with a fixed bed column packed with natural zeolite to decrease the concentration of heavy metals to acceptable levels for safe discharge. This combined system produced the permeate with high-quality treated wastewater suitable for agricultural purposes.

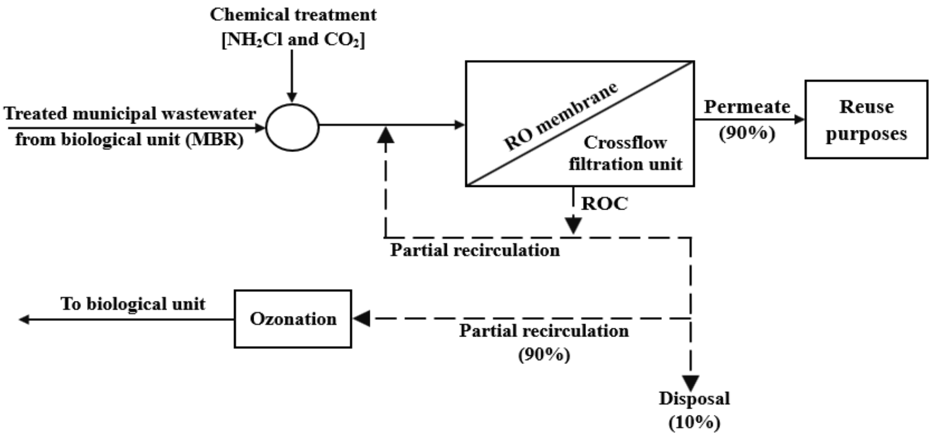

Fig. 3 A Schematic Diagram of an Integrated Treatment System, Adapted from [63].

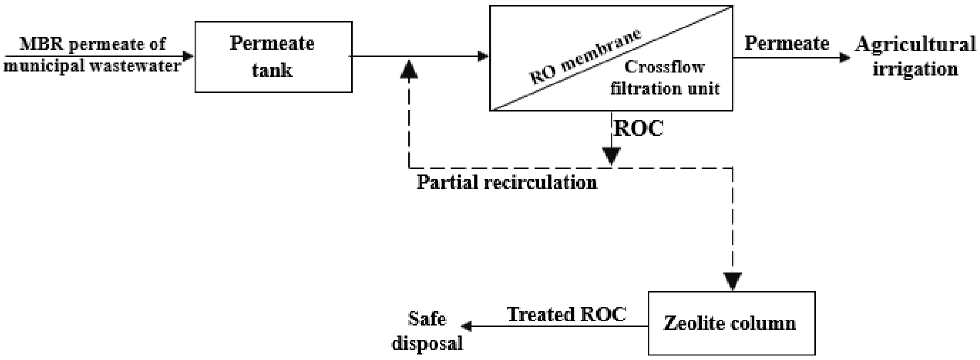

Fig. 4 A Schematic Diagram of a Combined Treatment System, Adapted from [64].

Hee and Tansel [36] applied an integrated system for treating membrane bioreactor (MBR) permeate of municipal wastewater, as shown in Fig. 5. In this system, the MBR permeate from its storage tank was fed for the crossflow filtration unit. The ROC was divided into two streams. The first stream was partially recycled and mixed with MBR permeate from municipal wastewater to be used together as influent feed for the crossflow filtration unit. The second stream was treated with two sequential processes. The first process was adsorption using powdered activated carbon, and the second process was chemical oxidation using persulfate in the presence of nano-sized zero-valent iron (nZVI) to generate the radicals of $HO^{\bullet}$ and $SO_4^{\bullet -}$ as reacting species to degrade the organic compounds in the ROC. These sequential processes showed effective performance in removing hazardous substances in the ROC. Table 2 compares the advantages and disadvantages of the complete and partial recirculation mode of operations for treatment ROC.

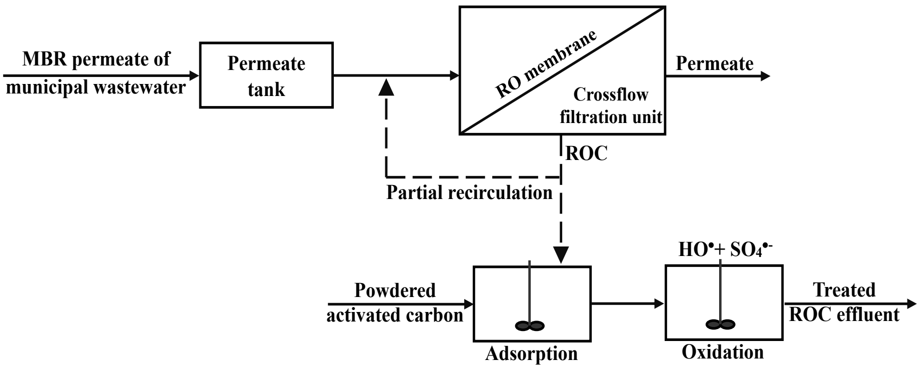

Fig. 5 A Schematic Diagram of an Integrated Treatment System, Adapted from [36].

Table 2 Advantages and Disadvantages of Complete and Partial Recirculation Modes of Operations for the Treatment of ROC.

Type of Operation Mode	Process Schematic	Advantages	Disadvantages	Reference
Complete recirculation	Fig. 1	• High Recovery rate	• High chemical materials are required. • Incomplete removal of pollutants • Membrane fouling • High operational cost.	[61]
	Fig. 2	• High recovery rate. • High removal efficiency of pollutants.	• High capital cost • High operational cost • Maintenance issues	[62]
Partial recirculation	Fig. 3	• 90% of the overall recovery rate obtained.	• High chemical materials are required. • Membrane fouling • High operational cost	[63]
	Fig. 4	• High recovery rate • High removal efficiency of heavy metals	Membrane fouling High operational cost	[64]
	Fig. 5	• High recovery rate • High removal efficiency of hazardous substances	Membrane fouling Maintenance issues	[36]

5.PROPOSED PRETREATMENT OPTIONS APPLIED TO THE ROC

Advanced oxidation processes (AOPs) have been successfully applied to treat municipal or industrial ROCs based on a batch system (no recirculation mode of operation). These processes can generate free hydroxyl radicals ($HO^{\bullet}$) as a strong and powerful oxidizing agent that reacts with a broad spectrum of dissolved organic constituents in these ROCs to degrade (oxidize) them [65–71]. In their literature review, Ganiyu et al. [72] showed that AOPs can be coupled with the membrane separation processes, producing an integrated treatment that can overcome the defects of either AOP or the membrane separation process when used alone. Further to the applied pretreatment processes (photolysis, chemical oxidation) using conventional oxidants (persulfate, monochloramine) and adsorption using powdered activated carbon) shown in Section (4), the following AOPs as promising options are proposed to be combined with the crossflow membrane separation process working on the complete recirculation mode of operation to produce an integrated technology for the treatment of ROC generated from municipal or industrial effluents. These proposed options will extend the application of ZLD as a strategic solution for offering alternative water resources.

5.1.Fenton Process

Fenton reagent and Fenton-like reagent are chemical oxidation processes that generate the $HO^{\bullet}$ through the reaction of iron ions as a catalyst from salts, such as ferrous ions (Fe^{2+}) and ferric ions (Fe^{3+}), respectively, in the existence of hydrogen peroxide (H_2O_2) as a conventional oxidant in an acidic solution. The reactions of Fenton reagent and Fenton-like reagent can be described by Eqs. (1–6), and the radical-radical reactions can be described by Eqs. (7–10) [71, 73, 74]:

$$\mathbf{Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + HO^- + HO^{\cdot}} \quad (1)$$
$$\mathbf{Fe^{2+} + HO^{\cdot} \rightarrow Fe^{3+} + HO^-} \quad (2)$$
$$\mathbf{Fe^{2+} + HO_2^{\cdot} \rightarrow Fe^{3+} + HO_2^-} \quad (3)$$
$$\mathbf{Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO_2^{\cdot} + H^+} \quad (4)$$
$$\mathbf{Fe^{3+} + HO_2^{\cdot} \rightarrow Fe^{2+} + O_2 + H^+} \quad (5)$$
$$\mathbf{Fe^{3+} + O_2^{\cdot-} \rightarrow Fe^{2+} + O_2} \quad (6)$$
$$\mathbf{HO^{\cdot} + H_2O_2 \rightarrow H_2O + HO_2^{\cdot}} \quad (7)$$
$$\mathbf{HO^{\cdot} + HO^{\cdot} \rightarrow H_2O_2} \quad (8)$$
$$\mathbf{HO_2^{\cdot} + HO_2^{\cdot} \rightarrow H_2O_2 + O_2} \quad (9)$$
$$\mathbf{HO_2^{\cdot} + HO^{\cdot} \rightarrow H_2O + O_2} \quad (10)$$

The Fenton process was used to treat ROC generated from the treated secondary effluent of municipal wastewater. For example, Westerhoff et al. [75] showed that Fenton reagent and Fenton-like reagents could remove up to 50% of DOC in the ROC. For the treatment of ROC from the treated secondary effluent petroleum industrial wastewater, Cai et al. [65] reported that the COD in the ROC was removed by 37% at the optimum oxidation conditions (Fenton reagent ratio (H_2O_2/Fe^{2+}) is 8 and pH 3). Generally, no more studies have been found in the literature relating to the application of the Fenton process for the treatment of ROC from industrial resources, despite the effective application of the Fenton process for the treatment of various types of industrial wastewaters (in particular, pharmaceutical type) [76]. However, a combination of the Fenton process and other processes is the most recent development in this field. Ren et al. [70] applied a combination of the Fenton process and the zero-valent iron (Fe^0) process for treating the ROC generated from the biologically treated secondary effluent industrial wastewater (amino acid production plant). Usman et al. [77] used integrated adsorption and Fenton processes to treat pharmaceutical wastewater. The performance of the Fenton process can be enhanced by connecting it with the photolysis process, i.e., using UV irradiation). This connection is referred to as the Photo-Fenton process or the photo-assisted Fenton process. The presence of UV irradiation catalyzes Fenton's reaction, resulting in more generation of $HO^{\bullet}$ via synergistic effect, thus enhancing the overall degradation performance [68, 78, 79].

5.2.Photolysis Process

The photolysis process works based on the ability of pollutants in their solution to absorb UV irradiation from sources of solar or artificial lights with sufficient energy to break down the covalent bonds of these pollutants [80, 81]. Generally, direct photolysis has a limited ability to degrade most compounds compared with other AOPs [82]. To overcome this drawback, conventional oxidants like H_2O_2 were used in conjunction with photolysis, i.e., UV), referred to as the UV/H_2O_2 process, to accelerate the photolytic reactions by increasing the rate of formation of $HO^{\bullet}$, thus enhancing the overall degradation [80, 83, 84]. The reactions of the UV/H_2O_2 process can be described by the Eqs. (11–16) [23, 85]:

$$\mathbf{H_2O_2 + h\nu \rightarrow HO^{\cdot} + HO^{\cdot}} \quad (11)$$
$$\mathbf{H_2O_2 + HO^{\cdot} \rightarrow HO_2^{\cdot} + H_2O} \quad (12)$$
$$\mathbf{H_2O_2 + HO_2^{\cdot} \rightarrow HO^{\cdot} + H_2O + O_2} \quad (13)$$
$$\mathbf{HO^{\cdot} + HO^{\cdot} \rightarrow H_2O_2} \quad (14)$$
$$\mathbf{HO_2^{\cdot} + HO_2^{\cdot} \rightarrow H_2O_2 + O_2} \quad (15)$$
$$\mathbf{HO^{\cdot} + HO_2^{\cdot} \rightarrow H_2O + O_2} \quad (16)$$

The UV/H_2O_2 process has been successfully applied to treat ROC generated from the treated secondary effluent of municipal wastewater. Some examples of these applications are shown in Table 3. While, for the application of the UV/H_2O_2 process in the treatment of ROC generated from the treated secondary effluent of municipal wastewater, no study has been found in the literature, despite the effective application of UV/H_2O_2 process for treating various types of industrial wastewaters, such as phenolic compounds [83], organic pollutants [86], polystyrene microplastic [80], and pharmaceuticals [84].

Table 3 Application of UV/H_2O_2 Process for Treating ROC.

Type of ROC	Parameter	Removal (%)	Reference
Municipal wastewater	DOC COD	25 46	[23]
Municipal wastewater	DOC COD	26-38 25-37	[87]
Municipal wastewater	DOC COD	45-46 44-48	[88]
Municipal wastewater	DOC COD Color	38 50-55 100	[89]
Municipal wastewater	DOC COD Color	15 15 86	[90]
Municipal wastewater	DOC Color	29 96	[91]
Municipal wastewater	DOC	14	[92]
Municipal wastewater	DOC COD Color	15 16 50	[93]

5.3.Photocatalysis Process

Photocatalysis is a combination process using a semiconductor metal oxide as a photocatalyst in the presence of a light source (usually UV) to accelerate chemical photoreactions under mild conditions (ambient temperature and pressure) [94]. This process has been one of the most widely applied AOPs for treating various pollutants in industrial wastewater [95]. One of the most common types of heterogenous photocatalysts is titanium oxide (TiO_2), which has attracted significant attention as an ideal and standard heterogenous photocatalyst due to its high reactivity, high chemical, thermal, and mechanical stability, less toxicity, and low cost [47, 96]. The TiO_2-heterogenous photocatalysis mechanism occurs by the photoactivation of TiO_2 via the absorption of photons with a certain level of energy (equal or higher than that of the band gaps of TiO_2) to generate a pair of charge carriers, positive holes (h_{VB}^+), and electrons (e_{CB}^-). These charge carriers can go through a series of redox reactions to generate the free hydroxyl radicals ($HO^•$) and other radicals like ($HO_2^•$) and ($O_2^{•-}$). These reactions can be presented by the following equations [97–100]:

$$TiO_2 + h_v \xrightarrow{\geq E_{BG}} h_{VB}^+ + e_{CB}^- \quad (17)$$
$$h_{VB}^+ + \text{pollutants} \rightarrow \text{oxidized pollutants} \quad (18)$$
$$h_{VB}^+ + H_2O \rightarrow HO^• + H^+ \quad (19)$$
$$e_{CB}^- + O_2 \rightarrow O_2^{•-} \quad (20)$$
$$O_2^{•-} + H^+ \rightarrow HO_2^• \quad (21)$$
$$e_{CB}^- + HO_2^• \rightarrow HO_2^- \quad (22)$$
$$HO_2^- + H^+ \rightarrow H_2O_2 \quad (23)$$
$$H_2O_2 + h_v \rightarrow 2HO^• \quad (24)$$
$$H_2O_2 + e_{CB}^- \rightarrow HO^• + OH^- \quad (25)$$
$$H_2O_2 + HO^• \rightarrow HO_2^• + H_2O \quad (26)$$

Photocatalysis (UV/TiO_2) has been successfully used to treat ROC, as shown in Table 4.

Table 4 Application of Photocatalysis (UV/TiO_2) Process for Treating ROC.

Type of ROC	Parameter	Removal (%)	Reference
Pharmaceutical wastewater	DOC	95	[75]
Municipal wastewater	DOC	50	[101]
Municipal wastewater	DOC	72	[102]

Figure 6 shows the proposed AOP as a pretreatment process in combination with a membrane separation process working on the complete recirculation mode of operation to generate an integrated technology for treating the treated effluent of either municipal or industrial wastewater.

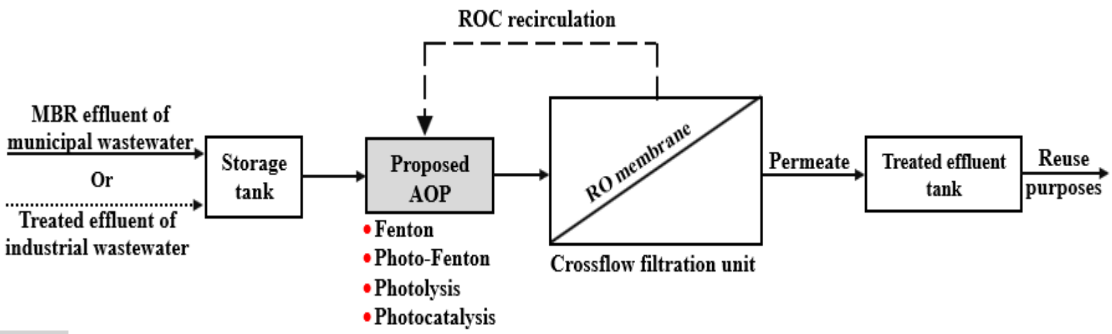

Fig. 6 A Schematic Diagram of a Proposed AOPs as a Pretreatment Process with the Membrane Separation Process.

Table 5 shows the advantages and disadvantages of advanced oxidation processes proposed to be combined with a crossflow membrane separation process for treating ROC generated from municipal or industrial effluents.

Table 5 Advantages and Disadvantages of Advanced Oxidation Processes Proposed.

Proposed process	Advantages	Disadvantages	Reference
Fenton	• High degradation rate • High effective performance • Operated in mild conditions • Minimal energy required	• High H_2O_2 required, increasing cost • High sludge formed • Formed sludge requires further treatment. • Restricted by pH range (2-3)	[103, 104]
Photolysis	• Simple and flexible process • Easily operated • No photocatalyst is needed. • No sludge formation • Eco-friendly process	• Conventional oxidant (H_2O_2) may be needed to increase the overall degradation efficiency. • Colloidal turbidity can affect its efficiency. • Relatively high operating cost • A continuous energy source is needed.	[105, 106]
Photocatalysis	• Efficient process in degrading a wide range of pollutants • No sludge formation • Operated in mild conditions • Eco-friendly process	• Separation and recycling of suspended photocatalysts are required. • Colloidal turbidity can affect its efficiency. • Optimum photocatalyst loading should be investigated experimentally to obtain an optimum photocatalytic performance. • A continuous energy source is needed. • High operational cost	[104, 107]

6.CONCLUSION

The direct disposal of huge volumes of untreated ROC generated from municipal and industrial resources into natural water bodies and soil is problematic due to its serious environmental threat. Thus, managing this issue is still a technological challenge. However, in the present review, integrated treatment technologies based on the application of the ZLD (or n-ZLD) concept have been addressed as a better option than direct disposal methods due to their recovery of treated wastewater effluent and production of a high-quality permeate that can be used for many reuse purposes. Several pretreatment processes, such as photolysis, chemical oxidation using conventional oxidants (persulfate, monochloramine), and adsorption using powdered activated carbon, showed promising performance in combination with a crossflow membrane separation process working on two modes of operation (complete and partial recirculation), producing an integrated technology for the treatment of ROC generated from municipal and industrial resources. To extend the concept of ZLD (or n-ZLD) in the real applications for the treatment of ROC produced from these resources as a sustainable solution, this paper suggests AOPs, such as Fenton, photo-Fenton, photolysis, and photocatalysis as pretreatment options applied successfully to treating ROC in a batch system, i.e., no recirculation, to be combined with the crossflow membrane separation process, producing an integrated technology with complete or partial recirculation modes of operation. The advantages of these integrated technologies are that they produce vast quantities of permeate, which can be considered an alternative and sustainable resource for conserving limited freshwater resources. Evaluation of the performance of these treatment technologies in large-scale applications is necessary. Future research should consider the economic costs in terms of operation, maintenance, and energy.

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