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Treatment of MSW landfill leachate by a thin gap annular UV/H_2O_2 photoreactor with multi-UV lamps

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Abstract

The treatment of leachate from landfills is a major disposal problem for municipal solid waste. The leachate is generally recalcitrant to be treated according to complicated characteristics and high color intensity resulting further threat for environment and human health. In this work, the designed thin gap annular photoreactor with 4-UV lamps in UV/H_2O_2 process was proposed to decolor and remove chemical oxygen demand (COD) from the landfill leachate for solving this environmental problem. Meanwhile, the operating parameters such as UV dosage, hydrogen peroxide concentration and leachate strength were evaluated. The landfill leachate treated with the maximum dosage of 4-UV lamps and 232.7 mM of hydrogen peroxide concentration achieved 72 and 65% of color and COD removal efficiencies in 300 min. As for less concentrated leachate of 20% strength, 91% of color and 87% of COD were removed within only 120 min. From the experimental results, the UV/H_2O_2 process in this work was an effective pre-treatment or treatment technology for landfill leachate.

Keywords: Landfill leachate; MSW; UV/H_2O_2 process; Decolorization; COD; Hydrogen peroxide

1. Introduction

One of the major environmental concerns of landfill disposal by municipal solid waste (MSW) is collection and treatment of leachate. Generally, the leachate results from the rainfall, runoff of surface drainage and groundwater percolating through the levels of solid waste and extracting the dissolved and suspended materials [1]. Corresponding to the mature age and the biochemical reaction in the landfill, the complex characteristics of leachate varies significantly consisting the organics, ammonia-nitrogen, heavy metals and dissolved solid to form high chemical oxygen demand (COD), nitrogen and color intensity. Thus, the treatment of landfill leachate is often complicated and expensive in order to comply with the effluent standards prior to discharge resulting into the challenge for environmental engineers. In Taiwan, the most common leachate treatment by conventional biological process such as rotary biological contactor (RBC) [2], sequential batch reactor (SBR) [2,3] and anaerobic process [3–6] were extensively investigated, which was substantially ineffective that most of the landfill facilities have difficulty in compliance with the regulated requirements. Thus, the treatment of landfill leachate is studied necessarily by the alternative physical and chemical treatment technologies. By sequential coagulation with Fenton oxidation followed by acid precipitation, about 90% of COD removal with the original of 3530 mg l^{-1} was demonstrated successfully [7]. Wang et al. [8] found 64% of COD removal by combining UV irradiation with $FeCl_3$ coagulation for leachate. By ozonation, Bila et al. [9] found the increase of BOD_5/COD from 0.05 to 0.3 to improve biodegradability, while Rivas et al. [10] also reported a moderate reduction of COD for a stabilized leachate. Moreover, Wu et al. [11] demonstrated significant enhancement of leachate biodegradability by applying ozone dosage of 1.2 g l^{-1} to remove 90% color, yet the removal of total organic carbon (TOC) was ineffective. Besides, some researches for leachate treatment by the other methods such as electro-Fenton [12],

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photo-Fenton [13] and membrane [14] demonstrated effectively.

UV/H_2O_2 process produces hydroxyl free radicals $^{\bullet}OH$ to enhance a high degradation rate of organics in aqueous system. This technology is widely used to decompose organic products in industrial wastewater and groundwater that the extensive literature review has been published [15]. Additionally, the wastewater treatment of colored and high strength by UV/H_2O_2 process has been studied effectively such that Shu et al. [16,17] expressed the feasibly effective decolorization of two prepared di-azo dyes in laboratory by this process while identifying the optimal operating parameters. Similarly, Kurbus et al. [18] demonstrated that the UV/H_2O_2 process can also successfully decolorize six vinylsulphone reactive dyes, yet this process is seldom applied on field industrial manufacturing effluent, neither the landfill leachate.

The purpose of this study was to evaluate the treatment feasibility for landfill leachate using the UV/H_2O_2 process with 4-UV lamps in the annular photoreactor design. The decolorization and COD removal efficiencies were studied by identifying the operating parameters such as hydrogen peroxide concentration, UV input power and wastewater strength.

2. Experimental

2.1. Materials

Hydrogen peroxide (35%, w/w) used was from Fluka Chemical. The wastewater sample was collected directly from the leachate-holding tank of Shalu landfill in central Taiwan. TS of 7750 mg l^{-1}, SS of 1150 mg l^{-1}, pH of 7.8, COD of 3750 mg l^{-1} and PtCo color unit of 8250 were measured from the original leachate.

2.2. Apparatus

In this work, the recirculation batch reactor system consisting four thin gap annular UV reactors was conducted that each reactor was equipped with a Sterilight® low pressure mercury arc UV lamp (wavelength 253.7 nm, input power of 36 W, light intensity reading at quartz tube was provided by manufacturer as 30,000 $\mu W\ cm^{-2}$) in the inner quartz tube shown in Fig. 1. The scheme comprised outside diameter of the quartz tube 2.2 cm, inside diameter of stainless shell of 3.2 cm, the reactor gap size of 0.5 cm and the length of annular reactor of 90.0 cm, so that made a hold up volume of 381.7 cm^3 for each annular reactor. The recirculation of leachate was pumped from a 2.0 dm^3 water reservoir through four annular reactors one by one sequentially with 1.0 $dm^3\ min^{-1}$ of flow rate, next flowing back to the mixing reservoir counter-clockwise while the flow passing by means of the thin gap between quartz tube and outer stainless steel shell of the annular reactors where the oxidation process occurs. The hold up volume of annular photoreactor was about 16% of total reaction volume. Thus, the real residence time was only about one-sixth of the appeared residence time.

2.3. Analyses

The true color of landfill leachate was measured by Platimun–Cobalt (PtCo color unit) Standard Method using Hack® DR/2500 Spectrophotometer. Chemical oxygen demand (COD) was measured by Standard Method 5220C.

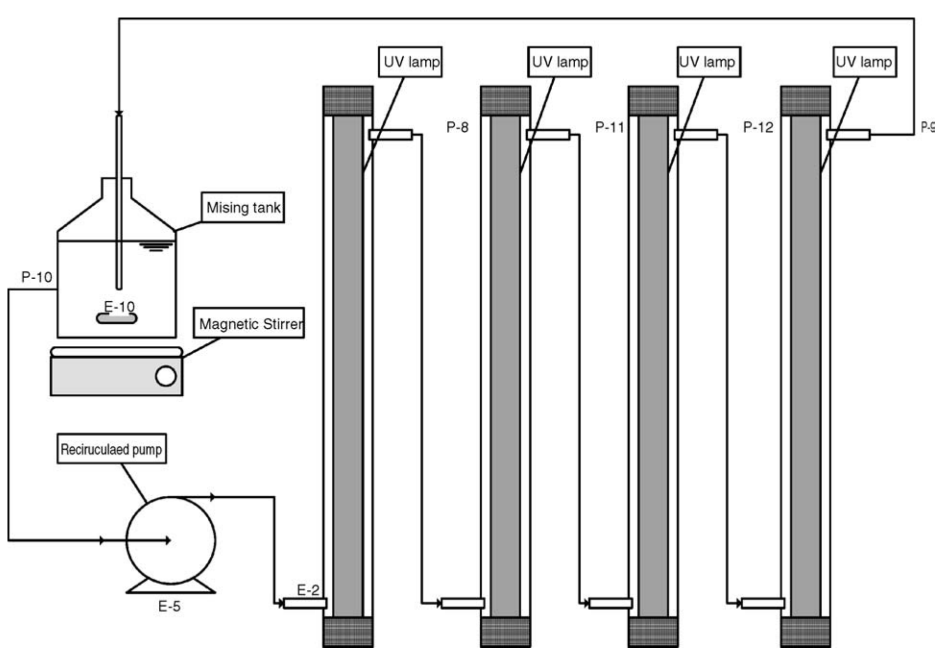

Fig. 1. The schematic drawing of the multi-UV lamps annular UV/H_2O_2 photoreactor.

3. Results and discussion

3.1. Decolorization and COD removal without UV power

Base on the previous study, dye wastewater with Acid Orange 10 was barely decolorized by hydrogen peroxide alone without UV power [19]. In this work, two strengths of leachate, such as 100 and 20%, were prepared for decolorization and COD removal by hydrogen peroxide alone without UV power while very long reaction time demand for decolorizing the leachate. The effect of water strength on the decolorization and COD removal by hydrogen peroxide alone was shown in Fig. 2, while the two leachate strengths of 100 and 20% as well as the hydrogen peroxide concentration of 232.7 mM. In reaction time of 600.0 min, decolorization and COD removal of leachate were shown in Fig. 2a and b, respectively. In the upper figure, the original leachate containing PtCo color units of 12,275 declined to 5985. Yet there was nearly residual color in 20% diluted water, which was decolorized more efficiently than that of original 100% strength leachate. In Fig. 2b, COD removal of the original leachate was removed from 4030 to about 1628 mg l^{-1}. Thus, for 100 and 20% leachate strength, the decolorization rate of 52.3 and 67.9% as well as the COD removal of 59.6 and 79.2%, respectively, was obtained. This was fairly ineffective by hydrogen peroxide alone in comparison with UV/H_2O_2 process for decolorization and COD removal. In spite of achieving more than 50% of color and COD removal efficiencies, the long reaction time demand of 600.0 min suggests the inappropriate application by hydrogen peroxide alone. During the analysis of leachate, the contribution of COD by hydrogen peroxide was taken into account of COD removal. Since the effluent standards require treated leachate to meet 150 mg l^{-1} standard, no matter the COD is from leachate or reaction reagent such as H_2O_2.

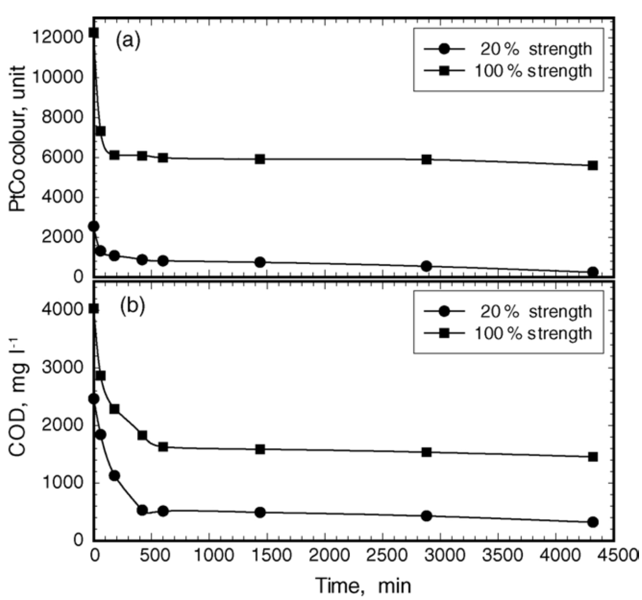

Fig. 2. The residual (a) PtCo color unit and (b) COD for 100 and 20% strength landfill leachate by reaction with hydrogen peroxide alone without UV irradiation. The operating condition was under H_2O_2 initial concentration of 232.7 mM and 4-UV lamps turn off. The PtCo color units were 12,275 and 2550 for 100 and 20% strength leachate, respectively. The COD values were 4030 and 2462 mg l^{-1} for100 and 20% strength leachate, respectively.

3.2. The effect of hydrogen peroxide concentration on decolorization

The leachate with high pollution strength is refractory to the free radical oxidation so that the higher hydrogen peroxide concentration was utilized in order to improve the oxidation in UV/H_2O_2 process. In Fig. 3a, the higher hydrogen peroxide dosage promoted the more significant decolorization such as 15.7, 25.9, 27.6 and 70.5% of decolorization achieving by 52.8, 116.4, 174.5 and 232.7 mM of hydrogen peroxide dosage in 150.0 min. The color removal efficiencies of leachate illustrated no significant differences by adding the lower hydrogen peroxide concentrations of 52.8, 116.4 and 174.5 mM that half of the best decolorization was obtained by adding 232.7 mM of hydrogen peroxide concentration. The decolorization was observed by adding 232.7 mM of concentration not only approaching the best but also incrementing sharply indicating possibly this was about the most suitable amount among studied cases. Less than 232.7 mM, the free radicals may not fully react in original leachate. Yet diluted water strength of 50% improved this situation shown in Fig. 3b that color removal efficiency was dependent on the hydrogen peroxide concentration proportional. For 150.0 min of reaction time, 43.9, 54.3, 77.6 and 92.9% of decolorization were obtained with hydrogen peroxide dosage of 58.2, 116.7, 174.5 and 232.7 mM, respectively. The decolorization was definitely increased in 50% strength of leachate than that of 100% strength of origin, as well as less reaction time demand. Though the optimum hydrogen peroxide concentration for deducing organic pollutants in the laboratory aqueous solution by UV/H_2O_2 process was demonstrated by some studies such that 46.53 mM of H_2O_2 for a C.I. Acid Blue 113 (29.30 μM) [17], 24.5 mM of H_2O_2 for a Disperse Red 354 (45.76 μM) [20]. In this work, hydrogen peroxide concentration of 232.70 mM were much higher than the above so that the leachate was much difficult to be treated. As long as the correct operating parameters are alerted, the UV/H_2O_2 process still was suggested an effective technology for the decolorization of landfill leachate.

3.3. The effect of hydrogen peroxide concentration on COD removal

Except that the effect on decolorization of leachate by hydrogen peroxide dosages in UV/H_2O_2 process was discussed above, the effectiveness of leachate treatment represented by COD removal were applied in this experiment while leachate strength of 100 and 50% shown in Fig. 4a and b, respectively. Similar to color removal, COD removal of

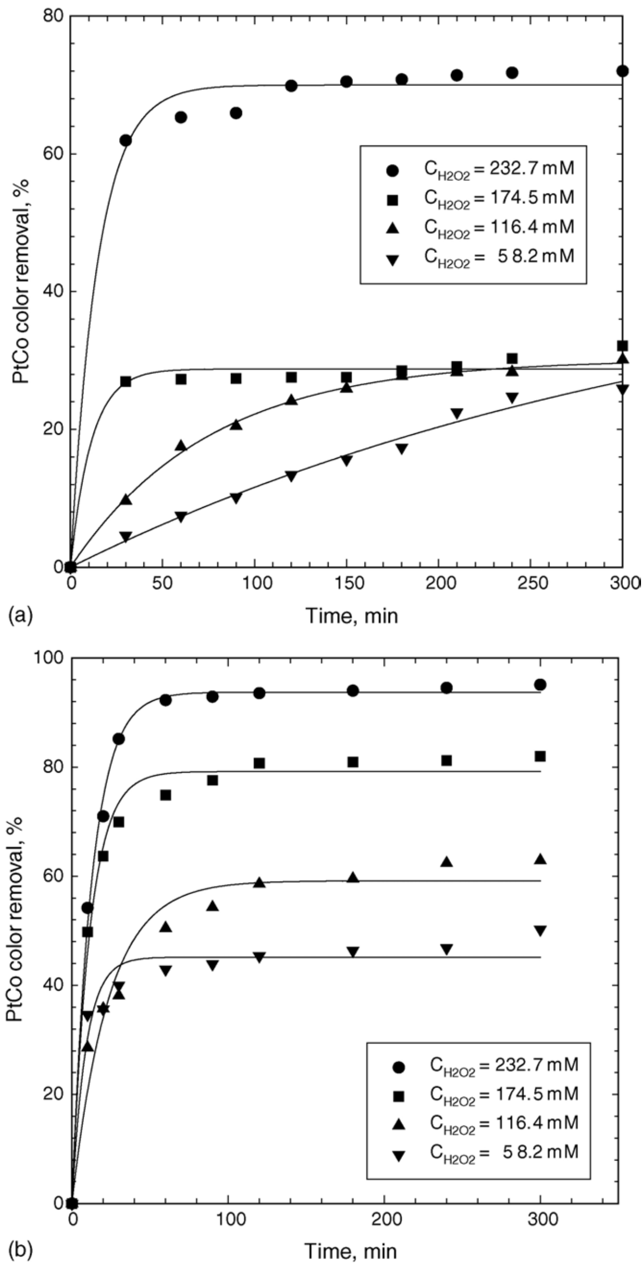

Fig. 3. The color removal efficiencies of leachate vs. time for various hydrogen peroxide concentrations in UV/H_2O_2 reactor system. The operating condition was under H_2O_2 initial concentration of 58.2–232.7 mM and 4-UV lamps turn on: (a) for 100% strength leachate and (b) for 50% strength leachate.

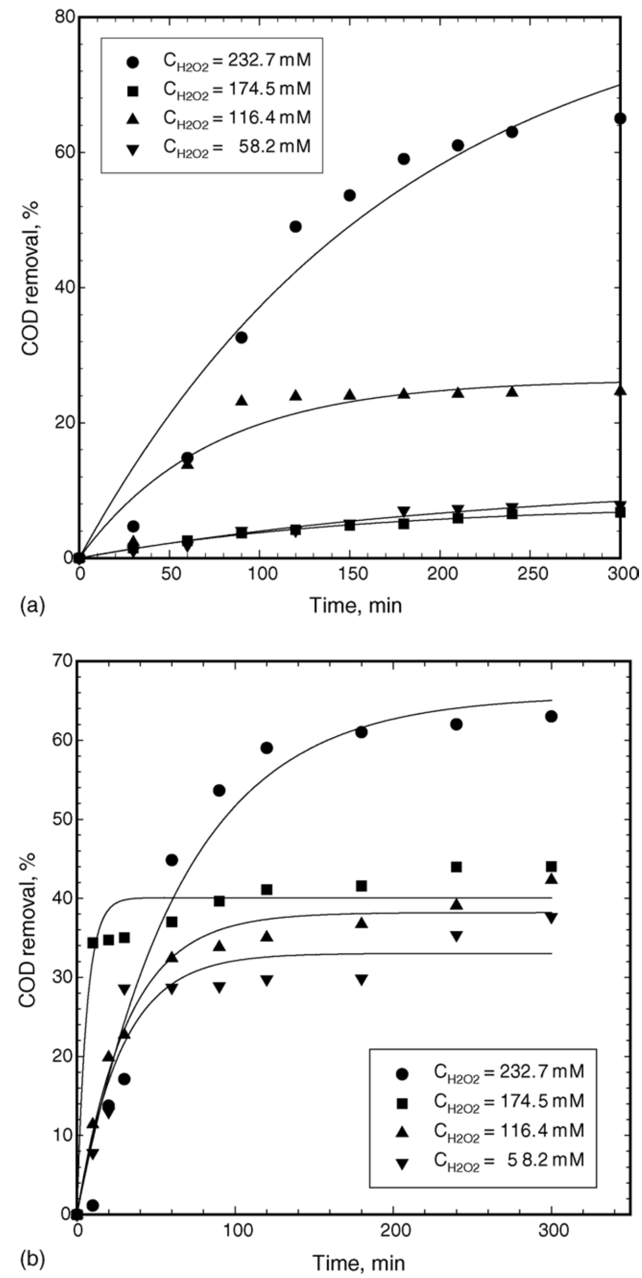

Fig. 4. The COD removal efficiencies of leachate vs. time for various hydrogen peroxide concentrations in UV/H_2O_2 reactor system for (a) 100% strength leachate and (b) 50% strength leachate. The operating conditions were the same as those given in Fig. 3.

leachate by this UV/H_2O_2 process was definitely visible at high hydrogen peroxide concentration of 232.7 mM. Once less than 174.5 mM, less effective COD removal resulted while 53.6 and 5.2% of COD removal achieving by hydrogen peroxide dosage of 232.7 and 58.2 mM, respectively, in 150 min. However, less reaction time demands reaching higher COD removal by lower strength (50%) leachate is shown in Fig. 4b, that the COD removal was dependent upon the hydrogen peroxide dosage. For example, in 90 min, COD removal of 53.6 and 28.9% appeared by adding hydrogen peroxide dosage of 232.7 and 58.2 mM, respectively. Hence, COD removal was more ineffective than decolorization using this UV/H_2O_2 process at the same strength leachate of 50%.

3.4. *The effect of UV power on decolorization and COD removal*

The wastewater with high strength color such as landfill leachate is generally regarded as to be treated rebelliously. Especially by the UV-irradiation technology, the

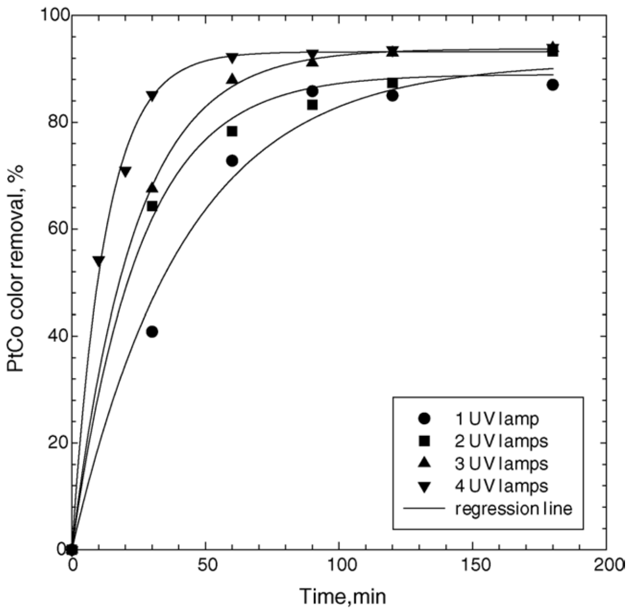

Fig. 5. The color removal efficiencies of 50% strength leachate vs. time under various UV power for UV/H_2O_2 reactor system. The H_2O_2 initial concentration was 232.7 mM. The PtCo color unit was 4150 and COD value was 1732 mg l^{-1}.

strong strength of wastewater absorbs UV resulting deduction of photo-efficiency, which irradiates hydrogen peroxide molecules into less free radical. Hence, the oxidation in UV/H_2O_2 process is promoted positively by increasing UV power in order to overcome the difficult treatment of leachate so that the reactor with four 36-W UV lamps was utilized in this work. The UV power resulting decolorization and COD removal was illustrated in Figs. 5 and 6, while the leachate of 50% strength with PtCo color of 4150 units, COD of 1732 mg l^{-1} and UV absorbance at 254 nm. The higher UV lamp power produced more and faster formation of $OH^{\bullet}$ free radicals so as to improve the decolorization rate. The mathematic equations were developed as the pseudo-first-order decolorization and COD removal of leachate in the UV/H_2O_2 process as follows:

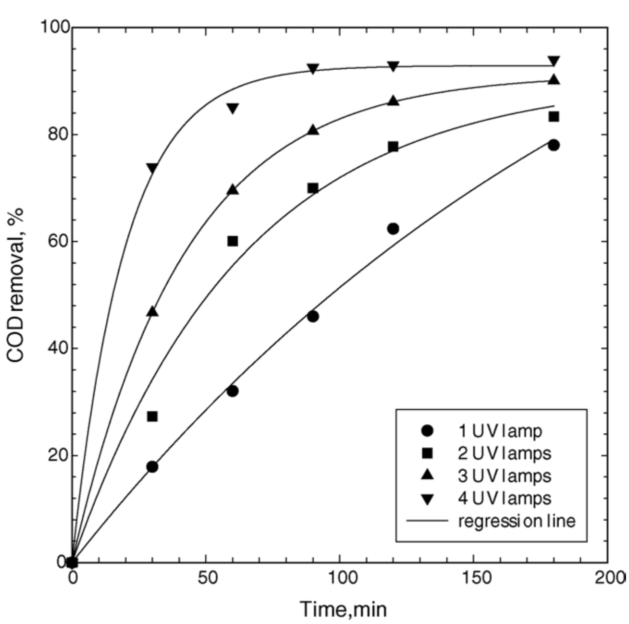

Fig. 6. The COD removal efficiencies of leachate vs. time under various UV power for UV/H_2O_2 reactor system. The operating conditions were the same as those given in Fig. 5.

$$\frac{C}{C_0} = e^{-kt} \tag{1}$$

$$-\ln\left(\frac{C}{C_0}\right) = k \times t \tag{2}$$

$$\text{Removal efficiency (\%)} = 100 \times \left(1 - \frac{C}{C_0}\right) = 100 \times (1 - e^{-kt}) \tag{3}$$

where k denotes the observed first-order reaction rate constant, t expresses the reaction time, C_0 the initial PtCo color unit (or COD) of leachate and C is the PtCo color unit (or COD) of leachate at any time t. Hence, k can be calculated by linear regression. Though part of color and COD were not able to be removed because of refractory property of leachate, the modified removal efficiency equation was developed as follows:

$$\text{Removal efficiency (\%)} = R_{\text{ultimate}} \times 100 \times (1 - e^{-kt}) \tag{4}$$

where R_{ultimate} denotes the ultimate removal efficiency of PtCo color unit (or COD).

In Fig. 5, the decolorization followed pseudo-first-order reaction obtaining the observed rate constant of 0.0801, 0.0431, 0.0397 and 0.0239 min^{-1} while addition of 232.7 mM of hydrogen peroxide concentration and 4-, 3-, 2- and 1-UV lamps, respectively. Significantly, the rate constant enlarged by incrementing UV power, which irradiating H_2O_2 to produce more $OH^{\bullet}$ radicals resulting into the growth of decolorization. On the other hand, the obtained rate constant can be used to design a real treatment process for landfill leachate. From Fig. 6, the trends of COD removal were fairly similar to that of color removal while the identical conditions used in Fig. 5. For example in 120.0 min, COD removal was 93.0, 86.1, 77.8 and 62.4% using 4-, 3-, 2- and 1-UV lamps, respectively. Hence, COD removal was proportionally dependent on the UV power.

3.5. The effect of leachate strength

The original leachate was prepared into various strengths of 20, 50 and 100%. The effect of leachate strength on decolorization was shown in Fig. 7, while hydrogen peroxide concentration of 232.7 mM and 4-UV lamps employed. From the figure, the decolorization of leachate by this UV/H_2O_2 process was effective, that the most difficult to be decolorized was occurred by water strength of 100%. In 60.0 min, the decolorization of 65.3, 92.3 and 88.9% was obtained by the

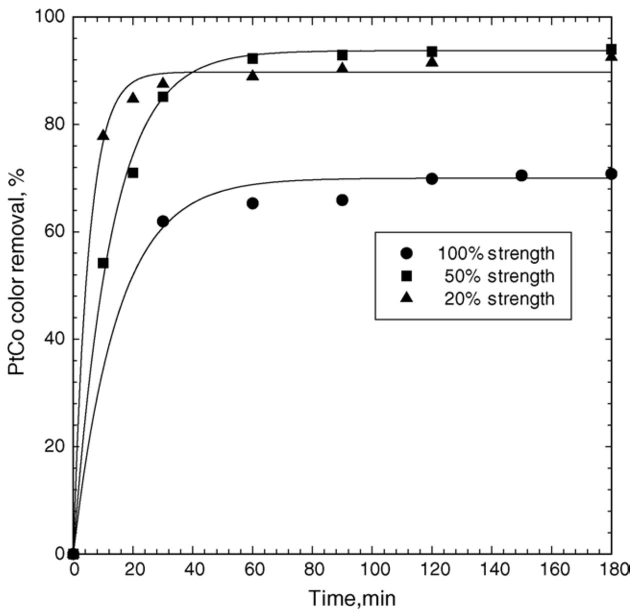

Fig. 7. The color removal efficiencies of leachate vs. time under various leachate strengths in UV/H_2O_2 reactor system. The operating condition was under H_2O_2 initial concentration of 232.7 mM and 4-UV lamps turn on.

water strength of 100, 50 and 20%, respectively. Furthermore, COD removal by various leachate strengths increased finitely shown in Fig. 8, that COD removal of 59.2, 61.5 and 88.1% was obtained while water strengths of 100, 50 and 20%, respectively, in 180.0 min. Therefore, it is significantly observable that raised removal efficiency of PtCo color and COD by declining leachate strength in this UV/H_2O_2 process.

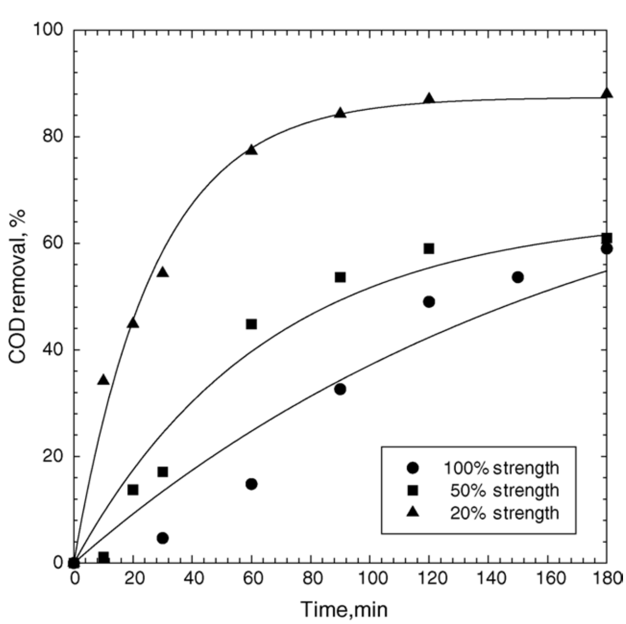

Fig. 8. The COD removal efficiencies of leachate vs. time under various leachate strengths in UV/H_2O_2 reactor system. The operating conditions were the same as those given in Fig. 7.

4. Conclusions

The thin gap annular UV/H_2O_2 reactor system with 4-UV lamps was designed to increase UV power input, which effectively decolorized and mineralized the landfill leachate. From the experimental results, moreover, the greatest removal efficiencies of both color and COD were observed while keeping the highest dosage of hydrogen peroxide and UV power input as well as the most diluted leachate. In addition, the lower water strength of leachate approached not only the better removal of both color and COD, but also the more rapid reaction, demanding less time. Hence, with the maximum dosage of 4-UV lamps and 232.7 mM of hydrogen peroxide concentration, the color and COD removal were 72 and 65% for original leachate in 300 min, while 91 and 87% for 20% of leachate strength in 120 min, respectively.

Acknowledgement

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