



Comparative evaluation of the elimination of taste and odor substances via UV/H₂O₂ and UV/Cl₂ advanced oxidation at a pilot-scale plant

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ABSTRACT

This study evaluated taste and odor substance removal and EEO (electrical energy per order) values in a pilot-scale UV/H₂O₂ and UV/chlorine process. Despite its effectiveness, the UV/H₂O₂ process had an operational problem related to the quenching system for residual chemicals. Only 5%–20% of the H₂O₂ in the UV reactor was consumed because of its low molar absorption coefficient ($\epsilon_{\text{H}_2\text{O}_2 \text{ at } 254 \text{ nm}} = 19.6 \text{ M}^{-1} \text{ cm}^{-1}$); therefore, a large volume of H₂O₂ is needed for oxidation of target compounds. However, a small amount of chlorine remains after UV/chlorine oxidation because free chlorine has a higher molar absorption coefficient ($\epsilon_{\text{HOCl at } 254 \text{ nm}} = 59 \text{ M}^{-1} \text{ cm}^{-1}$, $\epsilon_{\text{OCl}^- \text{ at } 254 \text{ nm}} = 66 \text{ M}^{-1} \text{ cm}^{-1}$) and results in higher quantum yields (average $\Phi_{\text{HOCl at } 254 \text{ nm}} = 1.25$, $\Phi_{\text{OCl}^- \text{ at } 254 \text{ nm}} = 1.03$) than H₂O₂. In the UV/chlorine process, the 2-MIB removal efficiency decreased as the concentration of injected chlorine increased, with a correlation coefficient of -0.95 without pH adjustment. The removal rate of 2-MIB and the EEO value both increased with a correlation coefficient of 0.97 . The EEO value, a factor used to evaluate the economic efficiency of the treatment process, decreased as the H₂O₂ injection concentration increased in the UV/H₂O₂ process.

Keywords: Taste and odor substance; UV/H₂O₂; UV/Cl₂; Electrical energy dose (EED); OH radical; Chlorine radical

1. Introduction

Taste and odor substances such as geosmin and 2-methylisoborneol (2-MIB) are the main reasons for consumer complaints about tap water due to both an extremely low odor detection concentration limit as well as the difficulty involved in completely removing these substances through conventional water treatment processes [1]. The control of taste and odor (T&O) substances in tap water is currently a major challenge to be solved by drinking water treatment

plants. Despite the use of activated carbon to solve the T&O problem during several decades, the T&O issue has persisted due to current environmental conditions such as climate change and algal blooms. Thus, we need more powerful and efficient technology to control the T&O substances in drinking water treatment plants. The advanced oxidation process (AOP) produces hydroxyl radicals (OH[•]), powerful oxidants which can chemically decompose and remove most organic contaminants in water as opposed to selectively reacting to

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pollutants such as ozone. Thus, the AOP technology has been widely applied to various fields, including water treatment. The removal efficiency of target compounds, however, can be impaired by the scavenging effect of the water matrix. Significant AOP technologies are ozone, ultraviolet (UV)/hydrogen peroxide (H_2O_2), UV/chlorine, and ozone/ H_2O_2 . The efficiency of UV AOP is directly related to the permeability of UV photons into the water matrix [2,3]. Furthermore, removal efficiency of contaminants can be affected by the molar light absorption coefficient, light intensity, wavelength range, and light transmittance. The UV/ H_2O_2 process is widely used for drinking and reuse water treatment since it has a strong disinfecting effect on *Giardia*, *Cryptosporidium*, and other viruses, as well as high removal efficiency for trace amounts of organic contaminants in water. Although H_2O_2 absorbs UV light in the wavelength range of 200–300 nm, a sufficient amount of H_2O_2 should be injected into water for ideal OH^\bullet generation because the molar light absorption coefficient of H_2O_2 is low in the general UV output wavelength range [2–4]. The injected H_2O_2 is photolyzed by UV radiation to generate OH^\bullet as shown in Eq. (1).



When H_2O_2 is introduced as an oxidizing agent to generate OH^\bullet , it overflows into the process following the UV reactor, and must, therefore, be quenched by chlorine injection if there is no granular activated carbon adsorption unit in the water treatment process. It is necessary to continuously measure the residual concentration of H_2O_2 to calculate the correct amount of chlorine to inject, but the concentration is difficult to measure continuously. Using H_2O_2 as a treatment agent is relatively expensive and can also increase the cost of water treatment.

Recently, the UV/chlorine process has been studied as an alternative to the UV/ H_2O_2 process in terms of the AOP technology. The UV/chlorine process additionally enables the removal of various pollutants and may be more economical for quenching facilities. For instance, activated carbon adsorption units are not necessary for the UV/chlorine process, and residual chlorine from the UV reactor can be used to chlorinate the subsequent process. The UV/chlorine process has also been reported effective for treating trace amounts of harmful substances, pharmaceutical and personal care products, trichloroethylene, geosmin, and 2-MIB [5–9]. The treatment mechanism of the UV/chlorine process is shown in Eqs. (2)–(5). When free residual chlorine in water is exposed to UV, OH^\bullet and Cl^\bullet are formed [10]. Meanwhile, Cl^\bullet can react with Cl^- to produce $\text{Cl}_2^{\bullet-}$. As a result, Cl^\bullet , OH^\bullet ,

and $\text{Cl}_2^{\bullet-}$ radicals are the main active components used to remove organic contaminants during the UV/chlorine process.



The radical chemistry of the UV/chlorine process is not as clear as in the UV/ H_2O_2 process because of the coexistence of various reactive radicals such as Cl^\bullet , OH^\bullet , and $\text{Cl}_2^{\bullet-}$ in the UV/chlorine process. In this study, the UV/ H_2O_2 and the UV/chlorine process were installed in a pilot scale in a large existing drinking water treatment plant to evaluate the 2-MIB removal efficiency. The consumption rates of oxidizing chemicals (H_2O_2 and chlorine), as well as the electrical energy per order value of each process, were examined to evaluate their economic values in future operations and also to establish basic data for the introduction of UV AOP to the conventional water treatment process.

2. Materials and methods

2.1. Pilot plant test

This experiment was performed on a pilot plant installed at the Incheon N water purification plant at a time when the concentration of 2-MIB was unusually high as a T&O substance. The capacity of the pilot plant was $2,000 \text{ m}^3 \text{ d}^{-1}$ and hydraulic retention time of UV reactor was 9.5 s. The N water purification plant takes raw water from the Han river and produces purified drinking water through conventional processes including mixing, coagulation, sedimentation, rapid sand filtration, and chlorine disinfection. The pilot plant was installed to collect water samples from the sedimentation and rapid sand filtration units of the conventional water purification plant. The sampled water passed through the UV AOP reactor and then through the granular activated carbon and filtration/adsorption (F/A) treatment units as shown in Fig. 1. The low pressure (LP) UV reactor consisted of two stages: the first stage reactor contained four 0.96 kW LP UV lamps (Light Sources Inc., USA) and the second stage reactor contained three identical lamps. The lamp output of the LP UV reactor could be

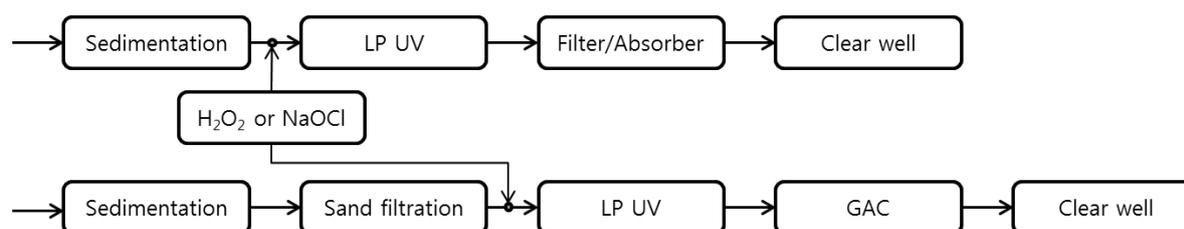


Fig. 1. Schematic diagram of the pilot scale UV AOP process.

adjusted within the range of 50%–100%. The reactors and the pilot plant were designed and installed by the ECOSSET Co., Ltd. of Korea.

As shown in the pilot plant schematic (Fig. 1), the sedimentation water and the rapidly filtered water could be sent to the LP UV reactors separately. H_2O_2 or sodium hypochlorite ($NaOCl$) was injected after sufficient mixing with water through the in-line mixer. This paper discusses the results of the LP UV treatment on sedimentation water and sand filtration water only. Table 1 summarizes the quality of the sedimentation water and the rapid filtration water entering the pilot plant. The turbidity of the sedimentation water was 0.26 NTU and that of the rapid filtration was 0.08 NTU, which showed the turbidity of the sedimentation water was three times higher than that of rapid filtration water while total organic carbon (TOC) and UV absorbance at 254 nm (UV254) were higher in the sedimentation water. The 2-MIB concentration was 36.2 ng L^{-1} in sedimentation water and 28.9 ng L^{-1} in rapid filtration water. The UV transmittance (UVT) value, which indicates the UV irradiation transmittance in water due to the metal ions and water turbidity, was 93.4% in the sedimentation water and 94.8% in the rapid filtration water. The rapid filtration water passed through one additional unit of sand filtration.

2.2. Analytical method

The TOC of the sample was measured using a TOC analyzer (Shimadzu TOC-V_{CPH}, Japan). UV254 was measured using a UV/Vis spectrophotometer (DR 5000, HACH, USA). The free residual chlorine was measured with the HACH® Pocket Colorimeter II using the standard method 4500-Cl G. The concentration of H_2O_2 was measured with PhotoLab® 7600 UV-VIS (WTW, Germany) using the neocuproine method. The T&O substances, geosmin and 2-MIB, were analyzed with a gas chromatograph (model: Agilent 7890B) and a mass spectrometer (model: Agilent 7000C) according to the standard method 6040D.

3. Results and discussion

3.1. Decay of H_2O_2 and free chlorine

In the UV AOP process, the removal rates of H_2O_2 and chlorine injected as an oxidant were calculated by measuring the amounts before and after using the UV reactor, which was in the range of 6%–18% for H_2O_2 with a mean removal rate of 10% according to the electrical energy dose (EED: the amount of electric energy input per unit volume (m^3) of water) to the UV reactor. The removal rate of H_2O_2 increased as the EED value increased (Fig. 2a), but there was no significant relationship between the H_2O_2 input concentration and the removal rate (Fig. 2b). The consumption rate was highest at 7 mg L^{-1} of the H_2O_2 injection concentration and lowest at 10 mg L^{-1} .

The injection of chlorine as an oxidizing agent is not only cheaper than H_2O_2 , but the residual chlorine remaining after the UV reaction without quenching or removing can also be used as a residual disinfectant in the subsequent process, thus maintaining the target disinfection chlorine level. In order to verify the UV AOP effect on the conventional water treatment process, the experiment was conducted in a large pilot scale on sedimentation water and rapid sand filtration water with pH 7.5–7.7 with no artificial pH adjustment. Although other studies [3,7,11] reported that 40%–80% of chlorine was consumed before and after the UV reactor, this study showed large deviations in chlorine consumption from 10% to 56% with the mean consumption rate of 26%, depending on the EED value and the injection concentration (Fig. 3a).

The pH of the treated water did not change when H_2O_2 was injected. Unlike liquid chlorine (Eq. (6)), when $NaOCl$ was used to increase the chlorine concentration the high alkalinity of the $NaOCl$, as shown in Eq. (7), increased the pH of the treated water gradually, resulting in pH 7.61 for 0 mg L^{-1} , pH 7.91 for 2 mg L^{-1} , pH 8.43 for 4 mg L^{-1} , pH 8.72 for 7 mg L^{-1} , and pH 9.22 for 10 mg L^{-1} . This gradual increase showed that the residual chlorine and pH were highly correlated (Fig. 3b).

Table 1
Average value of water quality parameters

Process	Water quality parameters (unit)	Average	Range
Sedimentation	pH	7.5	7.4–7.6
	Alkalinity (mg L^{-1} as CaCO_3)	38	36–40
	Turbidity (NTU)	0.26	0.19–0.30
	TOC (mg L^{-1})	1.56	1.41–1.71
	UV254 (cm^{-1})	0.024	0.023–0.024
	Geosmin (ng L^{-1})	2.2	1.2–4.5
	2-MIB (ng L^{-1})	36.2	30.8–41.6
Sand filtration	pH	7.6	7.5–7.7
	Alkalinity (mg L^{-1} as CaCO_3)	37	35–39
	Turbidity (NTU)	0.08	0.04–0.12
	TOC (mg L^{-1})	1.49	1.29–1.65
	UV254 (cm^{-1})	0.022	0.021–0.023
	Geosmin (ng L^{-1})	2.69	1.1–3.4
	2-MIB (ng L^{-1})	28.9	24.2–33.2

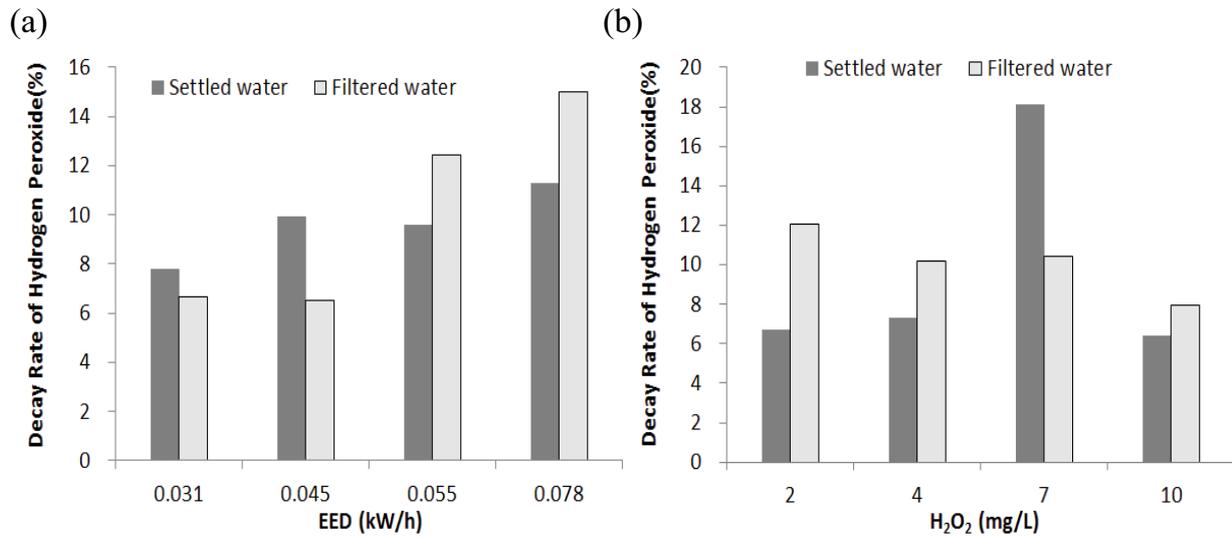


Fig. 2. Decay rate (%) of H₂O₂ by (a) EED and (b) H₂O₂ concentration.

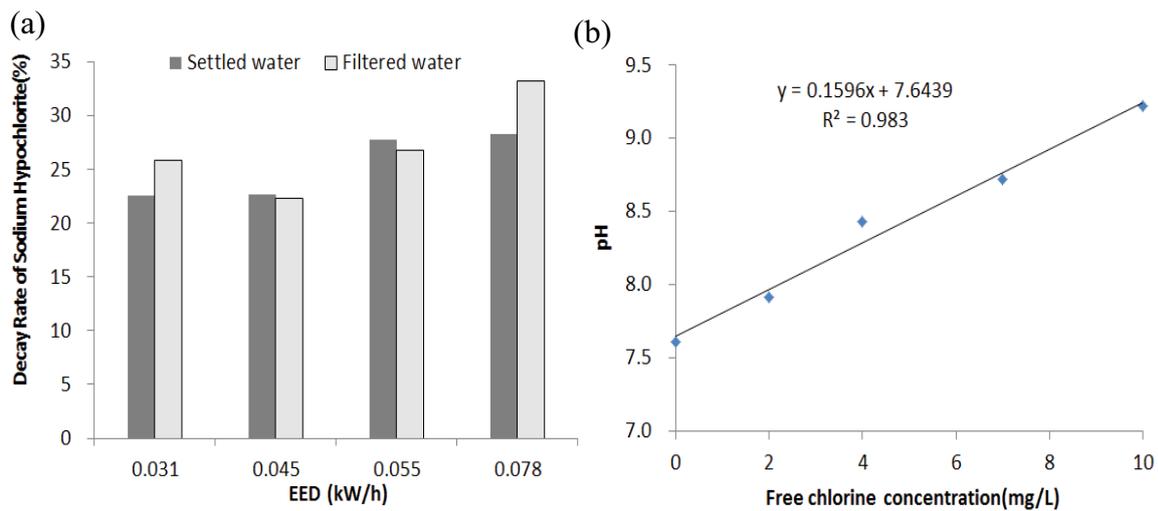


Fig. 3. Decay rate (%) of NaOCl by (a) EED and (b) free chlorine concentration.



3.2. 2-MIB control by the UV/H₂O₂ process

The 2-MIB is generally known to be more difficult to remove than geosmin, even when using AOP technology. The reason for different oxidation efficiencies in 2-MIB and geosmin is the steric hindrance caused by differences in the chemical structures of the two substances [12]. Therefore, if a process successfully removes 2-MIB, geosmin may be more easily controlled by that process. The 2-MIB selected for treatment had high concentrations ranging from 30 to 50 ng L⁻¹, while the geosmin treated during this experiment had a lower value of approximately 2 ng L⁻¹. The removal

efficiency of 2-MIB was investigated in the UV treatment and the UV/H₂O₂ process using various EED values and H₂O₂ injection concentrations.

The results of the test experiment (Fig. 4) on the sedimentation water and the sand filtration at LP UV showed that the removal efficiency of 2-MIB increased as the H₂O₂ concentration and the EED value increased. For the sand filtration water, the removal rate averaged 23.7% for the UV treatment alone. The removal rate was not strongly influenced by the EED value applied to the UV reactor. However, when injecting H₂O₂ to the UV reactor, the removal rates were increased to 53.5% at 2 mg L⁻¹, 72.9% at 4 mg L⁻¹, 86.9% at 7 mg L⁻¹, and 91.0% at 10 mg L⁻¹ of H₂O₂. The correlation coefficient between the 2-MIB removal rate and the H₂O₂ injection concentration was 0.93, and the correlation coefficient between the 2-MIB removal rate and the H₂O₂ injection concentration was 0.95. The results suggest

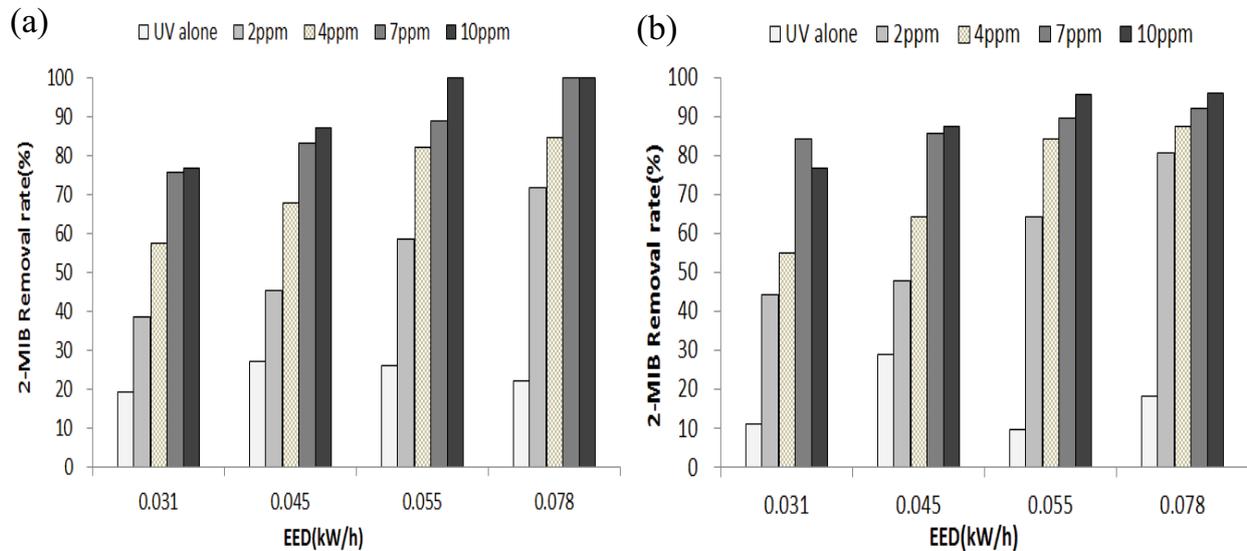


Fig. 4. 2-MIB removal rate of (a) sand filtered water and (b) settled water by EED in the UV/H₂O₂ process.

that oxidizing potential was enhanced by OH radicals generated through increases to both H₂O₂ concentration and electric energy inputs.

3.3. 2-MIB control by the UV/chlorine process

Using H₂O₂ as an oxidant in the AOP process to generate OH[•] is a more expensive and involved water treatment option, as the residual must be removed through quenching by an activated carbon filtration facility. When chlorine is used as an oxidizing agent in place of H₂O₂, the treatment process can be effective without activated carbon filtration after the UV AOP process. Further, the residual chlorine from the AOP reactor can be used to maintain the disinfection residual. From an economic standpoint, the chlorine process is considered preferable compared with the H₂O₂ process [10,13,14], since it requires no quenching facility. The treatment efficiency of the chlorine AOP process increased as the pH decreased, and the removal rate decreased as the pH increased [7,8,10,11]. This is because UV absorbance is higher for HOCl than for H₂O₂ while the scavenging rate of HOCl is lower than that of H₂O₂. The higher UV absorbance and lower scavenging rate indicate that HOCl is a better oxidant for the chlorine AOP than H₂O₂. In contrast, OCl⁻ scavenges OH[•] approximately four orders of magnitude faster than HOCl or H₂O₂, indicating that UV/chlorine AOP are more efficient as the water pH decreases. When chlorine is hydrolyzed in water, it produces hypochlorous acid (HOCl) and hypochlorite ion (OCl⁻) whose portions are functions of pH value. The HOCl form is favorable, more than 70% at pH 7 or lower, while the OCl⁻ form is dominant, greater than 70%, at pH 8 or higher, and OCl⁻ ion absorbs more than 4.5 times the MP UV light than HOCl. Therefore, odor substance removal efficiency was better at a lower pH [2,13].

In order to simulate the actual water treatment process, this experiment used the sedimentation water and the sand filtration from the conventional water treatment (pH 7.5–7.6) without artificial pH manipulation. When NaOCl was added

as an oxidizing agent, the average removal rate of 2-MIB was 14.1% for the sedimentation water and 15.2% for the sand filtration in the UV/chlorine AOP process (Fig. 5).

Other research results [3,6,15] showed that 40%–100% of the odor substances were removed depending on pH, contact time, UV irradiation intensity, and chlorine concentration, which was quite different from the aforementioned experiment. Most of the previous research was conducted using a collimated beam apparatus rather than a real plant. The poor removal rate was likely caused by the relatively short contact time in the UV reactor and the pH increase by OCl⁻ resulting from use of NaOCl. Wang et al. [3] conducted pilot- and full-scale UV/chlorination process evaluations and concluded that the UV/chlorine and UV/H₂O₂ processes differed only within 10% for destruction of taste and odor substances at pH 7.5 and 8.5. Unlike the result reported by Wang et al. [3], the results of this study showed the MIB removal efficiency to be much lower than that of UV/H₂O₂. These differences can be attributed to variations in water quality factors (e.g., pH, reactive radical scavenging) leading to radical generation and inhibition. Therefore, pH must be adjusted in the UV/chlorine process to improve the removal efficiency of 2-MIB. The experiments conducted on sand filtration water showed that 2-MIB removal efficiency decreased with increased concentration of injected chlorine with a correlation coefficient of -0.95 . The removal rate of 2-MIB increased as the EED value increased with a correlation coefficient of 0.97.

3.4. Electrical energy per order (EEO)

EEO (kWh m⁻³ order⁻¹) used to evaluate and compare the cost of water treatment is defined as the electrical energy required to reduce to 1 log or 90% the concentration of the target compound in 1 m³ of water. The EEO index was introduced as a basic concept by Dotson et al. [16] in 2001. While it is an excellent tool for comparing and evaluating the performance of full-scale UV AOP technology, the EEO index may

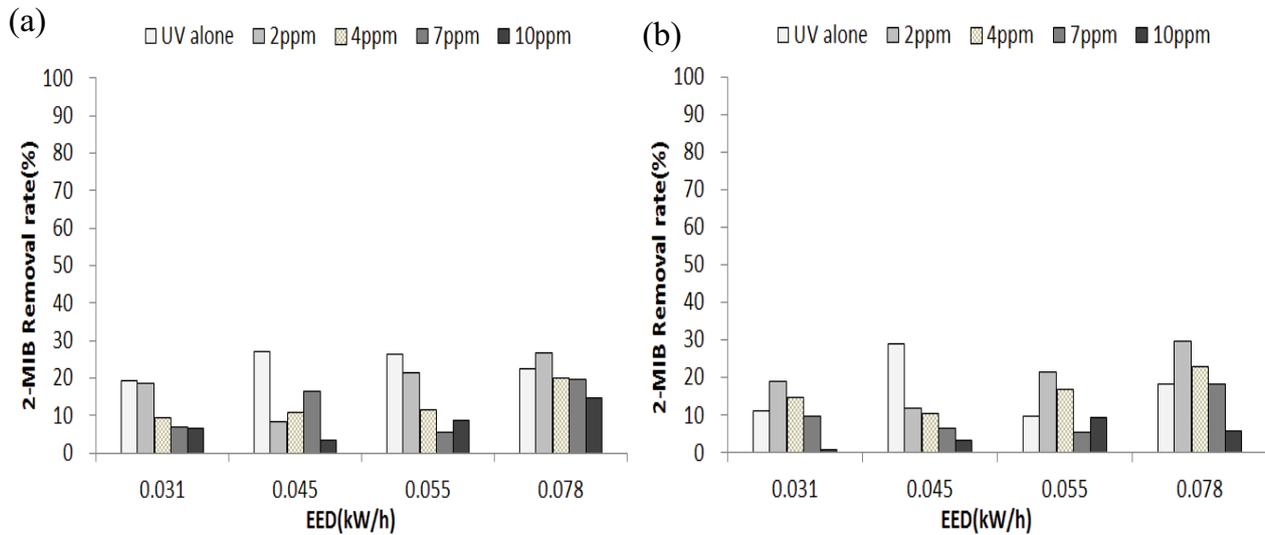


Fig. 5. 2-MIB removal rate of (a) sand filtered water and (b) settled water by EED in the UV/chlorine process.

Table 2
EEO value for the 90% removal rate of 2-MIB (unit: kWh m⁻³ order⁻¹)

Treatment	Oxidant input concentration (H ₂ O ₂ or chlorine)			
	2 mg L ⁻¹	4 mg L ⁻¹	7 mg L ⁻¹	10 mg L ⁻¹
UV/H ₂ O ₂ , settled water	0.109	0.094	0.063	0.048
UV/H ₂ O ₂ , filtered water	0.139	0.093	0.045	0.037
UV/chlorine, settled water	0.734	0.872	0.896	1.039
UV/chlorine, filtered water	0.620	0.868	0.990	2.042

not be effective for the collimated beam or bench scale test. Because the EEO value decreases as reactor size increases, the effect is more pronounced for the higher UVT [17]. Therefore, the EEO value for the relatively small pilot plant appears higher than the actual water treatment process. It is useful to calculate the EEO value which can be calculated from EED.

$$EED = \frac{(P \times T)}{(60 \times V)} \quad (8)$$

where P : power (kW), T : irradiation time (min), V : total system volume (m³).

$$EEO = \frac{EED}{\left(\log \frac{C_0}{C_e} \right)} \quad (9)$$

where EED: amount of electric energy input to 1 m³ of water (kWh m⁻³), C and C_e : concentration before and after UV reactor.

When experimental data are plotted as C_e vs. EED, the resulting slope will be equivalent to $-1/EEO$, which can be used to evaluate the EEO value for a given process. After setting the flow rate to the UV reactor, the UV output was adjusted to 0.031, 0.045, 0.055, and 0.078 kWh m⁻³, and the injection concentration of H₂O₂ was increased stepwise from

0 to 10 mg L⁻¹. The EEO values for the 90% removal rate of 2-MIB were compared for each process as shown in Table 2. The EEO values of the UV/H₂O₂ process decreased as the H₂O₂ injection concentrations increased for the sedimentation water, resulting in values of 0.109 kWh m⁻³ for 2 mg L⁻¹ of H₂O₂ and 0.048 kWh m⁻³ for 10 mg L⁻¹. The sedimentation water had lower UVT than the sand filtration, and because of the difference in UVT, the EEO value of sedimentation water was slightly higher at each H₂O₂ injection concentration except for the 2 mg L⁻¹ injection. The EEO value of the UV/chlorine process increased proportionally with the chlorine concentration, which was opposite to the trend observed in the H₂O₂ input. It was determined that the removal efficiency of 2-MIB decreased as a result of higher pH related to increased concentrations of NaOCl; OCl⁻ produced by this process acted as a scavenger. It is, therefore, recommended that pH be controlled prior to process initiation in order to remove 2-MIB through the UV/chlorine process. To achieve the target removal rate with the most economical operation, the EEO value obtained for each oxidant concentration can be used to determine the optimum operating point with regard to specific oxidant costs and electric charges.

4. Conclusions

This experiment involved installation of pilot plant scale UV/H₂O₂ and UV/chlorine AOP processes in a conventional

water treatment facility to remove 2-MIB, a T&O substance in drinking water. The study resulted in the following conclusions:

- H₂O₂ injected as an oxidizing agent had an average consumption rate of 10%, which was similar to other studies. The consumption rate tended to increase with the EED value. However, high input H₂O₂ concentration did not result in a higher rate of H₂O₂ consumption.
- The removal rate of 2-MIB can be expressed in pseudo-first-order kinetics as a function of first, the H₂O₂ injection concentration and second, the EED value. In the UV/H₂O₂ AOP process, 2-MIB removal rate increased proportionally with increases of EED and H₂O₂ concentration. The UV/H₂O₂ AOP process was capable of removing over 90% of 2-MIB through manipulation of H₂O₂ injection volume and EED value.
- In the UV/chlorine AOP process, the removal rate of 2-MIB increased as the EED value increased, while the removal rate decreased as the chlorine injection concentration increased. When NaOCl was added to increase the concentration of chlorine without artificially controlling the pH, the removal rate decreased proportionally as a result of the pH increase. Therefore, in order to increase the removal rate by the UV/chlorine process, water to be treated should be maintained at a lower pH in consideration of the pK_a value of NaOCl.
- The EEO value, used to evaluate the cost of water treatment, decreased as the H₂O₂ injection concentration increased in the UV/H₂O₂ process. Therefore, the EEO obtained at each H₂O₂ concentration could be utilized to identify the optimum operating point considering specific oxidant costs and electric charges.

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