

Elimination of DOC in RO concentrate from a wastewater reclamation plant: Cost comparison of different AOPs

Beom Joon Kim¹, Joon-Young Choi², Hye-Jin Lee^{1†}, Changha Lee^{1†}

¹School of Chemical and Biological Engineering, Institute of Chemical Process (ICP), and Institute of Engineering Research, Seoul National University, Seoul, 08826, Republic of Korea

²Hyorim Industries Inc., Gyeonggi-do, 13517, Republic of Korea

Received August 17, 2022 Revised October 2, 2022 Accepted October 20, 2022

ABSTRACT

Advanced oxidation processes (AOPs) have been reported as alternative ways for treating reverse osmosis (RO) concentrate which is the primary concern for water reclamation plants. AOPs are known to oxidize recalcitrant organic compounds by a highly reactive and nonselective oxidant, hydroxyl radical (*OH). Researchers showed that AOPs such as the conventional Fenton, UV/H₂O₂, ozonation, electrochemical oxidation processes could remove organics in RO concentrate. However, comparing different AOPs has been rarely performed for the treatment of RO concentrate. In addition, the cost of these processes has not been fully explored. In this study, five different AOPs (i.e., the conventional Fenton, UV/H_2O_2 , ozonation, anodic oxidation, and electro-Fenton processes) were evaluated for the removal of dissolved organic carbon (DOC) in RO concentrate. The operation costs for DOC removal by these AOPs were also calculated. The results indicated that the conventional Fenton and ozonation processes were the most cost-efficient among the processes tested.

Keywords: AOP, Oxidation, RO concentrate, Water reuse



Graphical Abstract

This is an Open Access article distributed under the terms $(\mathbf{\hat{r}})$ (cc) of the Creative Commons Attribution Non-Commercial License (http://creativecommons.org/licenses/by-nc/3.0/) which permits unrestricted non-commercial use, distribution, and reproduction in any medium, provided the original work is properly cited.

Copyright © 2023 Korean Society of Environmental Engineers

[†] Corresponding authors

E-mail: leechangha@snu.ac.kr (C.L.), leehj5138@snu.ac.kr (H.-J.L.) Tel: +82-2-880-8630 (C.L.), +82-2-880-8629 (H.-J.L.) Fax: +82-2-888-7259 (C.L.), +82-2-888-7259 (H.-J.L.) ORCID: 0000-0002-0404-9405 (C.L.), 0000-0002-0142-3406 (H.-J.L.)

1. Introduction

Water shortage problem and pollution caused by industrial development and population increase have been significantly intensified in recent decades [1, 2]. The water shortage problem is getting more severe, and this problem may threaten human life critically. Consequently, sustaining plenty of clean and high quality of water will be a main issue for future human beings worldwide. One of the ways to solve this problem is to reuse wastewater effluents from wastewater treatment plants (WWTPs); WWTP effluents can be supplied to industrial and agricultural sectors after proper treatment [3, 4]. The treatment using reverse osmosis (RO) membranes has been broadly used in wastewater reclamation plants because the RO process can produce high quality of reused water for different industrial purposes [5]. However, one of the disadvantages of this process is that the RO concentrate is inevitably produced as a byproduct. In the RO concentrate, recalcitrant organic compounds, which were not properly treated by biological processes in WWTPs, exist at a higher concentration than the WWTP effluent [6]. The RO concentrates were usually returned to the feed stream of WWTPs. However, this RO concentrate return can result in the accumulation of non-biodegradable organics in the biological processes, increasing the organic load in WWTPs [7, 8]. Therefore, proper treatment of recalcitrant organic compounds in RO concentrate is required.

Advanced oxidation processes (AOPs) are known to generate hydroxyl radical (\cdot OH), which is a strong oxidant capable of oxidizing a wide spectrum of organic compounds in water [9-11]. AOPs have been studied for treating the residual organic compounds in RO concentrate [12-20]. UV/H₂O₂ process removed approximately 75% dissolved organic carbon (DOC) in RO concentrate in 3 hours [12], anodic oxidation using BDD as anode almost completely removed chemical oxygen demand (COD) in RO concentrate in 1 hour [13], and microwave-Fenton oxidation removed approximately 84% COD in RO concentrate [20]. However, these studies did not compare the efficiency of AOPs by the same standard (e.g., COD or DOC) and same target RO concentrate. In addition, there was little information in previous literature comparing the cost for treating RO concentrate by AOPs.

In this study, five AOPs that are widely used in the field (i.e., the conventional Fenton, UV/H_2O_2 process, ozonation, anodic oxidation, and electro-Fenton processes) were chosen for the treatment of RO concentrate from a wastewater reclamation plant. The DOC removal of RO concentrate was examined, and the cost required for the unit DOC removal was comparatively evaluated for these AOPs.

2. Materials and Methods

2.1. Reagents and RO Concentrate

All chemicals were of a reagent grade and used as received without further purification. The chemicals used in this study include FeSO₄·7H₂O, H₂O₂ (30 wt%) and HClO₄ (70 wt%) solutions, and NaOH (all from Sigma-Aldrich). Deionized water (DI, 18.2 M $\Omega \times$ cm, Millipore) was used to prepare stock solutions.

Table 1. Water quality parameters of the RO concentrate sample

Parameters	Values
DOC (mg/L)	44
pH	7.8
UV absorbance at 254 nm	1.16
Conductivity (μ S/cm)	8,486
Na ⁺ (mg/L)	2,000
K ⁺ (mg/L)	143
Ca ²⁺ (mg/L)	1,000
Mg^{2+} (mg/L)	200
Cl ⁻ (mg/L)	1,414
NO ₃ ⁻ (mg/L)	111
SO ₄ ²⁻ (mg/L)	2,000
T-N (mg/L)	17.53
T-P (mg/L)	< 1.0

The RO concentrate was collected from a wastewater reclamation plant located in Daegu, Korea (refer to Table 1 for water quality parameters of sampled RO concentrate). The RO concentrate was filtered by a GF/C filter (1.6 mm, Whatman) to remove suspended solids, and stored at 4° C until use.

2.2. Experiments for the Conventional Fenton Process

The following procedures were performed for the experiments of the conventional Fenton process. First, the pH value of RO concentrate (100 mL) was adjusted as 3.0 (the optimum pH for the Fenton reaction [21]) using HClO₄. Second, aliquots of FeSO₄. 7H₂O and H₂O₂ were sequentially injected into the solution to initiate the reaction, and after 1 h, the solution pH was increased to 7.0 using NaOH to finish the reaction. Lastly, RuO₂ powder was added to the solution to quench residual H_2O_2 for the accurate DOC measurement; RuO₂ powder was immediately removed by a 0.45 μ m PTFE filter (Advantec). The experiments were performed at different initial concentrations of Fe(II) and H₂O₂ (i.e., [Fe(II)]₀ = 1.0 ,3.0 5.0 mM and $[H_2O_2]_0$ = 1.5, 4.5, 7.5 mM). Design Expert 7 software (Stat-Ease Inc., USA) was used for response surface methodology (RSM) which was applied to evaluate the optimal doses of Fe(II) and H₂O₂ for the highest DOC removal. A two-factor and three-level central composite face-centered design (CCFCD) was applied to depict regression model. The analysis of variances (ANOVA) was validated using the Design Expert 7.

2.3. UV/H₂O₂ and Ozonation Experiments

The UV/H₂O₂ experiments were performed in a 100 mL quartz reactor which was placed in a dark chamber equipped with six 4W low pressure mercury lamps ($\lambda_{MAX} = 253.7$ nm, Philips), a magnetic stirrer, and a cooling fan. The incident light intensity was measured to be 4.12×10^{-6} Einstein L⁻¹ s⁻¹ (equivalent to 1.982 J L⁻¹ s⁻¹) by the ferrioxalate actinometry [22]. The initial H₂O₂ concentrations for the experiment were 4.5 and 45 mM. The reaction was initiated by UV illumination, and after 2 h, samples were withdrawn for analyses and RuO₂ powder was additionally added for quenching residual H₂O₂ in the sample.

A bench scale ozone generator (LAB-II, Ozonetech) was used

to produce ozone-containing gas (ca. 5 wt%) for ozonation experiments. In order to initiate the reaction, the ozone gas was directly diffused into the reactor (100 mL). Samples were withdrawn after 15, 30, 60 min, which were equivalent to ozone doses of 50.17, 135, and 318.58 mg/L, respectively.

2.4. Experiments for Electrochemical Oxidation

The electrochemical oxidation of RO concentrate was tested in a small cell (25 mL solution) equipped with two plate electrodes $(7 \text{ cm}^2 \text{ each})$ connected to a potentiostat for supplying power (Bio-Logic SAS, VSP). The electrolysis was performed with and without Fenton's reagent (i.e., the electro-Fenton process and the anodic oxidation, respectively). In preliminary experiments, four anode materials (IrO2, RuO2, Pt, and BDD, obtained from Wescoelectrode) were tested for the anodic oxidation of RO concentrate, and thereafter BDD was chosen as the anode (Fig. S1). Stainless steel (SS) (Alfa Aesar) was used as a cathode material for all experiments. The solution pH was adjusted to 3.0, and FeSO4 · 7H₂O (1.0 mM) and H_2O_2 (7.5 mM) were used for the electro-Fenton experiments. The electrolysis was initiated by applying current into the cell, and samples were withdrawn after 1 h of reaction. RuO_2 powder was added in the samples which included H_2O_2 . Constant currents were applied at current densities of 5, 10, 20 mA/cm², and cell voltages were monitored during the electrolysis.

2.5. Analyses

DOC was measured using a TOC analyzer (Sievers M5310C, Suez) which employed the UV/Persulfate method. DOC of each sample was measured after dilution four times with DI to fit into the prepared calibration curve range. The concentration of H_2O_2 was measured by the titanium sulfate method [23]. The concentration of O_3 was determined by monitoring the decolorization of indigo trisulfonate at 600 nm [24]. The absorbance for these colorimetric methods was measured using a UV-Vis spectrophotometer (LAMBDA 465, Perfkin-Elmer).

2.6. Cost Evaluation

The costs for DOC removal by different AOPs were calculated based on the prices of chemicals, electricity, and sludge treatment (for the Fenton process) (Table 2). The prices of $FeSO_4 \cdot 7H_2O$, H_2O_2 , H_2SO_4 , and NaOH were from Korea Price Information [25]. Electrical costs were calculated by Korean Electricity Information for WWTPs [26]. The price of ozone was calculated based on

 Table 2. Cost references of chemicals, electricity, and sludge landfill for the RO concentrate treatment by AOPs

for the red concentrate treatment of reard		
Material	Cost	Reference
$FeSO_4 \cdot 7H_2O$	305.41 \$ / 1 ton	[25]
H_2O_2 (35%)	872.60 \$ / 1 ton	[25]
Sludge landfill	80.63 \$ / 1 ton	[28]
Electricity at WWTP	0.14 \$ / kWh	[26]
H_2SO_4	78.53 \$ / 1 ton	[25]
NaOH	708.55 \$ / 1 ton	[25]
O_3	4,205.06 \$ / 1 ton	[27]

electricity required to generate ozone gas in an industrial scale [27]. Sludge treatment costs were adopted from the Korean WWTP report [28].

3. Results and Discussion

3.1. Conventional Fenton Process

The DOC removal efficiency of RO concentrates by the conventional Fenton process was examined by varying concentrations of Fe(II) (1.0 - 5.0 mM) and H₂O₂ (1.5 - 7.5 mM). The obtained results were evaluated by RSM using the concentration of Fe(II) and H₂O₂ as two independent variables (Fig. 1). The experimental data was fitted to develop a model equation (Eq. (1)) and the adequacy of developed model was verified by the four representative estimates, i.e., *p*-value = 0.0002, lack-of-fit = 0.9317, R^2 = 0.982, adequate precision = 18.632.

 $\begin{array}{l} \text{DOC removal (\%)} = 36.23 + 8.82 [Fe(II)] + 1.2 [H_2 O_2] + \\ 0.8 [Fe(II)] [H_2 O_2] \text{-} 5.33 [Fe(II)]^2 - 1.88 [H_2 O_2]^2 \end{array} \ (1)$

Based on the RSM model, the optimal DOC removal efficiency was estimated to be 40.35% at 4.7 mM Fe(II) and 4.5 mM H₂O₂ under investigated experimental conditions. Increasing the Fe(II) dose significantly increased DOC removal efficiency, while increasing the H₂O₂ dose marginally impacts on the DOC removal as compared to that of Fe(II). The consumed amount of H_2O_2 during the reaction proportionally increased with increase in the initial Fe(II) concentration (Fig. S2). These results indicate that the production of oxidants (\cdot OH) through the decomposition of H₂O₂ (Eq. (2)) is encouraged to oxidize organic compounds in RO concentrate as the initial Fe(II) concentration increases. Unlike Fe(II), the change in H₂O₂ concentration exhibited insignificant impacts on the DOC removal efficiency although H₂O₂ consumption showed a positive relationship with increasing the input concentration of H_2O_2 . This result can be explained by $\cdot OH$ -scavenging effects of a high concentration of H₂O₂. It is well-established chemistry that produced \cdot OH can be scavenged by Fe(II) and H₂O₂ (Eq. (3),(4)). As such, increasing the concentration of Fe(II) and H₂O₂ does not necessarily result in enhanced oxidation of organic compounds [29-31]. In addition, the increase of Fe(II) dose presumably contributed to enhanced coagulation performance as all of the reactions were finished by increasing pH to a neutral condition (i.e., pH 7), resulting in a positive effect on DOC removal.

$$Fe(II) + H_2O_2 \rightarrow Fe(III) + OH^- + \cdot OH$$
 (2)

$$\cdot OH + H_2 O_2 \rightarrow HO_2 \cdot + H_2 O \tag{3}$$

$$\cdot \text{OH} + \text{Fe(II)} \rightarrow \text{Fe(III)} + \text{OH}^{-}$$
 (4)

3.2. UV/H₂O₂ Process

The DOC removal efficiency of the RO concentrate by the UV/H₂O₂ process was investigated at two different H₂O₂ concentrations (i.e., $[H_2O_2]_0 = 4.5$ and 45.0 mM) (Fig. 2a). DOC removal efficiency by UV/H₂O₂ process at 4.5 mM of H₂O₂ was only 14% in 2 hours.



Fig. 1. Effects of Fe(II) and H_2O_2 dose on the DOC removal efficiency in RO concentrate using conventional Fenton process (the red dots on the response surface represent the experimental data points).

This result indicates that most of photon (> 90% referred to Table. 1) was absorbed by RO concentrate rather than H_2O_2 that produces ·OH through H_2O_2 photolysis (Eq. (5)), directly showing a typical limitation of the UV/H_2O_2 process. As increasing H_2O_2 concentration to 45.0 mM, DOC removal efficiency was significantly enhanced by 61% after 2 hours of UV irradiation. This enhancement was attributed to the increase in the fraction of UV absorption by H2O2 (From 2.52% at 4.5 mM H2O2 to 35.33% at 45.0 mM H_2O_2) with increasing the input concentration of H_2O_2 (Fig. S3), thereby more photons would be absorbed by H₂O₂ that leads to higher production of ·OH to oxidize organic compounds in RO concentrate. DOC removal efficiency of the UV/H₂O₂ process can be also improved by increasing UV lamp intensity [32]. However, increasing the H_2O_2 concentration would be the most efficient way to improve the efficiency of the UV/H₂O₂ process, considering that a dominant portion of energy cost is generally contributed by UV lamp output.

$$H_2O_2 + hv \rightarrow 2 \cdot OH$$
 (5)

3.3. Ozonation

Ozonation was performed to evaluate the removal of DOC in RO concentrate at 50.17, 135, 318.58 mg/L of ozone doses, equivalent to ozone gas diffusing time of 15, 30, and 60 min, respectively. DOC removal efficiency and its regression line at those three ozone doses were depicted (Fig. 2b). DOC removal of RO concentrate was gradually increased from 22.1% to 56.1% by increasing the ozone dose from 50.17 mg/L to 135 mg/L. However, above 135 mg/L of ozone does, insignificant enhancement in removal efficiency was observed (i.e., from 56.1% at 135 mg/L to 61.7% at 318.58 mg/L). This obscure tendency at a high degree of ozone dose shows a disagreement with most of previous studies which reported that an increase in ozone dose leads to a linear enhancement of the removal of organics, such as phenol compounds and

E. coli. [33-35]. The discrepancy between our study and other literatures possibly results from different nature of organic substances in RO concentrate in terms of reactivity with ozone. At the low/intermediate dose levels of ozone, DOC in RO concentrate can be effectively removed through the oxidation of organics with a high reactivity towards ozone, while the high dose levels of ozone are not necessarily resulted in linear enhancement of DOC removal due to the slow oxidative reaction of organics with a low reactivity towards ozone. With this aspect, promoting the generation of \cdot OH, which is a secondary oxidant that was produced during ozone decay, would improve the DOC removal of RO concentrate by ozonation.

3.4. Electrochemical Oxidation

The DOC removal efficiency of RO concentrate by anodic oxidation and the electro-Fenton process was examined under various current density conditions (i.e., current density, j = 5, 10, 20 mA/cm²) and the results were depicted (Fig. 2c and 2d)). Cell voltages were also monitored at each current conditions during the experiment for the cost analysis. Overall, DOC removal efficiency was increased with increasing current densities for both anodic oxidation and electro-Fenton processes. Average cell voltage was proportionally increased in both anodic oxidation (ca. 6.82, 9.47, and 13.6 V for j = 5, 10, and 20 mA/cm², respectively) and electro-Fenton processes (ca. 6.25, 9.09, and 13.1 V for i = 5, 10, and 20 mA/cm², respectively) as the current density was increased. This result shows excellent agreement with previous literatures on the critical role of applied current density during the anodic oxidation and electro-Fenton processes [36-38]. DOC removals by electro-Fenton process were always higher under identical current density conditions as compared to those of anodic oxidation except for j =5 mA/cm² (e.g., 48% and 60% of DOC removal efficiency at j= 20 mA/cm^2 for anodic oxidation and electro-Fenton processes, respectively). In addition, electro-Fenton process exhibited much



Fig. 2. DOC removal efficiency of (a) UV/H₂O₂, (b) ozonation, (c) anodic oxidation, and (d) electro-Fenton processes at different oxidant doses for (a, b) or current densities for (c, d) (oxidant dose and current density were specified at 40% of DOC removal efficiency in each graph).

higher DOC removal efficiency (60% for $j = 20 \text{ mA/cm}^2$) as compared to that of the conventional Fenton process (21% of DOC removal efficiency) under the same experimental conditions except applying potential to the system (Table. 3). Collectively, enhanced oxidation performance in the electro-Fenton process as compared to the anodic oxidation and conventional Fenton processes is a result of greater generation of \cdot OH owing to (i) the combination of homogenous \cdot OH that produced through Fenton reaction (Eq. (2)) and heterogenous BDD(\cdot OH) that produced on the surface of BDD electrode (Eq. (6)) and (ii) acceleration of Fenton reaction by reducing Fe(III) into Fe(II) at cathode.

$$BDD + H_2O \rightarrow BDD(\cdot OH) + H^+ + e^-$$
(6)

3.5. Cost Evaluation

Based on obtained DOC removal data by different AOPs (Table 3), the cost for each AOP was calculated to achieve 40% of DOC removal of RO concentrate in 1 m^3 scale of pilot plants (Table.

4). While 40% of DOC removal by the conventional Fenton process was accomplished under different concentrations of Fe(II) and H_2O_2 , 4.49 mM Fe(II) and 4.81 mM H_2O_2 were selected for the cost calculation, whereby the cost consumption of Fenton's reagents was the lowest. Operating cost for the conventional Fenton process was estimated by considering the chemical expenses including Fenton's reagents (i.e., Fe(II) and H₂O₂) and pH control reagents (i.e., H_2SO_4 and NaOH), as well as the expense for the iron-sludge landfill. Total cost for the conventional Fenton process was calculated to be 1.62 $/m^3$. Operating cost for the UV/H₂O₂ process was estimated based on the consumable expense for H₂O₂ reagent and electrical power consumption for the UV-C lamp at UV dose of 13.88 kJ/L. The total cost for the UV/H_2O_2 process was estimated to be 2.81 \$/m³. Operating cost of ozonation was estimated considering the electrical power consumption of industrial ozone generator in tandem with the oxygen generator and calculated to be 0.37 \$/m³. Operating cost of electro-Fenton process was calculated based on the electricity cost for applying potential to the BDD electrode

Table 3. DOC remova	al in RO concent	trate from a wastewate	er reclamation plan	it by various AOPs				
AOP	Ηq	[Fe(II)] ₀ (mM)	[H ₂ O ₂] ₀ (mM)	UV-C Dose (kJ/L)	Current Density, <i>j</i> [mA/cm ²]	Reaction time (h)	O ₃ Dose (mg/L)	DOC removal [%]
	3.0	1.0	1.5	× 1		1.0		19.5
	3.0	1.0	4.5	•		1.0	ı	22.0
	3.0	1.0	7.5			1.0	ı	21.0
-	3.0	3.0	1.5			1.0	·	34.0
Conventional	3.0	3.0	4.5			1.0	·	36.1
renton	3.0	3.0	7.5			1.0	ı	35.0
	3.0	5.0	1.5			1.0		35.3
	3.0	5.0	4.5	•		1.0	ı	40.1
	3.0	5.0	7.5			1.0		40.0
	3.0		4.5	13.88	•	2.0	·	14.0
U V/H2U2	3.0		45.0	13.88		2.0	·	61.0
	7.8	•	•	•	•	0.25	50.17	22.1
Ozonation	7.8					0.5	135	56.1
	7.8					1.0	318.58	61.7
V	7.8				5	1.0	·	23.7
Allouic	7.8				10	1.0		33.0
OXIMINI	7.8	ı			20	1.0	ı	48.0
	3.0	1.0	7.5		Ð	1.0	·	17.0
Electro-Fenton	3.0	1.0	7.5		10	1.0		48.0
	3.0	1 0	7.5	I	20	1 0	,	60.0

(8.54 V) as well as all of the expense for the conventional Fenton process (aforementioned chemical and sludge landfill expenses), while only the electrical energy consumption to the BDD electrode (11.37 V) was considered for the operating cost of anodic oxidation. Interestingly, the total operating cost of anodic oxidation (6.53 \$/m³) was higher as compared to that of electro-Fenton process (3.18 \$/m³), indicating the electricity cost is a dominant cost driver in Korea as compared to other chemical and sludge landfill expenses. Conclusively, the ozonation process was verified as the

Table 4. Operating cost comparis	son of five	e different	AOPs
----------------------------------	-------------	-------------	------

removal among tested AOPs.

most cost-efficient process in terms of the operating costs for DOC

AOPs	Specific operating condition for 40% removal of DOC	Cost for 40% removal of DOC in RO concentrate (\$/m ³)
Conventio	$[Fe(II)] = 4.49 \text{ mM}, \ [H_2O_2] = 4.81 \text{ mM}$	
nal Fenton	Initial pH = 3.0, Volume = 100 mL Reaction time = 1 h	1.62
	$[H_2O_2] = 26.9 \text{ mM}$	
UV/H_2O_2	UV dose = 13.88 kJ/L	2.81
	Initial pH = 7.8, Volume = 100 mL	
	Reaction time = $2 h$	
	$[O_3] = 86.97 \text{ mg/L}$	
Ozonation	Initial pH = 7.8, Volume = 100 mL	0.37
	Reaction time = 0.36 h	
	Anode: BDD, Cathode: SS	
	Current Density = 14.84 mA/cm^2	
Anodic	Average Voltage = 11.37 V	6.53
Oxidation	Initial pH = 7.8 , Volume = 25 mL	
	Reaction time = 1 h	
	Anode: BDD, Cathode: SS	
F I .	Current Density = 9.58 mA/cm^2	0.40
Electro-	Average Voltage = 8.54 V	3.18
Fenton	$[Fe(II)] = 1.0 \text{ mM}, [H_2O_2] = 7.5 \text{ mM}$	
	Initial pH = 3.0 , Volume = 25 mL	
	Reaction time = $1 h$	

4. Conclusions

Five different AOPs (i.e., the conventional Fenton, UV/H₂O₂ process, ozonation, anodic oxidation, and electro-Fenton processes) were successfully validated for their oxidation capacity for the treatment of RO concentrate from a wastewater reclamation plant, resulting in 14% ~ 61.7% of DOC removal depending on the applied process and experimental conditions. Based on the operation cost analysis, ozonation and followed by the conventional Fenton process were identified as the most cost-efficient process. Meanwhile, the processes (i.e., UV/H₂O₂ process, anodic oxidation, and electro-Fenton processes) that require electrical energy consumption were evaluated as a less cost-effective process as compared to the conventional Fenton and ozonation processes.

Acknowledgments

This research has been performed as Project No Open Innovation R&D (Together PRO) supported by K-water and by a Korean Environment Industry & Technology Institute (KEITI) through Prospective Green Technology Innovation Project (20200031 60008).

Conflict-of-Interests

The authors declare that they have no conflict of interest.

Author Contributions

B.J.K (Ph.D. student) conceived and designed the study, performed the experiments, wrote, and edited the manuscript. H.-J.L. (Ph.D.) and J.Y.C. (Researcher) reviewed and edited the manuscript. C.L. (Professor) conceived and designed the study, reviewed, and edited the manuscript. All authors read and approved the manuscript.

References

- Miller GW. Integrated concepts in water reuse: managing global water needs. *Desalination*. 2006;187;65-75. https://doi.org/10.1016/j.desal.2005.04.068.
- Gude VG. Desalination and water reuse to address global water scarcity. *Rev. Environ. Sci. Biotechnol.* 2017;16;591-609. https://doi.org/10.1007/s11157-017-9449-7.
- Jeong H, Seong C, Jang T, Park. Classification of wastewater reuse for agriculture: A case study in south korea. *Irrig. Drain.* 2016;65;76-85. https://doi.org/10.1002/ird.2053.
- Jang T, Lee SB, Sung CH, Lee HP, Park SW. Safe application of reclaimed water reuse for agriculture in Korea. *Paddy Water Environ*. 2010;8;227-233. https://doi.org/10.1007/s10333-010-0203-9.
- Abdel-Jawd M, Al-Shammari S, Al-Sulaimi J. Non-conventional treatment of treated municipal wastewater for reverse osmosis. *Desalination*. 2002;142;11-18. https://doi.org/10.1016/S0011-9164(01)00421-0.
- Al-Rifai JH, Gabelish CL, Schafer AL. Occurrence of pharmaceutically active and non-steroidal estrogenic compounds in three different wastewater recycling schemes in Australia. *Chemosphere*. 2007;69;803-815. https://doi.org/10.1016/j.chemosphere. 2007.04.069.
- Kazner C, Jamil S, Phuntsho S, Shon HK. Forward osmosis for the treatment of reverse osmosis concentrate from water reclamation: process performance and fouling control. *Water Sci. Technol.* 2014;69(12);2431-2437. https://doi.org/10.2166/ wst.2014.138.
- Xiang Q, Nomura Y, Fukahori S, Mizuno T, Tanaka H, Fujiwara T. Innovative Treatment of Organic Contaminants in Reverse Osmosis Concentrate from Water Reuse: a Mini Review. *Curr. Pollut. Rep.* 2019;5;294-307. https://doi.org/10.1007/s40726-

019-00119-2.

- Buxton GV, Greenstock CL, Helman WP. Critical Review of rate constants for reactions of hydrated electrons, hydrogen atoms and hydroxyl radicals (•OH/•O') in Aqueous Solution. J. Phys Chem Red Data. 1988;17(2);513-886. https://doi.org/ 10.1063/1.555805.
- Kim I, Lee J. Comparison of ozonation removal for PPCPs in secondary treated sewage by microbubble generator and ejector. *Environ. Eng. Res.* 2022;27(2). DOI: https://doi.org/10.4491/ eer.2020.163.
- Jiao Z, Gong H, Peng Y et al. Application of copper sulfate based fenton-like catalyst in degradation of quinoline. *Environ. Eng. Res.* 2022;27(5). DOI: https://doi.org/10.4491/eer.2021.305.
- 12. Liu K, Roddick FA, Fan L. Potential of UV/ H_2O_2 oxidation for enhancing the biodegradability of municipal reverse osmosis concentrates. *Water Sci. Technol.* 2011;63(11);2605-2611. https://doi.org/10.2166/wst.2011.465.
- Chen M, Zhao X, Wang C, Pan S, Zhang C, Wang Y. Electrochemical oxidation of reverse osmosis concentrates using macroporous Ti-ENTA/SnO₂-Sb flow-through anode: Degradation performance, energy efficiency and toxicity assessment. J. Hazard. Mater. 2021;401;122395. https://doi.org/10.1016/j.jhazmat.2020.123295.
- Umar M. Roddick F, Fan L. Recent Advancements in the Treatment of Municipal Wastewater Reverse Osmosis Concentrate-An Overview. *Crit Rev Environ Sci Technol.* 2015;45;193-248. https://doi.org/10.1080/10643389.2013.852378.
- Cemre Birben N. Uyguner-Demirel CS, Bekbolet M. Organic Matrix in Reverse Osmosis Concentrate: Composition and Treatment Alternatives. *Curr. Org. Chem.* 2017;21;1084-1097. https://doi.org/10.2174/1385272821666170102151901.
- Westerhoff P, Moon H, Minakata D, Crittenden J. Oxidation of organics in retentates from reverse osmosis wastewater reuse facilities. *Water Res.* 2009;43;3992-3998. https://doi.org/10. 1016/j.watres.2009.04.10.
- Perez-Gonzalez A, Urtiaga AM, Ibanez R, Ortiz I. State of the art and review on the treatment technologies of water reverse osmosis concentrates. *Water Res.* 2012;46;267-283. https://doi.org/10.1016/j.watres.2011.10.046.
- Cai QQ, Wu MY, Li R, Deng SH, Lee BCY, Ong SL, Hu J.Y. Potential of combined advanced oxidation – Biological process for cost-effective organic matters removal in reverse osmosis concentrate produced from industrial wastewater reclamation: Screening of AOP Pre-treatment technologies. *Chem. Eng.* 2020;389;1-12. https://doi.org/10.1016/j.cej.2019.123419.
- Zhou T, Lim TT, Chin SS, Fane AG. Treatment of organics in reverse osmosis concentrate from a municipal wastewater reclamation plant: Feasibility test of advanced oxidation processes with/without pretreatment. *Chem. Eng.* 2011;166;932-939. https://doi.org/10.1016/j.cej.2010.11.078.
- Zhang A, Gu Z, Chen W, Li Q, Jiang G. Removal of refractory organic pollutants in reverse-osmosis concentrated leachate by Microwave-Fenton process. *Environ. Sci. Pollut. Res.* 2018;25;28907-28916. https://doi.org/10.1007/s11356-018-2900-7.
- 21. Hermosilla D, Merayo N, Ordonez R, Blanco A. Optimization of conventional Fenton and ultraviolet-assisted oxidation processes for the treatment of reverse osmosis retentate from a

paper mill. Waste Manage. 2012;32;1236-1243. https://doi.org/10.1016/j.wasman.2011.12.011.

- 22. Wegner EE, Adamson AW. Photochemistry of complex ions. III. Absolute Quantum Yields for the Photolysis of Some Aqueous Chromium(III) Complexes. Chemical Actinometry in the Long Wavelength Visible Region. J. Am. Chem. Soc. 1966;88:3,394-403. No DOI.
- Eisenberg G. Colorimetric determination of hydrogen peroxide. Ind. Eng. Chem., Anal. Ed. 1943;15(5);327-328. No DOI.
- Bader H, Hoigne J. Determination of ozone in water by the indigo method. Water Res. 1981;15(4);449-456. https://doi.org/10.1016/0043-1354(81)90054-3.
- Korea Price Information. Cost of FeSO₄·7H₂O, NaOH,H₂SO₄, NaOH. [Internet]. KPI; c2020 [cited 20 Octobor 2020]. Available from : http://www.kpi.or.kr/www/.
- Korea Electric Power Corporation. Cost of industrial electricity at WWTP. [Internet]. KEPCO; c2020 [cited 20 Octobor 2020].
- 27. Ozonetech. Cost of industrial ozone generator. [Internet]. Ozonetech; c2020 [cited 20 Octobor 2020]. Available from : http://ozonetech.co.kr/OZbbs/board.php?bo_table= oz_prod1&wr_id=15.
- 28. Lee JI. Method for Efficient Treatment of Sewage Sludge. 1st ed. Gyeonggi Research Institute; 2008. P. 23-24.
- Su CC, Chang AT, Bellotindos LM, Lu MC. Degradation of acetaminophen by Fenton and electro-Fenton Processes in aerator reactor. Sep. Purif. Technol. 2012;99;8-13. https://doi.org/10.1016/j.seppur.2012.07.004.
- Muruganandham M, Swaminathan M. Decolourisation of Reactive Orange 4 by Fenton and photo-Fenton oxidation technology. *Dyes Pigm.* 2004;63;315-321. https://doi.org/10. 1016/j.dyepig.2004.03.004.
- 31. Mohajeri S, Aziz HA, Isa MH, Bashir MJK, Mohajeri L, Adlan

MN. Influence of Fenton reagent oxidation on mineralization and decolorization of municipal landfill leachate. J. *Environ. Sci. Health A.* 2010;45;692-698. https://doi.org/10.1080/10934 521003648883.

- Malvestiti JA, Fagnani E, Simao D, Dantas RF. Optimization of UV/H₂O₂ and ozone wastewater treatment by the experimental design methodology. *Environ. Technol.* 2018;40(15); 1910-1922. https://doi.org/10.1080/09593330.2018.1432698.
- Deborde M, Rabouan S, Mazellier P, Dugnet JP, Legube B. Oxidation of bisphenol A by ozone in aqueous solution. *Water Res.* 2008;42;4299-4308. https://doi.org/10.1016/j.watres.2008. 07.015.
- 34. Paraskeva P, Graham NJD. Treatment of a secondary municipal effluent by ozone, UV and microfiltration: microbial reduction and effect on effluent quality. *Desalination*. 2005;186;47-56. https://doi.org/10.1016/j.desal.2005.04.057.
- 35. Zhang J, Dong B, Han Y, Zhan X, Ge S, He S. Phenol Removal Performance and Mechanism Using Catalytic Ozonation with the Catalyst of Cobalt-doped α-MnO₂. *Environ. Eng. Res.* 2023;28(3). https://doi.org/10.4491/eer.2022.156.
- Zhou M, Liu L, Jiao Y, Wang Q, Tan Q. Treatment of high-salinity reverse osmosis concentrate by electrochemical oxidation on BDD and DSA electrodes. *Desalination*. 2011;277;210-206. https://doi.org/10.1016/j.desal.2011.04.030.
- 37. Maharaja P, Boopathy R, Karthikeyan S. Advanced oxidation of catechol in reverse osmosis concentrate generated in leather wastewater by Cu-graphite electrode. *Int. J. Environ. Sci. Technol.* 2016;13;2143-2152. https://doi.org/10.1007/s13762-016-1044-x.
- Mohan N, Balasubramania N, Basha CA. Electrochemical oxidation of textile wastewater and its reuse. J. Hazard. Mater. 2007;147;644-651. https://doi.org/10.1016/j.hazmat.2007.01.063.