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Removal of Colour from Landfill by Solar Photocatalytic

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Abstract: This study was conducted to examine the percentage of colour removal using coagulation as pretreatment, followed by the effects of ZnO, pH, stirring time and solar photocatalytic as final treatment. The duration of photocatalytic reaction occurs after 30 min of exposure to sunlight at solar intensity of 0.13- 0.89 kW m⁻² that is from 11:00-1:00 p.m. The study showed that, pretreatment of leachate by coagulation at no pH adjustment, at its natural pH 8.68 with addition 10 g L⁻¹ FeCl₃ as coagulant can remove 67.2% colour. But, 35% colour removal can be obtained by coagulation at pH 5 with addition 1 g L⁻¹ FeCl₃. Using 1.0 g L⁻¹ ZnO at pH 5 capable of removing 97% colour after 120 min of exposure. However, when lower amount of ZnO (0.2 g L⁻¹) was used together with addition H₂O₂ (50 ml L⁻¹) at pH 5, the removal of colour was 95.3%.

Key words: Landfill, leachate, ZnO, coagulation, colour

INTRODUCTION

Leachate produced from landfill contains large quantities of organic and inorganic matters and heavy metals. For the removal of these matters, heterogeneous photocatalysis could be applied. In photocatalytic process, hydroxyl radicals can be generated when the photocatalyst, such as TiO₂ and ZnO, is illuminated and these ultra-reactive species lead to successful mineralization of pollutants to CO₂, H₂O and other minerals. The studies in which heterogeneous photocatalysis on TiO₂ was a useful method for the elimination of toxic and non-biodegradable and refractory organic and inorganic components from landfill leachate by their transformation into harmless species, have been reported by Cho *et al.* (2002).

Therefore, for this study a semiconductor, ZnO and H₂O₂, was chosen to treat leachate. The parameters that were focused in this study is colour.

Semiconductor, notably zinc oxide is known to be a photocatalyst. Illumination of these oxides will produce excited electrons that will react with water or hydroxyl ions to generate hydroxyl radicals (Saha and Chaudhuri, 2003). The hydroxyl radical is the primary oxidizing agent. The hydroxyl radicals react with the leachate, completely mineralizing most compounds, leaving carbon dioxide, water and mineral acids as products (Saha, 2001). Semiconductor photocatalysis has had its usefulness explored as a viable alternative technology to classical best technologies.

Hydrogen peroxide (H₂O₂) is a clear liquid, slightly more viscous than water. It has strong oxidizing properties and is therefore a powerful bleaching agent that has found use as a disinfectant and as an oxidizer. According to Rabindranathan *et al.* (2003), addition H₂O₂ enhances the rate of TiO₂ catalysed photodegradation of phosphamidon and the combination efficiency of the catalyst and H₂O₂ is more than the sum of the effect of H₂O₂ and the catalyst.

This study focuses on the leachate generated from Pulau Burung Landfill Site (PBLs), which has a semi-aerobic system. The characteristics of raw leachate from old detention pond at Pulau Burung Landfill Site (PBLs) are still very high. So, there must be treatment of leachate before it discharged to surface water. Table 1 shows details of the main characteristics of leachate used in the experiment.

Table 1: The characteristic of raw leachate from detention pond at Pulau Burung Landfill Site

Parameters	Values	Standard B*
pH	7.8-9.4	5.5-9.0
COD (mg L ⁻¹)	1533-3600	100
BOD (mg L ⁻¹)	48-1120	50
Turbidity (NTU)	50-450	-
Suspended solid (mg L ⁻¹)	159-1120	100
Colour (PtCo)	2430-8180	-
Zinc (mg L ⁻¹)	0.1-1.8	1
Copper (mg L ⁻¹)	0.1-0.4	1
Manganese (mg L ⁻¹)	0.6-1.1	1
Cadmium (mg L ⁻¹)	< 0.04	0.02
Iron (mg L ⁻¹)	0.32-7.5	5

*Standard B of the environmental quality (Sewage and Industrial Effluents) Regulations 1979, under the Environmental Quality Act of Malaysia, 1974

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MATERIALS AND METHODS

Coagulation as pretreatment process: There were two methods that have been done before coagulation. First, adjust pH to 5. One thousand milliliter raw leachate poured into 1 L high shape beaker for coagulation. Before coagulation, sulphuric acid (H₂SO₄) is used for pH adjustment raw leachate to pH 5. According to Hua (2005), the highest removal of COD by addition of ferric chloride was at pH 5 which the percentage of removal was 9%. This is similar with the optimum pH 4.9 found by Amokrane *et al.* (1997) for the removal of 55% COD in landfill leachate using ferric chloride as coagulant.

Secondly, without pH adjustment. To find optimal coagulant dose, ferric chloride was added to 500 mL raw leachate sample. The coagulant doses of ferric chloride used range from 1 to 15 g L⁻¹, at varying increments.

Coagulation studies on leachate were performed with jar test equipment (Jar Tester Model CZ150) comprising six paddle rotors (24.5×63.5 mm), equipped with 1 beaker of each. A pre-determined concentration of ferric-chloride was added to the conditioned leachate. The initial rapid mixing stage for the both experiments were 1 min at 80 rpm, followed with slow mixing stage for 15 min at 30 rpm. The final gravity settling stage lasted for another 1 h before sampling.

After 60 min of settling time (Aziz *et al.*, 2004), the colour of the supernatant were determined. The supernatant from this pretreatment step was then removed by using the pipet. Five hundred milliliter of this supernatant was taken and kept for the next step.

Treatment of leachate

Batch studies on different concentration of ZnO and/without H₂O₂: To study zinc oxide-mediated photocatalytic degradation, concentration of leachate was chosen. In each case, one light control (without ZnO) was used. Five hundred milliliter of the supernatant (without ZnO) from first pretreatment was taken and measured for colour.

Secondly, 500 mL of the supernatant were taken in a 1000 mL glass beaker to which zinc oxide (ZnO) and/without hydrogen peroxide (H₂O₂) was added and kept in suspension by constant stirring and exposed to sunlight to allow photooxidation to occur in 30 min to 2 h. A 20 mL aliquot will be drawn out at every 30 min interval and filtered through a 0.45 µm membrane filter. The filtrate will be diluted 1 in 10 and then tested for colour.

RESULTS AND DISCUSSION

Pretreatment by Coagulation-Flocculation Using FeCl₃: Leachate is dark brown or black in colour. Coagulation-

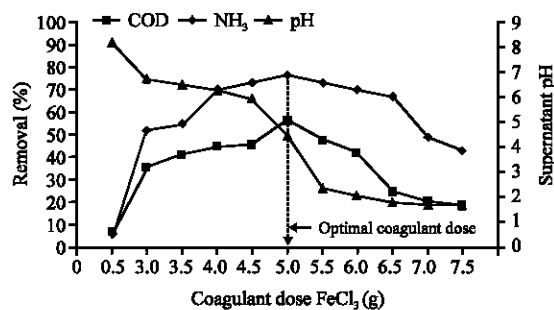


Fig. 1: pH of supernatant and removal (%) of COD and NH₃ vs. coagulant dose (with no pH adjustment) in 500 mL leachate

flocculation is to remove this dark colour first in order to allow for photooxidation to occur. Ferric chloride (FeCl₃) was chosen as a pretreatment for coagulation-flocculation process. The leachate samples were then adjusted to pH 5 before the addition FeCl₃ (1 g L⁻¹). The amount of colour, ammonia (NH₃) and COD removal were determined after coagulation-flocculation. In the next stage, no pH adjustment was made before the addition FeCl₃ (optimal coagulant dose) (Fig. 1). The amount of colour, ammonia (NH₃) and COD removed were again determined and compared to the first.

Based from the results, the coagulant dose that gave the highest percentage COD and ammonia removal was chosen as the optimum coagulant dose. The highest removal of COD and ammonia was obtained by addition 10 g L⁻¹ FeCl₃. The percentage removals were 55.8 and 75.8%, respectively.

The pH for the supernatant decreased continually with increasing coagulant dose and gave a value of 4.43 with optimal coagulant dose of FeCl₃. This pH decrease can be explained by the acidic character of Fe³⁺ (acid of Lewis), when reacting with OH⁻ ions to form iron precipitated in the forms of Fe(OH)₃ (Amokrane *et al.*, 1997).

With increasing coagulant dose, the dark brown colour of raw leachate turned to become clear yellow for the optimal coagulant dose near the optimum coagulant dose. For each coagulant dose tested, the settling times required were 2 h. These results were similar to that obtained by Amokrane *et al.* (1997).

Stage 1: Batch studies on photooxidation using different concentration of zinc oxide (ZnO): Figure 2, the final colour of the leachate dissolved was 120 Pt-Co which means a 97% reduction. It shows that 97% colour removal occurred in 90 min with ZnO dose of 1.0 g L⁻¹ and solar intensity of 0.13-0.89 kW m⁻².

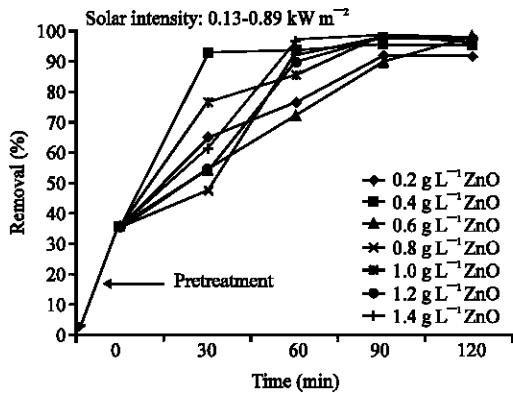


Fig. 2: Percent removal of colour using photooxidation and different concentration of ZnO

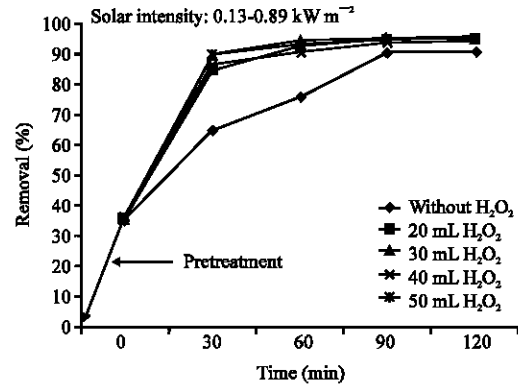


Fig. 3: Percentage removal of colour using 0.2 g L⁻¹ ZnO and H₂O₂

Stage 2a: Batch studies using 0.2 g L⁻¹ zinc oxide (ZnO) and different concentration of H₂O₂: Hydrogen peroxide corresponds to two-electron transformations of oxygen and water. It is formed under photocatalytic conditions. It is also degraded over band-gap irradiated semiconductors even at irradiating energies insufficient for its direct photolysis. At neutral or acidic pH, the reduction of superoxide via OH⁻ ions to produce OH• radicals is also improbable.

From the Fig. 3, preliminary studies have shown that addition of H₂O₂ enhances the rate of ZnO catalysed photodegradation of leachate is more than without adding H₂O₂. The final colour of the leachate was 195 Pt-Co-a 95.3% reduction in 120 min when using 50 ml L⁻¹ H₂O₂. If without using H₂O₂ the final results for colour was 91% (310 Pt-Co) reduction in 120 min.

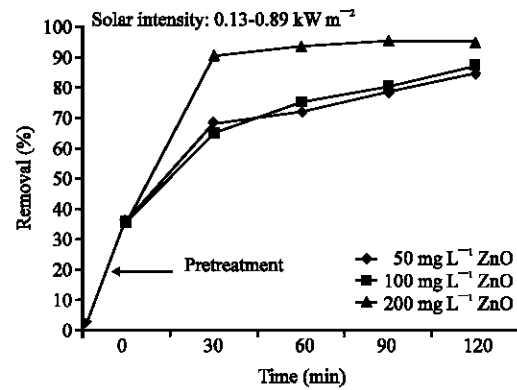


Fig. 4: Percentage removal of colour using 50 ml L⁻¹ H₂O₂ and ZnO

Stage 2b: Batch studies on 50 ml L⁻¹ H₂O₂ and different concentration of zinc oxide (ZnO): From the Fig. 4, it can be seen that when we can see that using 0.2 g L⁻¹ ZnO and 50 ml L⁻¹ H₂O₂ gives the best result for removing colour. Figure 4, the final colour of the leachate was 195 Pt-Co. It shows that 95.3% colour removal occurred in 120 min.

The present study clearly shows that semiconductor oxides can be used as effective photocatalysts for the degradation of leachate. ZnO increased the efficiency to degrade leachate parameters such as colour. To reduce the usage of zinc oxide (ZnO), hydrogen peroxide (H₂O₂) was added that resulted in lower amount of dissolved Zn ions.

The duration of photocatalytic reaction occurs after 30 min of exposure to sunlight at solar intensity of 0.13- 0.89 kW m⁻² that is from 11-1 pm. The reaction increases with increase in exposure to sunlight. Thirty minutes to 2 h exposure is sufficient to cause significant removal of colour.

pH is a very important parameter for photocatalytic removal for colour in landfill leachate. Thus, it could be easily expected that when landfill leachate is at acidic pH, it will reduced colour but it will also dissolved more Zn ions. At pH 5 degradation rates of colour was significantly higher than at alkaline pH.

The best amount of ZnO used for optimal removal of colour is 1 g L⁻¹ at pH 5. This result is 97% colour removal. However, when lower amount of ZnO was used together with the addition of H₂O₂ at pH 5, the removal of colour slightly reduced. When using 0.2 g L⁻¹ ZnO and 50 ml L⁻¹ H₂O₂, the removal for colour was 95.3%, respectively.

But, when this result compared to the sample which is in different concentration of FeCl₃ with no pH adjustment, there were significant differences. When 0.2 g L⁻¹ ZnO used together with addition of 50 ml L⁻¹ H₂O₂ at pH 5, the removal for colour is 95.3% (195 Pt-Co) using 1 g L⁻¹ FeCl₃.

CONCLUSION

The leachate pretreatment, coagulation was found to be effective in reducing the colour. The reactions between 50 ml L⁻¹ H₂O₂ and 0.2 g L⁻¹ ZnO for treated leachate at pH 5 using 1 g L⁻¹ FeCl₃ could remove the colour significantly. It seems that when ZnO added into leachate with no pH adjustment, at its natural pH 8.68, the Zn ions dissolved is lower than when ZnO added into leachate at acidic pH 5.

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