Research Article

Combination of Coagulation and VUV+H_2O_2 for the Treatment of Color and Organic Matter in Treated Effluent Wastewater from a Sugar Factory

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Abstract

The removal of color and organic matter in treated effluent wastewater from a sugar factory by coagulation, combined with advanced oxidation processes (AOPs), including UV, UV+ hydrogen peroxide (UV+H_2O_2), vacuum UV (VUV), and VUV+H_2O_2, was investigated in this study. The effect of pH and a dose of coagulants (aluminum sulfate (alum), polyaluminium chloride (PACl), ferric chloride ($FeCl_3$), and ferric sulfate ($Fe_2(SO_4)_3$)), as a pretreatment step prior to AOPs, was analyzed. The optimum pH and coagulant dose for coagulation was pH 6 and 600 mg/L, respectively. The iron-based coagulants generally provided the better removal efficiency than the aluminium-based coagulants. Among four coagulants, $FeCl_3$ was chosen for use in the pretreatment step since it provided the best performance for dissolved organic carbon (DOC) removal, UV transmission improvement, and cost-effectiveness. The removal efficiency and degradation rate of color, UV absorbance at 254 nm (UV254), and DOC increased along with the H_2O_2 dosage. The reduction of UV254 indicated a less aromatic portion of DOC after AOPs. The ratio of DOC:H_2O_2 at 1:3 yielded the highest DOC removal of 70 ± 0.42 and 73 ± 0.19% in 60 min by UV+H_2O_2 and VUV+H_2O_2, respectively. For energy efficiency, VUV+H_2O_2 (DOC:H_2O_2 =1:3) provided the lowest electrical energy per order (EEO) (26 kWh/m^3). After coagulation, DOC combined with VUV+H_2O_2 was reduced to the appropriate level for reuse (4 mg/L). The findings from this research demonstrate the promise of coagulation coupled with the VUV+H_2O_2 process as a technology for water reuse application in the sugar industry.

Keywords: DOC, H_2O_2, Reuse, Sugar factory, Treated effluent wastewater, VUV

1 Introduction

Sugar is one of the most important agro-food industries in Thailand's socio-economic growth. Sugar production requires a large volume of fresh water (1–2 m^3/t of raw sugar) [1], [2] and generates a significant amount of wastewater, which is composed of high organic contents, such as sugar and other carbohydrates [2], [3]. If this untreated effluent is discharged into the environment, it will affect aquatic life, plants, and humans, and also alter soil properties. As a result, the wastewater from sugar production must be treated before disposal or reuse. Generally, the treated effluent from the sugar factory's wastewater treatment system can be used for the irrigation of sugarcane fields. However, if this treated effluent could be reused in sugar processing or other parts of the process (i.e., boiler), it would be more beneficial to the factory in reducing the risk of water scarcity due to drought as well as mitigating water conflict by communities access to the same water sources.

Treated effluent from the sugar factory retains organic matter and its yellowish color from melanoidins formed during a non-enzymatic browning reaction (Maillard reaction) between amino acids and carbohydrates (sugars) under high temperature [4]. Color from melanoidins is difficult to decompose by biological treatment processes. As a result, the treated wastewater has an undesirable color for recycling. This makes it necessary to improve the quality of the wastewater before it can be recycled. There are several methods for removing color and organic matter from treated effluents, such as adsorption, electrochemical treatment, photocatalysis, etc. [5]–[11]. Chemical treatment is one of the most popular methods because it can effectively remove turbidity, color, and organic matter, such as coagulation [12]–[15]. The coagulation process involves the addition of chemical agents, commonly aluminum salt ions and iron salt ions, to destabilize suspended particles and form flocs, which can be settled due to their size and weight. The coagulation process performance could be improved by adjusting the type of coagulant, coagulant dose, and pH. Advanced oxidation processes (AOPs), such as ultraviolet (UV) and vacuum UV (VUV) combined with hydrogen peroxide (H_2O_2) produce very active radicals for the degradation of organic matter in water [16], [17]. UV radiation at 200 to 400 nm, especially at 254 nm, appears to be one of the most popular technologies for water treatment. Besides UV, a VUV process, which emits radiation at 100 to 200 nm, is a more attractive and efficient method than traditional UV [17]–[19]. VUV has an advantage over UV in that it can generate highly reactive oxidizing species (hydroxyl radical: $OH^\bullet$) *in situ* by water homolysis [Equation (1)] [20]. Direct photolysis is dependent on the ability of compounds or medium to absorb the emitted light. Fortunately, water can absorb light strongly in the VUV region [21].

$$H_2O + hv\ (\lambda < 190\ nm) \rightarrow H^\bullet + OH^\bullet \qquad (1)$$

According to a previous investigation, coagulation using iron-based coagulants is proven to be effective in removing a significant amount of the color (78–80% at pH 5) and dissolved organic carbon (DOC) (40–42% at pH 5) from high salinity municipal wastewater [22]. Another study reported that coagulation alone reduced the DOC fraction in industrial wastewater containing insecticide by only 1%, but when combined with the photo-Fenton ($UV/Fe^{2+}/H_2O_2$) process, the efficiency of DOC removal rose to 57% after 60 min of irradiation due to an increase in the amount of $OH^\bullet$ [23]. Thus far, there is no study on the use of coagulation and AOPs for the treatment of color and organic matter in sugar factory wastewater. A VUV process is a relatively new advanced oxidation process with the capability to degrade organic contaminants with a high energy wavelength and oxidative species generation. The performance of the VUV system could be more powerful with the addition of H_2O_2. VUV+H_2O_2 has the potential to remove color and organic matter from sugar factory wastewater. Therefore, this research aims to study the efficiency of coagulation combined with UV-based AOPs (UV, UV+H_2O_2, VUV, VUV+H_2O_2) for removing color and DOC from wastewater in the sugar industry. This study analyzes the effect of pH and a coagulant dose of aluminum sulfate (alum), polyaluminum chloride (PACl), ferric chloride ($FeCl_3$), and ferric sulfate ($Fe_2(SO_4)_3$) as a pretreatment step prior to AOPs. The electrical energy efficiency of color and organic matter removal in wastewater is also discussed.

2 Materials and Methods

2.1 *Wastewater sample*

Treated effluent wastewater was collected from a biological wastewater treatment plant at a sugar factory in Phu Khiao District, Chaiyaphum Province, Thailand. The location coordinate of the factory is 16.4835898886261, 102.12128006180285. The plant employs anaerobic followed by an aerobic treatment process (aerated lagoon) to remove wastewater organics. The samples were kept refrigerated at 4 °C to avoid organic degradation during storage. The apparent sample was clear and yellow in color. The pH, turbidity, and color of the sample were 8.19 ± 0.01, 11.20 ± 0.07 NTU, and 122.00 ± 1.41 ADMI, respectively. The characteristics of organic matter were measured in terms of UV absorbance at 254 nm (UV254) and

DOC of 0.810±0.001 1/cm and 27.10 ± 2.83 mg/L, respectively.

2.2 *Coagulation pretreatment*

A laboratory jar test apparatus was used to conduct the coagulation pretreatment of the sample. Four coagulants: aluminum sulfate octadecahydrate or alum ($Al_2(SO_4)_3.18H_2O$, Merck, Germany), polyaluminum chloride (PACl, Merck, Germany), ferric chloride hexahydrate ($FeCl_3.6H_2O$, Merck, Germany), and ferric sulfate heptahydrate ($Fe_2(SO_4)_3.7H_2O$, QRëC, New Zealand) were used in the experiments. A stock solution of 50 g/L for each coagulant was prepared with deionized (DI) water. For each jar, 1 L of the sample was poured into a 1-L beaker. The pH of the samples was adjusted to the desired values (5–9) using 1 N hydrochloric acid (HCl, RCI labscan, Thailand) or 1 N sodium hydroxide (NaOH, QRëC, New Zealand). Five different concentrations were used to define the coagulant dose: 100, 200, 400, 600, and 800 mg/L. The experiment consisted of three steps: 1) a period of fast mixing at 150 rpm for 1 min; 2) a period of slow mixing at 30 rpm for 20 min; and 3) 60 min of sedimentation. Afterward, the supernatants were collected to analyze the turbidity, color, UV254, and DOC. The experiments were performed at least twice to determine the average removal percentage and standard deviation.

2.3 *AOPs treatment*

The AOPs reaction was performed in a 2 L graduate cylinder (82×470 mm). Low-pressure mercury UV lamps (model GPH383T5/L/HO) and VUV (model GPH383T5/VH/HO, Universal Lights Source, Inc., San Francisco, CA, USA) were used for UV, and VUV experiments, respectively. The UV lamps emit only 254 nm, while the VUV lamp emits polychromatic light wavelengths at 185 nm and 254 nm. The power input of each lamp was 30 W. The lamps were turned on for 10 min before starting each batch experiment to ensure constant light output. All experiments were operated in a batch mode using 2 L of the water sample. The H_2O_2 (Fisher, UK) was added in different concentrations based on the DOC mass concentration of mg DOC:mg H_2O_2 (1:1, 1:2, 1:3). The solution was mixed thoroughly for 60 min of reaction time. Samples of 50 mL were taken at time intervals of 0, 5, 10, 15, 30, and 60 min for analyses.

The color, UV254, and DOC removal were investigated using the first order kinetic model, as shown in the following equation:

$$lnC/C_0 = -kt \tag{2}$$

where, C and C_0 are final and initial concentrations of color (ADMI), UV254 (1/cm), and DOC (mg/L), k is the first order rate constant for the decay of color, UV254, and DOC (1/min), and t is the reaction time (min). The plots of ln (C/C_0) versus time were performed. The values of k from the slope of the plots at different reaction times and the coefficient of determination values (R^2) were obtained.

2.4 *Electrical energy per order*

Electrical energy per order (E_{EO}) is in kilowatt-hours (kWh), required to degrade a contaminant C by one order of magnitude in a unit volume (1000 L) of contaminated water. E_{EO} values (kWh/m^3/order) can be calculated using the following formula:

$$E_{EO} = \frac{Pt1000}{V \times log\ (C_i/C_f)} \tag{3}$$

where P is the power input of the lamp (kW) (In this experiment, the power input of the lamp is 30 W; therefore, the power is 0.03 kW), V is the volume (L) of water treated in time t (h), log is the symbol for the decadic logarithm, C_i is the initial concentration, C_f is the final concentration (M or mol/L), and the factor of 1,000 converts L to m^3 [24].

2.5 *Analytical methods*

The pH was measured with a bench-scale pH meter (SensION 2, Hach, USA). Turbidity was read directly from a turbidity meter (2100P, Hach, USA) in the nephelometry turbidity unit (NTU). Color was measured with a spectrophotometer (DR6000, HACH, USA) in the ADMI (American Dye Manufacturers Institute) unit. The UV254, an indicator of unsaturated/aromatic carbon, was measured by a spectrophotometer (DR6000, HACH, USA). The DOC concentration was measured by the total organic carbon and total nitrogen

(TOC&TN) analyzer (multi N/C 2100, Analytik Jena, Germany). The sample for DOC analysis was filtered through a prewashed 0.45 µm pore-size cellulose acetate membrane (Filtrex, India).

3 Results and Discussion

3.1 *Efficiency of coagulation treatment*

3.1.1 Effect of pH

The effects of pH in the range of 5–9 for a fixed coagulant concentration of 500 mg/L were examined. Figure 1(a) shows that alum, $FeCl_3$, and $Fe_2(SO_4)_3$ effectively eliminated turbidity in the pH range of 7–9 to 5.43 ± 0.33 to 3.47 ± 0.16 NTU (52 ± 2.90 to 69 ± 1.39% removal). The turbidity removal efficiency increased at this pH range, which was consistent with a previous investigation [25]. All coagulants were ineffective for removing turbidity at pH 5–6. Among four coagulants, PACl provided the lowest turbidity removal in all pHs. The highest efficiency of PACl for removing turbidity was only 43 ± 1.33% at pH 8. Since Thailand has no standard values for the characteristics of wastewater reuse, the water quality parameters were compared with the international guidelines or criteria. The guidelines provided by the Environmental Protection Agency (EPA) suggest a turbidity level ≤ 2 NTU for indirect potable reuse (IPR), such as groundwater recharge, by spreading or injecting into a potable aquifer and the augmentation of surface water supply reservoirs [26]. In this experiment, the turbidity of treated effluent after coagulation with $FeCl_3$ at pH 6 was approximately 3 NTU, which does not meet the EPA guidelines. However, the turbidity criteria for reclaimed water in agricultural irrigation recommended by the European Union (EU) is ≤ 5 NTU [27]. Therefore, the treated effluent meets the EU criteria.

Figure 1(b) presents that color removal was better with a pH of 6 (70 ± 1.74 to 92 ± 0.00% removal). As the pH increases, the decolorization efficiency tends to decrease. $Fe_2(SO_4)_3$ and $FeCl_3$ were the most effective coagulants with color removal at pH 6 of 92 ± 0.00% and 89 ± 1.16%, respectively.

Ultraviolet absorbance at 254 nm is a surrogate parameter for determining the aromatic structures or unsaturated carbon in the water [28], [29]. Figure 1(c) shows that UV254 can be easily removed at a pH of 6 (39 ± 2.27 to 72 ± 6.90%). Removal efficiency tended to decrease as the pH increased. $Fe_2(SO_4)_3$ was the best coagulant for removing UV254 (72 ± 6.90%). $FeCl_3$ and alum were equally effective with 54 ± 0.52% and 53 ± 3.23% of UV254 removal, respectively. PACl provided the least removal among coagulants (39 ± 2.27%). For pH 5, the UV254 values of wastewater treated by $FeCl_3$ and $Fe_2(SO_4)_3$ were higher than those of raw wastewater. This was due to the structure of chromophores changing as the pH decreased, causing greater UV absorbance at 254 nm. Melanoidins and low MW molecules (less than 350 Da) were further

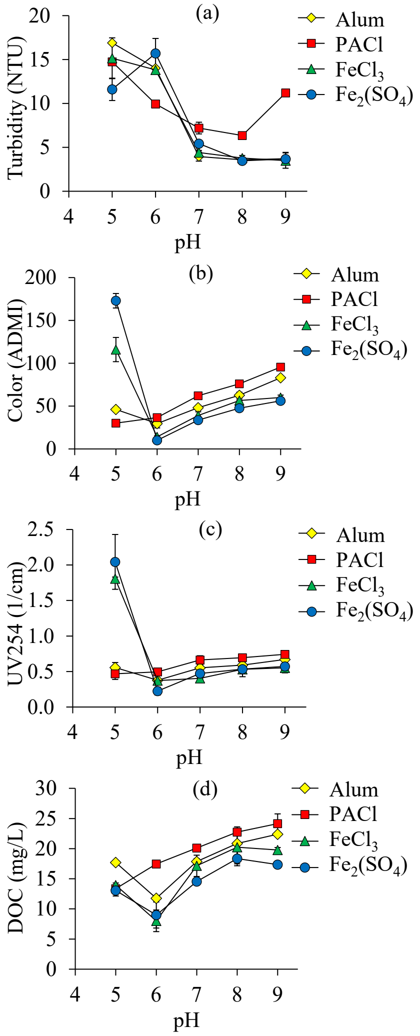

Figure 1: Effect of pH on (a) turbidity, (b) color, (c) UV254, and (d) DOC removal.

reduced by iron-based coagulants. Since anionic organic materials in molasses wastewater, such as melanoidins, amino acids, proteins, and sugars can form stable complexes with metal cations, such as Fe^{3+} in an acidic media, the development of certain higher MW color-causing chemicals, most likely Fe-organic complexes, offers the greatest potential [30].

Figure 1(d) presents the effect of pH on DOC removal efficiency. As the pH increases, DOC removal efficiency tends to decrease. The DOC removal efficiency at pH 6 of four coagulants in descending order were as follows: $FeCl_3 > Fe_2(SO_4)_3 >$ alum > PACl equating to 70 ± 0.34, 67 ± 0.39, 57 ± 0.44, and 36 ± 0.63%, respectively. This result is consistent with a previous work where PACl shows the lowest DOC removal [31].

According to a previous study, the optimal turbidity removal occurs at pH levels closer to neutrality due to the complex Al ion being less soluble, while the optimal coagulation of organic matter occurs at acidic pH levels between 4.5 and 6.0 (area with charge neutralization mechanism predominance) [32]. The elimination of organic matter occurs predominantly through charge neutralization under slightly acidic conditions [33]. Due to the lack of OH^- at acidic conditions, the hydrolysis of Al was restrained, existing in the monomeric and dimeric form as a positive charge. Therefore, charge neutralization was the dominating mechanism under acidic conditions [34].

The optimal pH for removal of color, UV254, and DOC was pH 6 at a coagulant concentration of 500 mg/L. $Fe_2(SO_4)_3$, while $FeCl_3$ provided the better efficiency than other coagulants. A previous study also reported that DOC removal was the most efficient at pH 6 [31]. When the pH was increased or decreased from pH 6, the removal efficiency of color, UV254, and DOC tended to decrease. At pH 5, turbidity, color, and UV254 with iron-based coagulants were higher than the initial values due to the solubility of iron at low pH. As the pH was higher, more soluble iron complexes were produced [22]. Based on the results of this experiment, pH 6 was chosen to investigate the effect of the coagulant dosage in the next step. The treated water with pH 6 may be too low for direct reuse. Since, pH can affects corrosion or precipitation, therefore, pH needs to be adjusted prior to reuse. However, it can be indirectly reused, as suggested in the EPA guidelines on water reuse [26] and the standard for wastewater reuse in the Eastern Mediterranean Region reported by the World Health Organization (WHO) [35]. The recommended pH for agricultural reuse (e.g., spray irrigation of food crops, pasture irrigation), industrial reuse (e.g., once-through cooling, recirculating cooling tower), and groundwater recharge is pH 6–9.

3.1.2 Effect of coagulant dosage

The effect of coagulant dosage on turbidity, color, UV254, and DOC removal at pH 6 is shown in Figure 2. All coagulants removed turbidity well in the concentration range of 400–600 mg/L [Figure 2(a)]. As the coagulant dosage increased to 800 mg/L, the turbidity removal efficiency tended to decrease (except for PACl). The highest turbidity removal of each coagulant was found at the following concentration dosage: alum 400 mg/L (75 ± 0.76%), PACl 800 mg/L (55 ± 9.15%), $FeCl_3$ 600 mg/L (73 ± 0.13%), and $Fe_2(SO_4)_3$ 600 mg/L (73 ± 0.13%).

Figure 2(b) presents the efficiency of color removal at pH 6. As the concentration of coagulant rises from 100 to 800 mg/L, the decolorization efficiency tends to increase. However, the decolorization efficiency increased slightly between 600 and 800 mg/L of coagulants. At 600 mg/L, the color removal efficiency was 80 ± 1.16, 63 ± 2.32, 89 ± 3.48, and 90 ± 1.74% for alum, PACl, $FeCl_3$, and $Fe_2(SO_4)_3$, respectively. At 800 mg/L, the color removal efficiency was 80 ± 4.06, 71 ± 4.64, 90 ± 1.16, and 81 ± 4.06% for alum, PACl, $FeCl_3$, and $Fe_2(SO_4)_3$, respectively.

The removal efficiency of UV254 tends to increase along with the concentration of coagulants [Figure 2(c)]. The UV254 removal efficiency of all coagulants at 600 mg/L in descending order is as follows: $Fe_2(SO_4)_3 > FeCl_3 >$ alum > PACl, equating to 77 ± 2.62, 76 ± 2.01, 62 ± 2.36, and 42 ± 2.18%, respectively. The UV254 removal efficiency of all coagulants at 800 mg/L increased slightly (except $Fe_2(SO_4)_3$) in descending order as follows: $FeCl_3 > Fe_2(SO_4)_3 >$ alum > PACl, equating to 81 ± 2.71, 73 ± 2.79, 65 ± 2.88, and 46 ± 2.27%, respectively.

Figure 2(c) presents the DOC removal efficiency at pH 6. It was discovered that as the concentration of coagulants increased, so did the removal efficiency. The DOC removal efficiency of all coagulants at

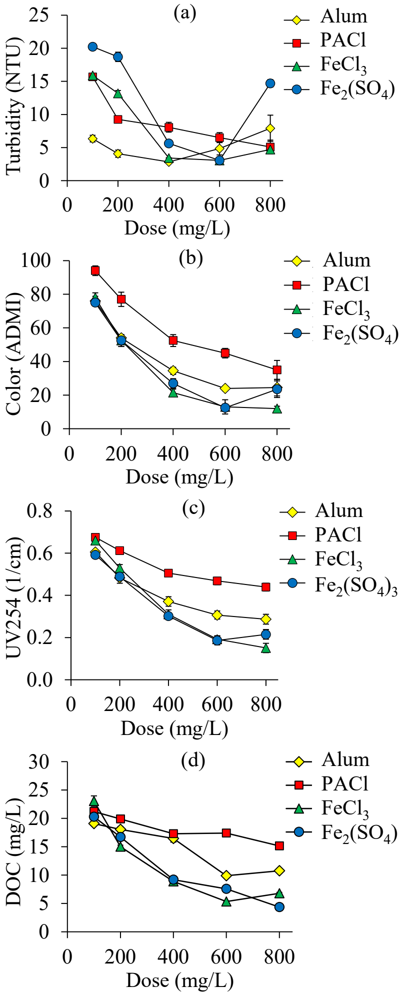

Figure 2: Effect of coagulant dosage on (a) turbidity, (b) color, (c) UV254, and (d) DOC removal.

600 mg/L in descending order is as follows: $FeCl_3$ > $Fe_2(SO_4)_3$ > alum > PACl, equating to 72 ± 1.25, 80 ± 0.47, 63 ± 0.47, and 36 ± 0.52%, respectively. The DOC removal efficiency of all coagulants at 800 mg/L in descending order is as follows: $Fe_2(SO_4)_3$ > $FeCl_3$ > alum > PACl, equating to 84 ± 0.78, 75 ± 0.60, 60 ± 1.30, and 44 ± 1.83%, respectively.

The removal efficiencies of color, UV254, and DOC follow the same trend. Increasing coagulant dosage tends to decrease color, UV254, and DOC removal. However, the greater removal efficiency was observed in color and UV254 than DOC. This is because coagulation can effectively remove color and aromatic compound (represented by UV254 value) but not other forms of any dissolved organic compound.

The removal of organic matter was significantly affected by the dose of coagulants. However, excessive doses had a limited effect [29]. Therefore, the most economical dose for turbidity, color, UV254, and DOC removal was selected as 600 mg/L at pH 6. $Fe_2(SO_4)_3$ and $FeCl_3$ provided the better turbidity, color, UV254, and DOC removal performance than alum and PACl. Umar *et al.* also reported higher performances for iron-based coagulants over alum-based coagulants on color and DOC removal from reverse osmosis concentrate in community wastewater [22]. Iron-based coagulants are more effective than aluminum-based coagulants because the floc of iron-based coagulants is larger. As a result, they can absorb more organic materials and a wider variety of organic compounds than aluminum-based coagulants [36]. In comparing the two iron-based coagulants, the removal efficiency of $Fe_2(SO_4)_3$ and $FeCl_3$ is quite similar. However, $FeCl_3$ is 14% cheaper than $Fe_2(SO_4)_3$. Therefore, $FeCl_3$ is suitable for use as a coagulant in coagulation treatment as the primary step for the removal of color and organic matter prior to further removal by UV or VUV-based AOPs.

3.2 *Efficiency of UV and VUV based AOPs treatment*

3.2.1 Effect of coagulation as pretreatment

Figure 3 shows the effect of $FeCl_3$ primary treatment on the removal efficiency of DOC by AOPs. The results indicate that pretreatment by $FeCl_3$ had a minor effect on DOC removal with UV, VUV, and H_2O_2 alone, but more DOC removals were observed by UV+H_2O_2 and VUV+H_2O_2 processes. This is because the coagulation process can eliminate colloidal particles, resulting in the increased transmission of light. The result shows that the percentage of UV transmission (%UVT) increased from 17 ± 0.20% (VUV+H_2O_2 without pretreatment) to 97 ± 0.79% (VUV+H_2O_2 with pretreatment). Pretreatment by $FeCl_3$ removed some parts of DOC, which, in turn, reduced the scavenging effect on $OH^•$. Therefore, H_2O_2 in the process of UV+H_2O_2 and VUV+H_2O_2 receives UV or VUV radiation and breaks down into $OH^•$, which enhances DOC removal. The coagulation with VUV+H_2O_2 resulted in a significantly higher percentage reduction

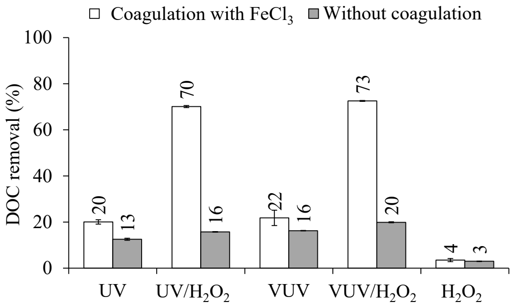

Figure 3: Effect of pretreatment by coagulation with $FeCl_3$ in DOC removal (DOC:$FeCl_3$=1:22, DOC:H_2O_2 =1:3, 60 min reaction time).

for DOC (73 ± 0.19%) than VUV alone (22 ± 3.33%). The findings from this study are consistent with earlier works, which reveal that the application of coagulation combined with UV/H_2O_2 for the treatment of post-pharmaceutical wastewater results in much greater reduction rates for DOC (by 75%) and UV254 (by 92%) than coagulation or UV/H_2O_2 oxidation alone [37]. This suggests that pretreatment with coagulation is required prior to AOPs. Therefore, the wastewater sample used in sections 3.2.2 to 3.2.4 was pre-treated with 600 mg/L of $FeCl_3$ at pH 6. The initial and final pH of treated wastewater were approximately 6.0 ± 0.3 and 5.0 ± 0.3, respectively.

3.2.2 Color improvement

Effluent pre-treated by coagulation still has an unpleasant color, making it unsuitable for reuse or recycling. Therefore, it should be further treated by AOPs to remove the yellowish color from melanoidins and humic acids. The color removal efficiency and reaction constants for VUV, VUV+H_2O_2, UV, and UV+H_2O_2 processes are shown in Figure 4 and Table 1, respectively. According to the experiment, the decolorization efficiency tends to increase along with the treatment time. A rapid reaction rate was observed during the first 5–15 min, after which it slowly increased. VUV+H_2O_2 outperformed VUV alone. Decolorization efficiency by VUV-based AOPs at 60 min was in the following order: VUV < VUV+H_2O_2(1:1) < VUV+H_2O_2(1:2) < VUV+H_2O_2(1:3), equating to 52 ± 2.95, 83 ± 1.31, 84 ± 1.18, and 87 ± 1.26%, respectively. The amount of H_2O_2 present in the VUV and UV processes influences the efficiency and rate of decolorization because it increases the amount of $OH^{\bullet}$ available for decomposing the organic substances that cause the change in color [38].

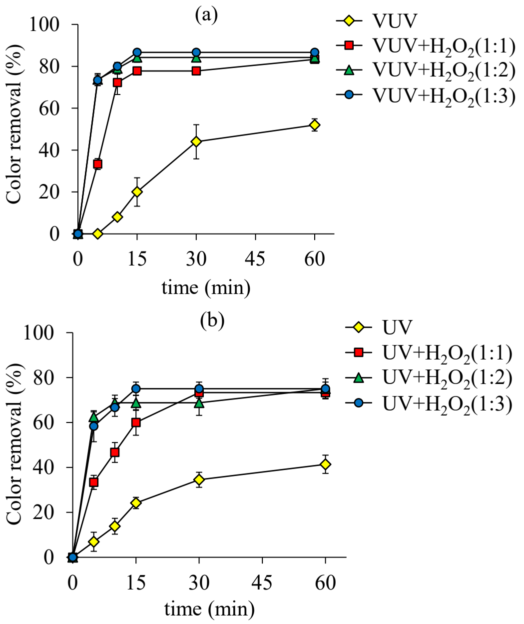

Figure 4: Effect of AOPs treatment on color removal: (a) VUV-based AOPs and (b) UV-based AOPs.

Table 1: Removal efficiency and reaction rate constant for the change in color (kADMI) by AOPs

Experiment (DOC:H_2O_2 mg/mg)	**Removal Efficiency (%)**		**k_{ADMI} (1/min)**	**R^2**
	0.97×10^3 mJ/cm^{2*}	**3.89×10^3 mJ/cm^{2**}**		
VUV	20 ± 6.8	52 ± 2.95	–0.017	0.948
VUV+H_2O_2(1:1)	78 ± 1.75	83 ± 1.31	–0.107	0.982
VUV+H_2O_2(1:2)	84 ± 1.18	84 ± 1.18	–0.178	0.941
VUV+H_2O_2(1:3)	87 ± 1.26	87 ± 1.26	–0.182	0.951
UV	24 ± 2.53	41 ± 4.06	–0.015	0.999
UV+H_2O_2(1:1)	60 ± 5.68	73 ± 2.53	–0.067	0.988
UV+H_2O_2(1:2)	69 ± 5.61	75 ± 4.49	–0.132	0.945
UV+H_2O_2(1:3)	75 ± 2.97	75 ± 2.97	–0.123	0.957

*UV dose at t = 15 min, ** UV dose at t = 60 min

The degradation rate constants were determined based on initial degradation rates (during the first 10–30 min) because no strong linear relationship (lnC/C_0 vs t) existed in the overall degradation kinetics. The pseudo first order rate law is given for the degradation of organic matter under AOPs treatment since the steady-state concentration of OH$^\bullet$ is considered to be constant with respect to pollutant concentrations [39]. Considering the reaction rate constant, which indicates the speed of the decolorization reaction, the rate constant of the VUV+H_2O_2 process was 1.1–1.9 times higher than that of UV+H_2O_2. This is because the VUV lamp emits wavelengths of 185 nm, which can decompose color molecules and generate OH$^\bullet$ through H_2O dissociation [20], [40]. Compared with a previous study by Puspita *et al.* [14], which removed color in municipal wastewater by VUV+H_2O_2 and UV+H_2O_2 processes, the ratio of DOC:H_2O_2 at 1:3 provided the maximum decolorization efficiency, which was similar to the results of this current study.

3.2.3 UV254 removal

The UV254 parameter represents the degree of aromaticity or organic matter content of unsaturated carbon-carbon bonds (C=C, C≡C) in the wastewater sample. This is because double-bound carbon molecules efficiently absorb UV radiation at this wavelength. A high level of UV254 indicates a significant amount of unsaturated organic compounds that are chromophores. These compounds are strongly bonded and difficult for biodegradation. Previous research has reported that approximately 44% of UV254 in industrial wastewater could be removed through a sequencing batch reactor (SBR) system [41]. VUV-based AOPs typically generate OH$^\bullet$ or use photons to break the bond or reduce the aromaticity of organic matter to form a structure that becomes easily biodegradable.

The efficiency and rate constants of UV254 reduction by VUV, VUV+H_2O_2, UV, and UV+H_2O_2 processes are shown in Figure 5 and Table 2, respectively. A fast reaction removal rate of UV254 occurred in the first 5–15 min, after which it slowly increased and reached a plateau. This suggested that the unsaturated and/or aromatic carbon molecules were preferentially oxidized. At 30 min, the UV254 removal efficiency was more than 50%. UV254 was removed more effectively by VUV than UV. This is due to the 185 nm wavelength of VUV enhancing the degradation of UV254 containing organic compounds. The removal efficiency of UV254 by VUV+H_2O_2 (96 ± 1.13%) was better than UV+H_2O_2 (94 ± 1.38%). The reaction rate constant for UV254 removal by VUV+H_2O_2 was also higher than UV+H_2O_2 (Table 2). UV254 removal efficiency by VUV-based AOPs at 60 min was in the following order: VUV < VUV+H_2O_2(1:1) < VUV+H_2O_2(1:2) < VUV+H_2O_2(1:3), equating to 80 ± 3.61, 91 ± 1.98, 93 ± 1.03, and 96 ± 1.13%, respectively. The rate of UV254 reduction increased linearly with the H_2O_2 dose. Specifically, UV254 indirectly represents the existence of unsaturated carbon bonds, including aromatic compounds, which have high electron density and are therefore preferentially attacked by OH$^\bullet$ [42].

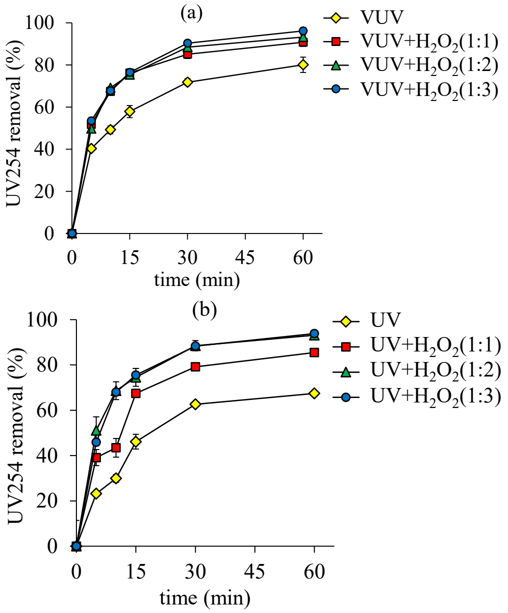

Figure 5: Effect of AOPs treatment on UV254 removal: (a) VUV-based AOPs (b) UV-based AOPs.

Table 2: Removal efficiency and reaction rate constant for UV254 (kUV254) removal by AOPs

Experiment (DOC:H_2O_2 mg/mg)	**Removal Efficiency (%)**		**k_{UV254} (1/min)**	**R^2**
	0.97×10^3 mJ/cm^{2*}	**3.89×10^3 mJ/cm^{2**}**		
VUV	58 ± 2.88	80 ± 3.61	–0.064	0.967
VUV+H_2O_2(1:1)	76 ± 1.87	91 ± 1.98	–0.104	0.982
VUV+H_2O_2(1:2)	75 ± 0.80	93 ± 0.43	–0.104	0.982
VUV+H_2O_2(1:3)	76 ± 0.87	96 ± 1.13	–0.121	0.983
UV	46 ± 3.25	67 ± 1.11	–0.035	0.986
UV+H_2O_2(1:1)	67 ± 1.03	85 ± 1.60	–0.058	0.969
UV+H_2O_2(1:2)	74 ± 2.82	93 ± 0.91	–0.080	0.962
UV+H_2O_2(1:3)	76 ± 2.92	94 ± 1.38	–0.080	0.968

*UV dose at *t* = 15 min, ** UV dose at *t* = 60 min

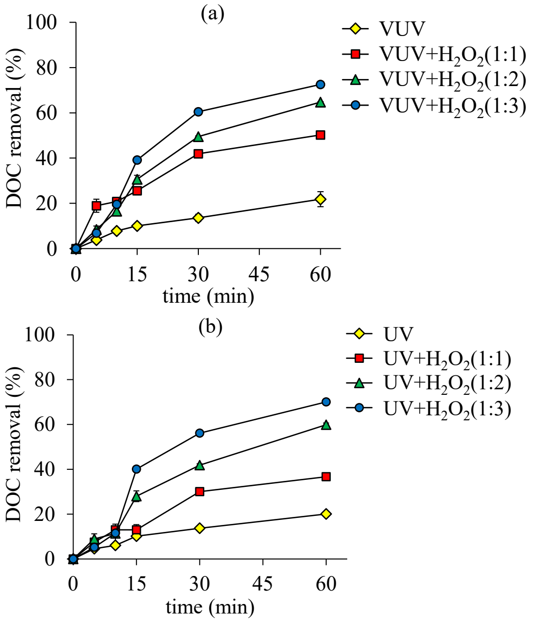

Figure 6: Effect of AOPs treatment on DOC removal: (a) VUV-based AOPs and (b) UV-based AOPs.

3.2.4 DOC removal

The removal efficiency and reaction rate constants of DOC by AOPs processes are shown in Figure 6 and Table 3, respectively. The results indicate that DOC removal gradually increases with reaction time (Figure 6). At 60 min, DOC removal by VUV and UV was 22 ± 3.33 and 20 ± 0.95%, respectively. The addition of H_2O_2 in VUV and UV improved DOC mineralization at 60 min to 73 ± 0.19 and 70 ± 0.42%, respectively (at DOC:H_2O_2 = 1:3). This finding appears consistent with previous research, which found that VUV and UV treatments exhibited small changes of DOC concentrations throughout the reaction time [17]. Adding H_2O_2 improved the DOC removal percentage as well as the degradation rate constant due to greater $OH^{\bullet}$ generation [43]. The reaction rate constants of VUV+H_2O_2 and UV+H_2O_2 at the ratio of DOC:H_2O_2 of 1:3 was found to be approximately 2 and 2.6 times higher than the ratio of 1:1, respectively. Noticeably, the addition of a higher concentration of H_2O_2 increased DOC removal efficiency. In this experiment, a DOC:H_2O_2 ratio of 1:3 provided the best DOC removal, which was consistent with previous experiments that employed VUV+H_2O_2 and UV+H_2O_2 processes to remove DOC in wastewater. Previous studies found that DOC removal in treated community wastewater [16] and reverse osmosis concentrate wastewater [22] were high at DOC:H_2O_2 ratios of 1:3 and 1:5, respectively.

Table 3: Removal efficiency and reaction rate constant of DOC removal (kDOC) by AOPs

Experiment (DOC:H_2O_2 mg/mg)	**Removal Efficiency (%)**		**k_{DOC} (1/min)**	**R^2**
	0.97×10^3 mJ/cm^{2*}	**3.89×10^3 mJ/cm^{2**}**		
VUV	10 ± 0.91	22 ± 3.33	–0.007	0.996
VUV+H_2O_2(1:1)	26 ± 0.81	50 ± 1.19	–0.019	0.967
VUV+H_2O_2(1:2)	31 ± 1.71	65 ± 0.16	–0.023	0.995
VUV+H_2O_2(1:3)	39 ± 0.89	73 ± 0.19	–0.030	0.986
UV	10 ± 0.26	20 ± 0.95	–0.006	0.965
UV+H_2O_2(1:1)	13 ± 2.35	37 ± 1.51	–0.012	0.987
UV+H_2O_2(1:2)	28 ± 2.39	60 ± 0.79	–0.018	0.984
UV+H_2O_2(1:3)	40 ± 1.28	70 ± 0.42	–0.027	0.958

*UV dose at t = 15 min, ** UV dose at t = 60 min

Notably, UV and VUV-based AOPs removed UV254 at a higher rate compared to DOC. This is because UV254 is a subgroup of unsaturated carbon organic compounds. These are electron-dense bonds and very sensitive to oxidants, while the reduction of DOC refers to the process of mineralization, which transforms organic matter into carbon dioxide. Moreover, the reduction of UV254 is an indicator of the increased biodegradability in the organic compound since AOPs can break down unsaturated carbon bonds [44].

In this study, the level of color, UV254, and DOC after pretreatment with $FeCl_3$ followed by VUV+H_2O_2 was decreased to 2 ADMI, 0.014 1/cm, and 4.41 mg/L, respectively. Quality requirements for reclaimed water used in toilet flushing are almost colorless and transparent [26]. The treated wastewater was still slightly colored. It might be further treated using a membrane to produce colorless wastewater. Several States in the USA have provided guidelines on the total organic carbon (TOC) ≤ 2 mg/L for IPR, while there is no standard for UV254 [26]. However, the level of color and DOC in our treated wastewater is close to the level in surface water and higher than the guidelines. Therefore, it might require further treatment, such as biological activated carbon filtration prior to reuse as raw water in the production of tap water [45].

Table 4: Electrical energy per order (E_{EO}) of UV and VUV AOPs with and without the pretreatment of $FeCl_3$

Experiment	Color (10 min)		DOC (60 min)		UV254 (10 min)	
	E_{EO} (kWh/m^3)	Removal (%)	E_{EO} (kWh/m^3)	Removal (%)	E_{EO} (kWh/m^3)	Removal (%)
UV	454	7 ± 0.39	252	13 ± 0.46	325	10 ± 0.08
UV+H_2O_2*	337	10 ± 2.64	200	16 ± 0.12	210	15 ± 0.14
VUV	742	5 ± 0.51	195	16 ± 0.09	443	8 ± 0.12
VUV+H_2O_2*	138	22 ± 2.16	157	20 ± 0.20	138	22 ± 0.21
$FeCl_3$+UV	233	14 ± 3.55	154	20 ± 0.95	97	30 ± 1.72
$FeCl_3$+VUV	414	8 ± 0.45	142	22 ± 3.33	51	49 ± 1.87
$FeCl_3$+UV+H_2O_2*	31	67 ± 3.96	29	70 ± 0.42	30	68 ± 0.82
$FeCl_3$+VUV+H_2O_2*	21	80 ± 1.89	26	73 ± 0.19	31	68 ± 0.99

* DOC:H_2O_2=1:3

3.3 *Evaluation of energy efficiency and economics*

The treatment methods used in AOPs for the removal efficiency of color, DOC and UV254, and the electrical energy per order (EE0) are compared in Table 4. The results demonstrate that pretreatment with $FeCl_3$ prior to AOPs consumes less energy than without pretreatment. The EE0 values for VUV+H_2O_2 processes with and without pretreatment for DOC removal equate to 26 and 157 kWh/m^3, respectively. Under H_2O_2-added conditions, the E_{E0} values for removing color, UV254, and DOC were less than those without the addition of H_2O_2. This suggests that adding H_2O_2 helps to increase energy efficiency. The DOC removal efficiency with VUV and UV processes was higher than VUV or UV alone with the addition of H_2O_2. It can be observed that pretreatment prior to VUV+H_2O_2 and UV+H_2O_2 processes consumes the least amount of energy. The total cost of VUV+H_2O_2 and UV+H_2O_2 processes for eliminating DOC at 60 min equated to 76.31 and 83.31 baht per 1 m^3 of wastewater, respectively (excluding the chemicals used for pH adjustment). The total cost was calculated from the highest amount of electricity at the time-of-use cost equating to 2.61 baht per unit (68 Thai baht for VUV+H_2O_2 and 75 Thai baht for UV+H_2O_2), the cost of $FeCl_3$ (1,000 mg/L = 7 Thai baht), and the cost of H_2O_2 (47 mg/L at DOC:H_2O_2(1:3) = 1.31 Thai baht)). This is only a preliminary laboratory study and an account of AOPs treatment. To scale up based on the process, we recommend taking into account the energy required to pump, aerate, and mix when evaluating a pilot or full-scale system.

4 Conclusions

This study demonstrates the technical and economic efficiency of turbidity, color, UV254, and DOC removal in treated sugar factory effluents with and without pretreatment by coagulation combined with VUV, VUV+H_2O_2, UV, and UV+H_2O_2 processes. The treated effluent from the biological wastewater treatment system at the sugar factory was found to be characteristically yellowish-brown in color with high DOC, making it unsuitable for recycling. The optimal pH and coagulant dose using coagulation as a pretreatment step were pH 6 and 600 mg/L (DOC:$FeCl_3$ = 1:22), respectively. The suitable coagulants for removing turbidity, color, UV254, and DOC were iron-based ($FeCl_3$ and $Fe_2(SO_4)_3$). $FeCl_3$ and $Fe_2(SO_4)_3$ have similar turbidity, color, and UV254 removal efficiency, but $FeCl_3$ was better at removing DOC. Pretreatment using the coagulation process improved the efficiency of AOPs in removing color and organic matter. This is because it helps to eliminate the suspended colloidal particles obstructing light dispersion, allowing light to better penetrate the water, while also eliminating some of the DOC. The VUV and UV technique is even more effective for removing color, UV254, and DOC with the addition of H_2O_2. VUV+H_2O_2 (DOC:H_2O_2 = 1:3) provided the greatest reduction in color (87 ± 1.26%), UV254 (96 ± 1.13%), and DOC (73 ± 0.19%) and also exhibited the highest energy efficiency.

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Author Contributions

P.K.: investigation, data curation, writing an original draft, reviewing and editing; N.P.: investigation, methodology, data analysis; T.R.: conceptualization, research design writing—reviewing and editing, funding acquisition, project administration. All authors have read and agreed to the published this version of the manuscript.

Conflicts of Interest

The authors declare no conflict of interest.

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