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Advanced Oxidation Processes and Nanofiltration to Reduce the Color and Chemical Oxygen Demand of Waste Soy Sauce

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Abstract: Currently, the ozone (O_3) oxidation efficiency in the treatment of waste soy sauce provides 34.2% color removal and a 27.4% reduction in its chemical oxygen demand (COD). To improve the O_3 oxidation efficiency, hydrogen peroxide (H_2O_2) is used to cause a H_2O_2/O_3 process. In H_2O_2/O_3 process experiments, a previously optimized pH of 11 and applied O_3 dose of 50 mg L^{-1} were used and the H_2O_2/O_3 ratio was varied between 0.1 and 0.9 in intervals of 0.2. The results show that an H_2O_2/O_3 ratio of 0.3 results in the highest efficiencies in terms of color removal (51.6%) and COD reduction (33.8%). Nanofiltration (NF) was used to pretreat the waste soy sauce to improve color removal and COD reduction. The results showed that NF with an NE-70 membrane results in 80.8% color removal and 79.6% COD reduction. Finally, the combination of NF and H_2O_2/O_3 process resulted in the best treatment efficiency: 98.1% color removal and 98.2% COD reduction. Thus, NF & H_2O_2/O_3 process can be considered as one of the best treatment methods for waste soy sauce, which requires high intrinsic color removal and COD reduction efficiencies.

Keywords: waste soy sauce; AOPs; H_2O_2/O_3 process; oxidation; nanofiltration; membrane

1. Introduction

Waste soy sauce has a high chemical oxygen demand (COD). It has an intense dark brown color due to the presence of caramel pigments and the occurrence of the Maillard reactions producing melanin or melanoidin [1,2]. Several processes are typically used to treat soy sauce wastewater including nitrification and denitrification, an activated sludge process, and biological treatments. However, it is difficult to control the stability of the soy sauce treatment processes because of annual fluctuations in the amount of waste soy sauce generated (47,914 tons were produced in 2016 compared to 37,232 tons in 2015 and 40,461 tons in 2014) [3]. Biological treatment removes only a small fraction of the organic matter that contributes to the color of the effluents [4–6]. Therefore, the wastewater discharged to the environment is colored, which is aesthetically displeasing; moreover, the wastewater could affect the ecosystem due to its non-biodegradability and recalcitrance [7]. Therefore, it is necessary to decolorize waste soy sauce before it is discharged.

In our earlier study, overcoming the limitations of ozone (O_3) oxidation to reduce the color and COD of waste soy sauce was intensively investigated [8]. Up to 34.2% color removal and 27.4% COD reduction were achieved by O_3 oxidation at a pH of 11 with an applied O_3 dose of 50 mg L^{-1} [8]. However, the color removal and COD reduction were not thoroughly investigated as the salt water regenerated from the waste soy sauce was reused. Therefore, a combination of different techniques must be considered as an alternative treatment method for removing the color and reducing the COD of waste soy sauce as completely as possible. In addition, a high concentration of salt can be recovered from waste soy sauce through the use of advanced technologies.

Many researchers conduct O_3 oxidation by adding hydrogen peroxide (H_2O_2) to boost oxidation to raise efficiency of the color removal and COD reduction in wastewater. Fahmi, M. R et al. reported that advanced oxidation processes (AOPs) better facilitate the removal of reactive red 120 than ozonation [9]. This indicates that H_2O_2 accelerates O_3 activation by forming hydroxyl radicals, which quickly oxidize reactive red 120 [9]. Rollon, A. A. et al. reported that the AOPs combination of H_2O_2 and O_3 are most effective, followed by UV/H_2O_2-treatment, UV/O_3-treatment, O_3-treatment, and UV-C treatment (which is the least effective) [10]. However, it is difficult to improve the efficiency of AOPs without creating oxidative conditions. This is because the O_3 oxidation at the optimum applied O_3 dose involves a large number of hydroxyl radicals reacting instantaneously with O_3-demanding species (the target substances) [8]. In other words, there is a high instantaneous ozone demand (IOD) in high concentration COD products such as waste soy sauce. Therefore, a pretreatment is required to overcome this IOD that is generated during oxidation.

Other researchers have proposed the use of membrane-based filtration techniques to remove color and reduce the COD of high-organic-content wastewater. Zheng, Y. et al. reported that under optimum conditions (pressure of 0.8 bar and volume-concentrating factor of 4.0), a submerged nanofiltration (NF) system exhibited a steady permeate flux of 5.15 L m^{-2} h, a color reduction of 99.3%, and a COD reduction of 91.5% [11]. Abid, M. F. et al. (2012) achieved 93.77%, 95.67%, and 97% removal of red, black and blue dyes using a NF membrane under a pressure of 8 bar [12]. However, it is difficult to remove the color and organic matter completely using typical membrane processes other than reverse osmosis because chromophoric dissolved organic matter generally has various molecular weights [13–15]. Furthermore, despite research progress to date, membrane filtration has yet to be applied in removing the intense color to reduce the high COD of waste soy sauce. Membrane systems have been considered promising for the pretreatment of waste soy sauce before oxidation. However, there remain some technological challenges to be solved for this application, including developing a physicochemical technique of membrane filtration and an oxidation method that can overcome the limitations of biological treatments. In this study, we applied H_2O_2/O_3 process to enhance color removal and COD reduction by oxidation and the H_2O_2/O_3 ratio was optimized. In addition, NF was combined with H_2O_2/O_3 process to maximize the color removal and COD reduction. Herein, we identify the best method for treating waste soy sauce from five different treatment methods that will guide future wastewater treatment strategies to allow the saline wastewater to be reused and mitigate the impact on the environment when it is discharged.

2. Materials and Methods

2.1. Sampling Procedure

The waste soy sauce used in this study was provided by Monggo Foods, Inc. (Changwon-si, Gyeongsangnam-do, Korea).

2.2. Characterization

All samples were analyzed after removing the impurities with a 1.2 μm GF/C filter (Whatman). A seven compact™ pH/Ion S220 instrument (Mettler-Toledo GmbH, Greifensee, Switzerland) was used to measure the pH. The biochemical oxygen demand (BOD_5), chemical oxygen demand (COD_{cr}), total nitrogen (TN), and total phosphorus (TP) were measured using a US/DR3900 (320−1100 nm, Hach Company, Düsseldorf, Germany) kit assay according to the methods for testing water pollution set forth by the Ministry of Environment of Korea. The total organic carbon (TOC) was analyzed using a Shimadzu JP/TOC−5000A (Kyoto, Japan). The color was calculated as the transmittance of wavelength at 390, 400, 456 nm as measured using an Agilent Cary60 UV−VIS spectrophotometer (Santa Clara, CA, USA). The salinity was measured using an SB1500PRO instrument (0%−10%, HM Digital, Seoul, Korea) and a table salinometer.

2.3. Size Exclusion Chromatography (SEC)

SEC analysis was performed with high performance liquid chromatography (HPLC, LC600 Shimadzu) with UVA (SPD-6A Shimadzu) and DOC (Modified Sievers Turbo total organic carbon analyzer) detectors. The columns employed were a TSK-50S column, a Polyacrylamide Bio-Gel P-6 column, and a Waters Protein-Pak 125 silica column (Table 1) [16].

Table 1. Characteristics of tested columns.

Type	Column Packing	Particle Size (μm)	Separation Range (Da)	Column Size (cm)
Hydroxylated organic	Protein PAK 125	10	1000–30,000	0.78 × 30
Polyacrylamide	TSK-50S	30	$<5 \times 10^6$	2 × 25
Silica	Bio-Gel P-6	90–180	1000–6000	0.5 × 90

Molecules that were larger than the pore size of the packing material were excluded and eluted first, at the void volume. Smaller molecules were able to penetrate through the porous infrastructure and were attenuated, corresponding to a higher retention time [16].

2.4. H_2O_2/O_3 Process

A schematic of the benchtop-scale reactor system used herein is shown in Figure 1. Experimental runs were performed in a 1 L Pyrex reactor with 500 mL of sample. The reactor was filled with 500 mL of waste soy sauce and agitated with a magnetic stirrer at 150 rpm. The optimum conditions for color removal and COD reduction by O_3-based oxidation were a pH of 11 and an applied O_3 concentration of 50 mg L^{-1} [8]. The O_3-containing gas was supplied continuously for 30 min through a gas diffuser at the bottom of the reactor. An O_3 trap containing a 2% potassium iodide solution was connected in series with the reactor to verify the O_3 gas concentration in the outlet gas stream. A 0.1 N $Na_2S_2O_3$ (sodium thiosulfate) solution was used as the reducing agent for the reverse titration in the trap. Subsequently, 0.1 N H_2SO_4 (sulfuric acid) was used to facilitate the reaction of the O_3 in the liquid phase with the I_2 [8]. All experiments were performed in duplicate at room temperature under a fume hood (for safety due to the presence of O_3 gas). The pH, applied O_3 concentration, and reaction time were kept constant while the H_2O_2/O_3 ratio was varied between 0.1 and 0.9 in intervals of 0.2. Table 2 summarizes the parameters for the H_2O_2/O_3 process experiments. In addition, the calculated vent gas of ozone by Equation (1) is shown in Table 3. The resulting color change and COD reduction were observed in 50 mL samples collected from the supernatant.

$$\frac{[a]\mathrm{eqNa_2S_2O_3}}{\mathrm{L}}\,\frac{1\mathrm{eqO_3}}{2\mathrm{eqNa_2S_2O_3}}\,\frac{48\mathrm{gO_3}}{1\mathrm{eqO_3}}\,\frac{[b]\mathrm{mL}}{[c]\mathrm{min}}\,\frac{1}{\mathrm{Reactor\ volume\ (L)}} \tag{1}$$

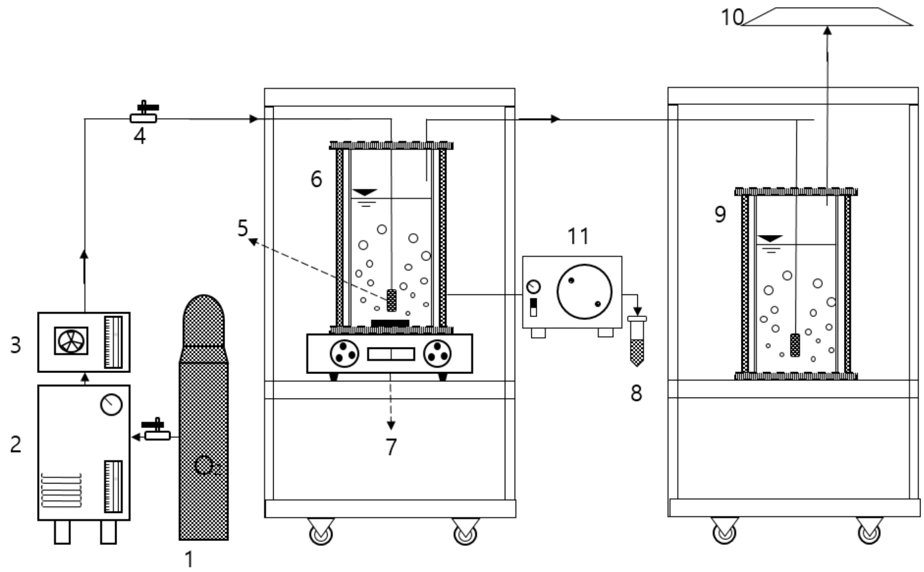
where [a]: $Na_2S_2O_3$ concentration, [b]: $Na_2S_2O_3$ consumption, [c]: Contact time.

Table 2. The experimental conditions for the H_2O_2/O_3 process.

Parameter	Value
pH	11
Applied O_3 conc. (mg L^{-1})	50
H_2O_2/O_3 ratio (wt/wt)	0.1, 0.3, 0.5, 0.7, and 0.9
Reaction time (min)	30
Mixing speed (rpm)	150
Sample volume (mL)	500

Table 3. Quantitative assessment of O_3 consumption.

Parameter	1st Experiment	2nd Experiment
$Na_2S_2O_3$ conc.	0.1	
$Na_2S_2O_3$ consumption	1.4	2.9
Vent O_3 conc. (mg L^{-1} min^{-1})	1.12	2.32



1. Oxygen gas
2. Ozone generator
3. Injection ozone monitor
4. Ozone injection valve
5. Diffuser
6. Oxidation reactor
7. Magnetic stirrer
8. Sampling port
9. Trap solution (KI)
10. Fume hood
11. Peristaltic pump

Figure 1. Schematic of the O_3-oxidation experimental system.

2.5. Nanofiltration (NF) System

The NF membranes used in this study were thin-film composite NF membranes, NE-70 and NE-90 (Toray Chemical, Korea). These membranes have different top layers, zeta potentials, wettabilities, and roughnesses as detailed in Table 4. The characteristics of the NF membrane were investigated in previous study [17].

Table 4. The characteristics of the NF membrane.

Membrane	Material	MWCO (Da)	Zeta Potential at pH 7 (mV)	Contact Angle (°)	Roughness (nm)
NE-70	Sulfonated polyethersulfone	350	–47.2	22.6 ± 1.9	8.69
NE-90	Meta-phenylene diamine	210	–38.7	41.5 ± 3.7	48

A laboratory-scale dead-end NF membrane was used in this experiment. The membrane surface area was 0.015 m^2. Waste soy sauce was fed into the filtration module by a gear pump. The filtration experiments were performed with a commercial NF module. Figure 2 shows a schematic of the NF system. In all experiments, a pressure of 1.5 MPa (15 bar) was applied at room temperature. The evolution of flux and rejection progressed over a period of 6 hours. For analysis and comparison, the measurements were taken after 1 h of filtration.

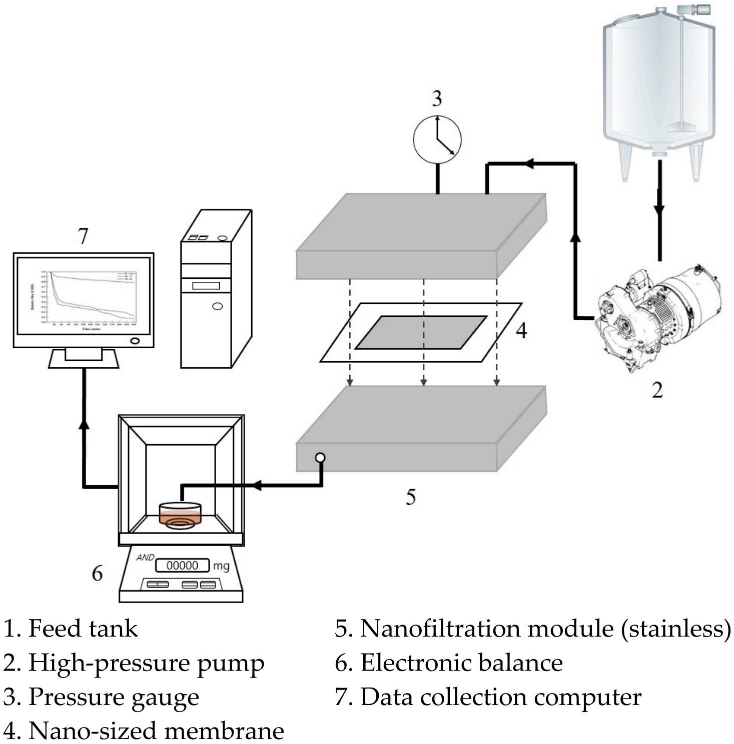


Figure 2. Schematic of the NF experimental system.

3. Results and Discussion

3.1. Characteristics of Waste Soy Sauce

The chemical properties of the sample were evaluated according to the guidelines for testing water pollution developed by the Ministry of Environment of Korea [18]. Table 5 shows the characteristics of the waste soy sauce. The pH value of 4.6 indicates that the waste soy sauce was acidic. The color was found to be 3810 TCU and the COD was measured to be 231.5 g/L. These results are similar to those reported in a previous study: 4038 TCU color and 229.1 g/L COD [8]. The T-N and T-P concentrations were measured to be 10.4 and 2.8 g/L, respectively. The salinity, 16.4%, was much higher than that found in other types of organic wastewater. In general, the high-salinity wastewater does not exhibit high removal efficiencies with biological treatment systems. This is because the performances of biological treatment processes are adversely affected by the negative effects of salt on microbial flora [19]. Moreover, the TOC, which is used as an indicator of water pollution, was found to be 57.6 g/L.

Table 5. Chemical characteristics of waste soy sauce.

Parameter	Value
pH	4.4 ± 0.2
[1] COD_{cr} (g/L)	231.5 ± 0.9
[2] BOD_5 (g/L)	129.4 ± 6.6
[3] TN (g/L)	10.4 ± 0.7
[4] TP (g/L)	2.8 ± 0.3
[5] TOC (g/L)	57.6 ± 2.7
Salinity (%)	16.4 ± 0.2
Color (TCU)	3810 ± 130

[1] COD: Chemical oxygen demand; [2] BOD: Biochemical oxygen demand; [3] TN: Total nitrogen; [4] TP: Total phosphorus; [5] TOC: Total organic carbon.

3.2. Optimization of H_2O_2/O_3 Ratio

Our previous study revealed that a pH of 11 and an applied O_3 concentration of 50 mg L^{-1} are optimal for color removal and COD reduction; these conditions were used in this study. H_2O_2 was combined with O_3 to accelerate the oxidation of the organic molecules present in the wastewater. The H_2O_2/O_3 ratio was varied between 0.1 and 0.9 to optimize the color removal by H_2O_2/O_3 process; the results of this experiment are shown in Figure 3. As the H_2O_2/O_3 ratio increased from 0.1 to 0.3, the color removal increased gradually from 41.2 to 51.6%. The H_2O_2/O_3 ratio of 0.3 resulted in the greatest color removal. The color removal decreased sharply from 51.6 to 17.1% as the H_2O_2/O_3 ratio increased above 0.3.

The lower residual color after oxidation with H_2O_2/O_3 as the H_2O_2/O_3 ratio increased was attributed to the increased formation of OH radicals [20–23]. Other studies have reported increased H_2O_2/O_3 ratios and biodegradability after wastewater treatment with O_3 and H_2O_2 [24,25]. The decreased color removal with H_2O_2/O_3 ratios above 0.3 was attributed to the strong scavenging effects of carbonate ($CO_3{}^{2-}$) and bicarbonate ($HCO_3{}^{-}$). The results showed that an H_2O_2/O_3 ratio of 0.3 was optimal for color removal (Yielding 51.6% color reduction) because of the high oxygenation capacity resulting from the suitable amount of hydroxyl radicals. Thus, it was confirmed that the H_2O_2/O_3 process are more efficient (51.6%) in removing color than in O_3-based oxidation (34.2%) [8].

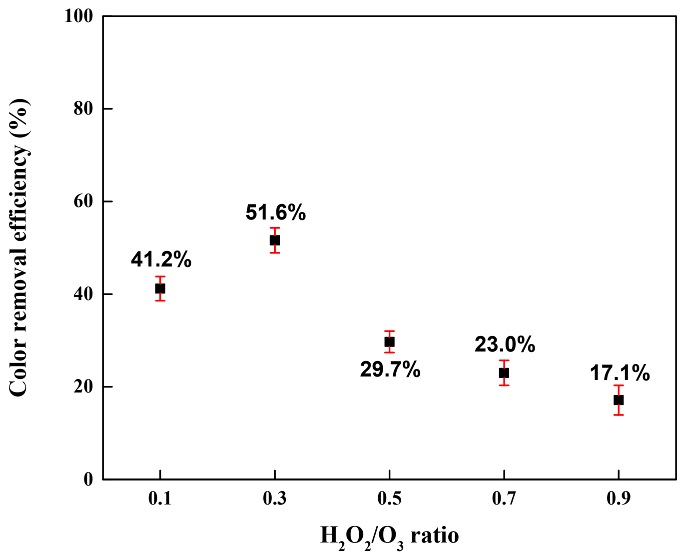


Figure 3. Removal efficiency of color with various H_2O_2/O_3 ratios during H_2O_2/O_3 process.

Figure 4 shows the influence of the H_2O_2/O_3 ratio between 0.1 and 0.9 on the COD reduction due to the O_3 treatment of waste soy sauce. The results show that the COD reduction as a function of the H_2O_2/O_3 ratio followed a similar trend to that of the color removal shown in Figure 3. The COD reduction was optimal (a 73.6 g/L reduction from 217.73 g/L) at an H_2O_2/O_3 ratio of 0.3. This result confirms that a sufficient amount of OH radicals were produced at an H_2O_2/O_3 ratio of 0.3. The H_2O_2 addition facilitated the production of OH radicals, resulting in a synergistic effect between the applied O_3 concentration and the COD reduction [26]. The COD reduction effect decreased distinctly from 33.8 to 16.6% as the H_2O_2/O_3 ratio increased above 0.3 as observed with the color removal. This was likely due to the OH radicals being consumed by the excessive amounts of H_2O_2 [27–29].

The results shown in Figures 3 and 4 confirm that the color removal and COD reduction strongly depend on the H_2O_2/O_3 ratio. However, the H_2O_2/O_3 ratio influences color removal more than COD reduction. Moreover, an H_2O_2/O_3 ratio of 0.3 was the most effective in terms of both color removal and COD reduction.

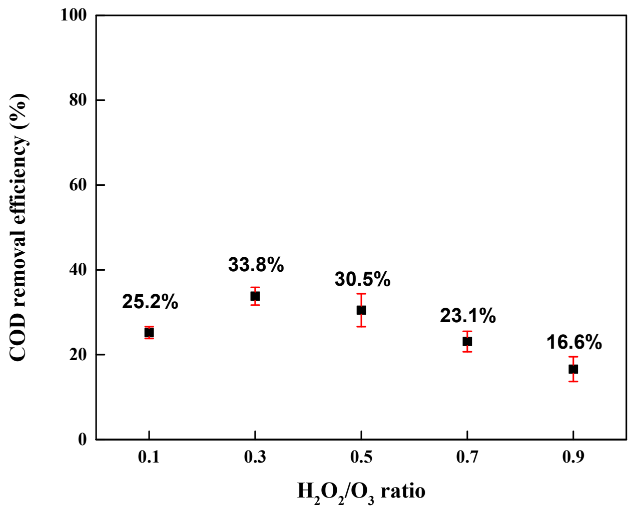


Figure 4. Removal efficiency of COD with various H_2O_2/O_3 ratios during H_2O_2/O_3 process.

3.3. Nanofiltration (NF) System

3.3.1. Size Exclusion Chromatography Analysis

The COD reduction was not significant despite the use of a H_2O_2/O_3 process because the COD of waste soy sauce is considerably higher than that of other types of waste and wastewater. In addition, there is a high IOD due to the high COD of the waste soy sauce. Thus, the initial COD of waste soy sauce makes it unsuitable for the oxidation treatment [30]. In addition, it is known that H_2O_2 interferes with the COD reduction by this process by consuming oxidation agents including potassium dichromate [31]. Thus, the molecular weight distribution of the total organic carbon was measured to determine the best membrane-based filtration pretreatment in the interest of improving the COD reduction effect of the H_2O_2/O_3 process.

Figure 5 shows the molecular weight distribution in the waste soy sauce as measured via size-exclusion chromatography (SEC) using a method for detecting organic matter wherein the adsorption of the sample was measured at an ultraviolet (254 nm) wavelength [32]. The main peak shows the molecular weight of the organic matter ranged 750–2200 dalton.

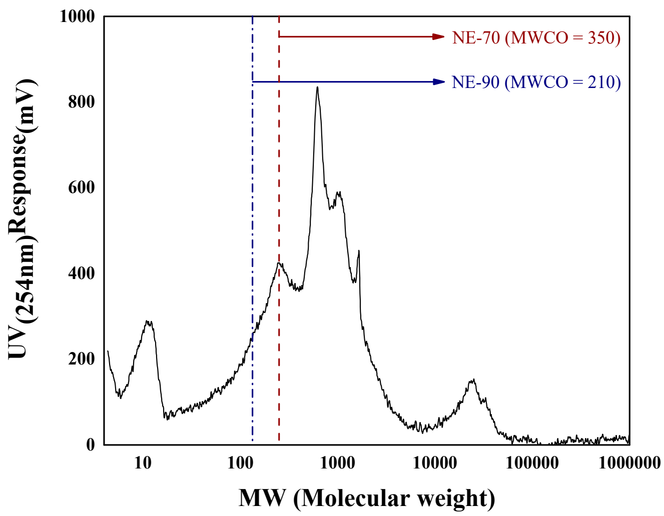


Figure 5. Molecular weight distribution of waste soy sauce.

From this data, the appropriate molecular weight cutoff (MWCO) for the NF membranes (NE-70 and NE-90) was selected to approve the COD reduction effect by H_2O_2/O_3 process. To facilitate stable treatment by oxidation, the COD reduction by the membrane pretreatment was calculated to be over 70%. Thus, experiments were conducted using two membranes, NE-70 (71.8% of the expected COD removal) and NE-90 (73.2% of expected COD removal), confirming the effectiveness of the pretreatment in terms of color removal and COD reduction.

3.3.2. Color Removal and COD Reduction by Nanofiltration (NF)

In our earlier study, O_3-based oxidation exhibited low color removal (34.2%) and COD reduction (27.4%) efficiencies at pH 11 with an O_3 injection dose of 50 mg L^{-1} [8]. In this study, we used H_2O_2/O_3 process and found that the removal efficiency was higher than that with than O_3-based oxidation under the same conditions. However, the color removal and COD reduction were not complete even with H_2O_2/O_3 process. Thus, we applied NF as a pretreatment to enhance the color removal and COD reduction by oxidation method.

Table 6 shows that color removal and COD reduction by NF was similar with the NE-70 and NE-90 membranes. The results show that the color removal and COD reduction were similar with both membranes even though the MWCO of the NE-90 membrane (210 daltons) is lower than that of the NE-70 membrane (350 daltons). The NE-90 membrane, which yielded a color reduction of 81.3% and a COD reduction of 80.7%, was slightly more effective for removing color and reducing the COD than the NE-70 membrane, which yielded a color reduction of 80.8% and a COD of 79.6%. However, the NE-70 is of sustainability usable than NE-90 for experiment. It can further be concluded that the NE-70 membrane is suitable as a pretreatment for waste soy sauce treatment as it improves the color removal and COD reduction.

Table 6. Color removal and COD reduction by nanofiltration (NF).

Classification	Type	Amount of Removal (Removal Efficiency)	
		Color (TCU)	COD (g/L)
Treated waste soy sauce (Removal efficiency)	NE-70	2908.8 (80.8%)	173.3 (79.6%)
	NE-90	2926.8 (81.3%)	175.7 (80.7%)

3.3.3. Flux Variation

To predict the lifetime of the nanofiltration (NF) membranes for waste soy sauce treatment, flux experiments were conducted. Figure 6 shows the flux decline of the two NF membranes during 4 hours of operation. The flux decreased sharply initially then steadily decreased after 45 min of operation. The flux of the NE-70 membrane was lower than that of the NE-90 membrane. This was attributed to the properties of the membranes: NE-90 has a higher contact angle (41.5 ± 3.7°) than NE-70 (22.6 ± 1.9°), showing that its surface is more hydrophobic [17,33,34]; this would cause hydrophobic organic compounds to adsorb onto the surface [17]. In addition, NE-90 has a higher surface roughness than NE-70, which results in a greater repulsive force between the membrane surface and the solute and, thus, a lower solute permeability [17].

These results show that NE-70 is more suitable for this application as its flux decline is lesser during the waste soy sauce treatment than that of NE-90 although the color removal and COD reduction were similar for the two membranes.

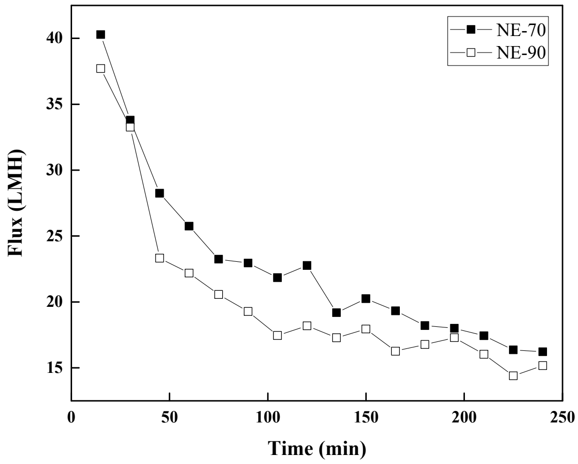


Figure 6. Flux decline of NE-70 and NE-90 in filtration of waste soy sauce.

3.3.4. Comparison of Treatment Methods

To enhance the effectiveness of the O_3 oxidation, we aimed to increase the OH radical production by adding H_2O_2 and applying NF as a pretreatment to overcome the limitation (i.e., the high COD concentration) of the H_2O_2/O_3 process. Figure 7 and Table 7 show comparisons of color removal and COD reduction obtained by various treatment methods for waste soy sauce treatment. The results show that the NF & H_2O_2/O_3 process resulted in the greatest color removal and COD reduction (98.1% and 98.2%, respectively) of the five methods considered. However, the residual color was 74.4 TCU and the residual COD was 4.2 g/L, which are considerably higher than those in ordinary wastewater. Therefore, additional treatment methods should be investigated in order to further improve the process.

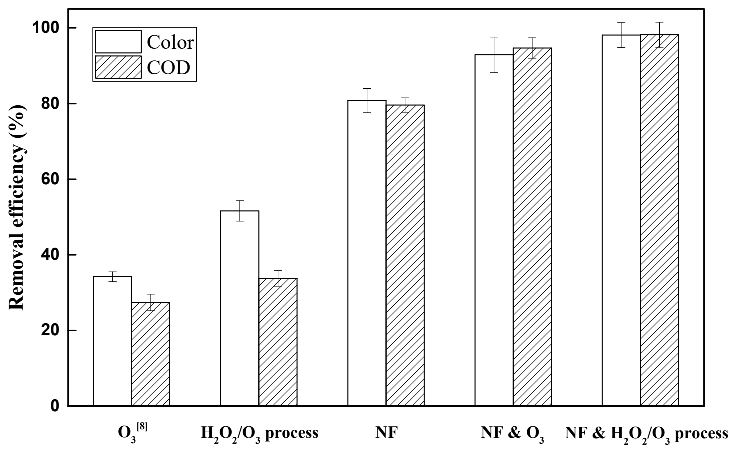


Figure 7. Comparison of removal efficiencies from five treatment methods.

Table 7. Color removal and COD reduction by various treatment methods.

Parameter	Amount of Removal (Removal Efficiency)	
	Color (TCU)	COD (g/L)
O_3 [8]	1333.8 (34.2%)	63.3 (27.4%)
H_2O_2/O_3 process	1857.6 (51.6%)	73.6 (33.8%)
NF (NE-70)	2908.8 (80.8%)	173.3 (79.6%)
NF & O_3	3344.4 (92.9%)	206.2 (94.7%)
NF & H_2O_2/O_3 process	3531.6 (98.1%)	213.8 (98.2%)

4. Conclusions

In this study, the H_2O_2/O_3 process were optimized for removing the color and reducing the COD of waste soy sauce. The H_2O_2/O_3 process were conducted under optimized conditions (H_2O_2/O_3 ratio or 0.3, pH of 11.0, and applied O_3 dose of 50 mg L^{-1}), resulting in 51.6% color removal and 33.8% COD reduction. This was primarily due to the high oxidation capability of O_3 in the presence of the hydroxyl radicals introduced by the addition of H_2O_2. Moreover, an appropriate membrane was selected for waste soy sauce pretreatment based on the molecular weight distribution of the wastewater. The addition of this pretreatment resulted in 98.1% color removal and 98.2% COD reduction. Comparing with alternative methods, NF & H_2O_2/O_3 process can be considered one of the best treatment methods for waste soy sauce, which requires particularly high degrees of color removal and COD reduction. These results can ultimately guide future research into the best wastewater treatment techniques that would allow the wastewater to be reused and mitigate the environmental impacts when it is discharged.

Author Contributions: H.-H.J. conceived and designed the experiments, analyzed the data, participated literature review, and preparation of the manuscript. G.-T.S. provided methodology of the research, participated in analyzed the data, and preparation of the manuscript. D.-W.J. offered suggestions on the concept and preparation of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

References

1. Lertsiri, S.; Maungma, R.; Assavanig, A.; Bhumiratana, A. Roles of the Maillard reaction in browning during moromi process of Thai soy sauce. *J. Food Process. Preserv.* **2001**, *25*, 149–162. [CrossRef]
2. Nursten, H.E. *The Maillard Reaction: Chemistry, Biochemistry, and Implications*; Royal Society of Chemistry: London, UK, 2005.
3. KOrean Statistical Information Service (KOSIS). Available online: http://kosis.kr/statHtml/statHtml.do?orgId=145&tblId=TX_14503_B016&conn_path=I3 (accessed on 13 April 2018).
4. Pooja, R.N. Removal of Organic Matter from Wastewater by Activated Sludge Process—Review. *Int. J. Sci. Eng. Technol. Res.* **2014**, *3*, 1260–1263.
5. Zheng, C.; Zhao, L.; Zhou, X.; Fu, Z.; Li, A. *Treatment Technologies for Organic Wastewater*; Intech: London, UK, 2013; pp. 249–286.
6. Pipe-Martin, C. Dissolved organic carbon removal by biological treatment. In Proceedings of the Ninth International Conference on Water Pollution: Modelling, Monitoring, and Management, Alicante, Spain, 9–11 June 2008; pp. 445–452.
7. Yoon, Y.; Hwang, Y.; Kwon, M.; Jung, Y.; Hwang, T.M.; Kang, J.W. Application of O_3 and O_3/H_2O_2 as post-treatment processes for color removal in swine wastewater from a membrane filtration system. *J. Ind. Eng. Chem.* **2014**, *20*, 2801–2805. [CrossRef]
8. Jang, H.H.; Seo, G.T.; Jeong, D.W. Investigation of Oxidation Methods for Waste Soy Sauce Treatment. *Int. J. Environ. Res.* **2017**, *14*, 1190. [CrossRef] [PubMed]
9. Fahmi, M.R.; Abidin, C.Z.A.; Rahmat, N.R. Characteristic of colour and COD removal of azo dye by advanced oxidation process and biological treatment. In Proceedings of the International Conference on Biotechnology and Environment Management, Singapore, 2–4 September 2011; pp. 8–13.
10. Rollon, A.A.; Dionisio, M.G.G. Treatment of Distillery Wastewater using Combinations of Advanced Oxidation Processes: UV/O_3, UV/H_2O_2 & H_2O_2/O_3. In Proceedings of the 14th International Conference on Environmental Science and Technology, Rhodes, Greece, 3–5 September 2015.
11. Zheng, Y.; Yu, S.; Shuai, S.; Zhou, Q.; Cheng, Q.; Liu, M.; Gao, C. Color removal and COD reduction of biologically treated textile effluent through submerged filtration using hollow fiber nanofiltration membrane. *Desalination* **2013**, *314*, 89–95. [CrossRef]

12. Abid, M.F.; Zablouk, M.A.; Abid-Alameer, A.M. Experimental study of dye removal from industrial wastewater by membrane technologies of reverse osmosis and nanofiltration. *Iranian J. Environ. Health Sci. Eng.* **2012**, *9*, 17. [CrossRef] [PubMed]
13. Shahata, A.; Omata, T.; Urase, T. Removal of Color from Molasses Wastewater Using Membrane Bioreactor with Acidic Condition. *J. Water Environ. Technol.* **2013**, *11*, 539–546. [CrossRef]
14. Sharma, P.; Joshi, H. MF as Pretreatment of RO for Tertiary Treatment of Biologically Treated Distillery Spentwash. *Int. J. Environ. Sci. Technol.* **2016**, *7*, 172. [CrossRef]
15. Guo, W.D.; Huang, J.P.; Hong, H.S.; Xu, J.; Deng, X. Resolving excitation emission matrix spectroscopy of estuarine CDOM with parallel factor analysis and its application in organic pollution monitoring. *J. Environ. Sci.* **2010**, *31*, 1419–1427.
16. Her, N.; Amy, G.; Foss, D.; Cho, J.; Yoon, Y.; Kosenka, P. Optimization of method for detecting and characterizing NOM by HPLC-size exclusion chromatography with UV and on-line DOC detection. *Environ. Sci. Technol.* **2002**, *36*, 1069–1076. [CrossRef] [PubMed]
17. Kaewsuk, J.; Seo, G.T. A Study on Application of NF Membrane Process for Enhanced Drinking Water Quality. Ph.D. Thesis, Changwon National University, Gyeongsangnam-do, Korea, 2011.
18. Ministry of Environment. Available online: http://www.me.go.kr/home/web/index/do?menuld=70 (accessed on 10 March 2018).
19. Ching, Y.C.; Redzwan, G. Biological Treatment of Fish Processing Saline Wastewater for Reuse as Liquid Fertilizer. *Sustainability* **2017**, *9*, 1062. [CrossRef]
20. Al−Kdasi, A.; Idris, A.; Saed, K.; Guan, C.T. Treatment of textile wastewater by advanced oxidation processes—A review. *Glob. Nest J.* **2004**, *6*, 222–230.
21. Kommineni, S.; Zoeckler, J.; Stocking, A.; Liang, P.S.; Flores, A.; Rodriguez, R.; Brown, T.; Per, R.; Brown, A. *3.0 Advanced Oxidation Processes*; Center for Groundwater Restoration and Protection National Water Research Institute: Los Angeles, CA, USA, 2000; pp. 110–208.
22. Hao, X.; Li, R.; Wang, J.; Yang, X. Numerical simulation of a regenerative thermal oxidizer for volatile organic compounds treatment. *Environ. Eng. Res.* **2018**, *23*, 397–405. [CrossRef]
23. Yuan, R.; Liu, D.; Wang, S.; Zhou, B.; Ma, F. Enhanced photocatalytic oxidation of humic acids using Fe^{3+}-Zn^{2+} co-doped TiO_2: The effects of ions in aqueous solutions. *Environ. Eng. Res.* **2017**, *23*, 181–188. [CrossRef]
24. Cortez, S.; Teixeira, P.; Oliveira, R.; Mota, M. Evaluation of Fenton and ozone-based advanced oxidation processes as mature landfill leachate pre-treatments. *J. Environ. Manag.* **2011**, *92*, 749–755. [CrossRef] [PubMed]
25. Li, G.; He, J.; Wang, D.; Meng, P.; Zeng, M. Optimization and interpretation of O_3 and O_3/H_2O_2 oxidation processes to pretreat hydrocortisone pharmaceutical wastewater. *Environ. Technol.* **2015**, *36*, 1026–1034. [CrossRef] [PubMed]
26. Jang, W.J.; Roh, H.S.; Jeong, D. W. An important factor for the water gas shift reaction activity of Cu-loaded cubic $Ce_{0.8}Zr_{0.2}O_2$ catalysts. *Environ. Eng. Res.* **2018**, *23*, 339–344. [CrossRef]
27. Yetilmezsoy, K.; Sakar, S. Improvement of COD and color removal from UASB treated poultry manure wastewater using Fenton's oxidation. *J. Hazard. Mater.* **2008**, *151*, 547–558. [CrossRef] [PubMed]
28. Yang, D.M.; Yuan, J.M. COD and Color removal from real dyeing wastewater by ozonation. *Water Environ. Res.* **2016**, *88*, 403–407. [PubMed]
29. Kwon, M.; Kye, H.; Jung, Y.; Yoon, Y.; Kang, J.W. Performance characterization and kinetic modeling of ozonation using a new method: ROH, O_3 concept. *Water Res.* **2017**, *122*, 172–182. [CrossRef] [PubMed]
30. Kim, K.S.; Kim, S.B.; Kim, J.B.; Jeon, Y.B.; Jang, E.S.; Lee, S.J.; Kim, S.W. Pilot Plant Study for Evaluation of High Concentration Industrial Wastewater by Ozone Oxidation System. *Korean Soc. Environ. Eng.* **2004**, 901–902.
31. Lee, E.; Lee, H.; Kim, Y.K.; Sohn, K.; Lee, K. Hydrogen peroxide interference in chemical oxygen demand during ozone based advanced oxidation of anaerobically digested livestock wastewater. *Int. J. Environ. Sci. Technol.* **2011**, *8*, 381–388. [CrossRef]
32. Albrektienė, R.; Rimeika, M.; Zalieckienė, E.; Šaulys, V.; Zagorskis, A. Determination of organic matter by UV absorption in the ground water. *J. Environ. Eng. Landsc.* **2012**, *20*, 163–167. [CrossRef]

33. Kaewsuk, J.; Seo, G. T. Verification of NOM removal in MIEX−NF system for advanced water treatment. *Sep. Purif. Technol.* **2011**, *80*, 11–19. [CrossRef]
34. Hassan, A.R.; Rozali, S.; Safari, N.H.M.; Besar, B.H. The roles of polyethersulfone and polyethylene glycol additive on nanofiltration of dyes and membrane morphologies. *Environ. Eng. Res.* **2018**, *23*, 316–322. [CrossRef]