

## Landfill Leachate Treatment by a Coagulation–Photocatalytic Process

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**Abstract.** This study was conducted to investigate the efficiency of coagulation-photooxidation processes for removing color from a landfill leachate by using *Moringa Oleifera* as a coagulant as a pre-treatment. Then, followed by the effects of ZnO, pH, stirring time and solar photocatalytic as final treatment. Final treatment was examined the effectiveness of zinc oxide (ZnO) as a catalyst to enhance the removal of color under sun exposure. Based on the observation towards the colours of the samples during the experiment, sample with 0.2g of ZnO has the lightest yellow colour, almost colourless. The duration of photocatalytic reaction occurs after 30 minutes of exposure to sunlight that is from 12 – 2 p.m. Result indicated that 0.2g of ZnO gives the highest removal of color which is 90.1% at 60 minutes.

**Keywords:** Leachate, coagulation-flocculation, *Moringa oleifera*, color

### 1. Introduction

Sanitary landfill for the disposal of municipal solid wastes continues to be widely accepted and used in the several countries. This method generally offers lower cost of operation and maintenance as compared to others. However, leachate migration from the landfill could be a potential source of surface and groundwater contaminations.

As we know, landfill leachate is a very dark colored liquid formed primarily by the percolation of precipitation through open landfill or through the cap of the completed site. The decomposition of organic matter such as humic acid may cause the water to be yellow, brown or black (Zouboulis, A.I. et al., 2004). Combinations of physical, chemical, and biological treatments are usually used to improve the treatment efficiency of landfill leachates (Kargi, F., and Pamukoglu, M.Y., 2004). There are several techniques used for colour removal. These include chemical precipitation, adsorption through granular activated carbon, nanofiltration, ozonation, radiation, UV photolysis, chemical coagulation, biological treatment with various additives, anaerobic process, fluidized bio film process, and advanced oxidation with UV/H<sub>2</sub>O (Ahmedna et al., 2000, Kadirvelu et al., 2003 and Manu, B., 2002).

However, there is no specific guideline for the treatment of color in landfill leachate, especially in Malaysia. Coagulation followed by flocculation process is an effective way for removing high concentration of organic pollutants (Wang, Z.P. et al., 2002) as well as the color in leachate.

In this field of research, the performance of color removal will be evaluated. Addition of ZnO as a catalyst was found to give a significant effect and enhancing the removal of color from landfill leachate samples after coagulation-flocculation process.

### 2. Materials and Methods

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## **2.1. Materials**

### **2.1.1. Preparation of *Moringa oleifera* seeds powder**

The *Moringa oleifera* was obtained from Kampung Santan, Perlis area. Good quality seeds were identified from those, which were not rotten, old, infected with diseases, brownish and dried once opened. The seeds were dried in the oven (Memmert, ULE 400, Germany) for 24 h at 50 °C. The kernels were crushed and ground to a medium fine powder with a domestic food blender[(Katayon et. Al., 2006)].

### **2.1.2. Preparation of *Moringa oleifera* seeds extract**

A 5000 mg of *Moringa oleifera* seeds powder was placed in a beaker containing 0.2 l of distilled water. The mixture was blended using domestic blender for 2 min at high speed to extract the active ingredient of *Moringa oleifera*. The suspension was then filtered through a muslin cloth in a beaker and the filtrate made up to 0.5 l to give a stock solution of 10,000 mg/l. 10,000 mg/l of *Moringa oleifera* stock solution was used for jar test that were conducted to determine optimum dosages of *Moringa oleifera* in leachate treatment. [(Katayon et. Al., 2006)].

### **2.1.3. Preparation of leachate samples**

Landfill leachate sample were collected from the landfill site at Pauh, Perlis area. Samples were collected in leachate sample was collected in four bottles of PE (polyethylene) and transported to the laboratory and stored in the chemical refrigerator.

## **2.2. Methods**

### **2.2.1. Coagulation test**

Coagulation test was carried out using jar test (BIBBY Stuart Scientific, UK). The study involved rapid mixing, slow mixing and sedimentation in a batch process. Six glass beakers of 500 ml capacity were filled with the water samples and agitated simultaneously, and the rotational speed were varied accordingly, allowing simulation of different mixing intensities and resulting flocculation process.

### **2.2.2. Experimental runs**

Adsorption studied was conducted by varying pH and adsorbent dose in pre-treatment process while varying catalyst dose and contact time during final treatment process. Pre-treatment process was conducted to find the optimum dosage of adsorbent (MO) and optimum pH of leachate. This studied was carried out using jar test method. Final treatment process was carried to find out the optimum dosage of catalyst (ZnO) and optimum contact time in removal of COD from leachate. This final treatment process has been carried out under sun exposure. The contact time varied from 0 to 120 minutes. The percentage removal of COD landfill leachate was calculated according to the following equation:

$$\% \text{ Removal} = \frac{C_i - C_f}{C_i} \times 100$$

Where  $C_i$  and  $C_f$  are the initial and final COD concentration (mg/L) respectively.

### **2.2.3. Pre-Treatment**

#### **2.2.3.1. Determination of optimum pH**

First, sets of experimental runs were carried out for determination of optimum pH of leachate sample. Beakers were filled with 500 ml of the leachate. Solution of  $H_2SO_4$  and NaOH were being added into the

samples to obtain pH : 1, 2, 3, 4, 5, 6, 8, 10. In order to obtain the accurate pH, pH meter was being used during adjusted the pH level. Samples were placed on the floc illuminator and agitated at the preselected intensity of rapid mixing (Table 1.0). During rapid mixing (80 rpm), the coagulant dosage of *Moringa oleifera* was added into each beaker simultaneously for 20 ml for each beaker. After 1 minute of rapid mixing, the preselected intensity of slow mixing (30 rpm) was quickly established and the beakers were then carefully removed from the floc illuminator after 15 minutes and left for the sedimentation phase. After settling, 100 ml of sample was taken from the middle of each beaker and was filtered through filter paper (pore size 0.45  $\mu\text{m}$ ) for COD measurement. The pH that gave the highest 5 removal of COD was the optimum pH for leachate treatment.

### **2.2.3.2. Determination of optimum dosage of *Moringa oleifera***

Beakers were filled with 500 ml of the leachate with optimum pH. The pH of samples was maintained at a desired value by adding 1.0 M NaOH or H<sub>2</sub>SO<sub>4</sub>. During rapid mixing, different amount of *Moringa oleifera* stock solution with 5ml, 10ml, 15ml, 20ml, 30ml, 40ml, 50ml, 60ml, 70ml, 80ml, 90ml, and 100ml were added into each beaker and the samples were placed on the floc illuminator and agitated at the preselected intensity of rapid mixing.

After rapid mixing, the preselected intensity of slow mixing was quickly established and the beakers were then carefully removed from the floc illuminator and left for the sedimentation phase. After settling, 100 ml of sample was taken from the middle of each beaker and was filtered through filter paper (pore size 0.45  $\mu\text{m}$ ) for COD measurement. The dosage that gave the highest % removal of COD was the optimum dosage for leachate treatment. After evaluating the optimum dose, jar test experiments were conducted at the optimum dose.

### **2.2.4. Final Treatment**

#### **2.2.4.1. Determination of optimum dosage of Zinc oxide (ZnO) and optimum contact time**

Beakers were filled with 500ml of leachate with optimum dosage of *Moringa oleifera* and optimum pH of leachate sample. Optimum dosage of *Moringa oleifera* and optimum pH of leachate sample have been investigated in pre – treatment process above. The samples were placed on the floc illuminator and agitated at the preselected intensity of rapid mixing. After rapid mixing, the preselected intensity of slow mixing was quickly established and the beakers were then carefully removed from the floc illuminator and left for the sedimentation phase. After settling, supernatant sample was taken from the middle of each beaker and placed into another beaker. Sample was added with 0.1g of zinc oxide (ZnO) and the beaker was placed on the stirrer. The sample was being exposed to the sun (light intensity 1997 flux) from 12pm to 2pm with the speed level of the stirrer was set at 7. Sample was taken from the middle of each beaker every 30 minutes and was filtered through filter paper (pore size 0.45  $\mu\text{m}$ ) for COD measurement using DR2800 spectrophotometer. The above steps were being repeated by using different amount of zinc oxide (ZnO) which is 0.2g, 0.3g, 0.4g, 0.6g, 0.8g, 1.0g, 1.2g, and 1.4g.

## **3. Results and Discussions**

### **3.1. Result before treatment**

The initial raw leachate for color was measured before the treatment process, which is 2330 (Pt-Co).

### **3.2. Pre-Treatment**

#### **3.2.1. Determination of optimum pH and optimum dosage of *Moringa oleifera***

In this pre-treatment process, leachate was found to be effectively treated when the optimum dosage of *Moringa oleifera* at 20ml with the optimum pH is at pH 2.

### **3.3. Final treatment**

### 3.3.1. Determination of ZnO dosage and optimum contact time in removal of color

Based on the Fig. 1, sample without addition of ZnO gives the percent removal of color at 88.8% at 90 minutes and the removal being constant after 90 minutes until 120 minutes. It showed that, the sample was achieved equilibrium state and there was no change of color removal after 90th minutes. Any further addition of time beyond the optimum time did not cause any significant change in % removal of color. Eventhough the experiment is conducted without the addition ZnO, the process of coagulation flocculation using *Moringa Oleifera* as the coagulant itself does reduce the original dark brown colour of the leachate sample to yellowish.

In Fig. 1 below, the optimum contact time in removing color for ZnO dosage at 0.1g and 0.2g were at 60 minutes while for ZnO dosage at 0.4g the optimum contact time was at 90 minutes. Samples with ZnO dosage at 0.8g obtained the optimum time in removing color at 30 minutes. Percentage removal of color were decreased beyond the optimum contact time for all samples. Based on the result, there is no sample that reached the optimum contact time on the 120 minute.

Based on the result, for ZnO dosage at 0.1g to 0.4g, % removal of color were very high ranging from 88.8% to 90.1%. For ZnO dosage at 0.8g, % removal of color decreases slightly to 85.4%. From the observations during the experiment, the color of supernatant was changed when 0.2g of ZnO was added into the sample. Supernatant color turn from slightly yellow to very clear light yellow. The initial color of sample is dark brown. After the coagulation-flocculation process the color of sample turn from dark brown to yellowish. Addition of ZnO not only helps in reducing the color from yellowish to clear light yellow, it also increases the rate of removal of color. Based on the observations during the experiment, it can be seen that additional of ZnO beyond 0.4g produces cloudy yellow end product.

Based on observation towards the colours of the samples during the experiment run, sample with 0.2g of ZnO has the lightest yellow colour, almost colourless. Based on the result, it has been proved that 0.2g of ZnO gives the highest percentage removal of color which is 90.1% at 60 minutes. Any further addition of ZnO beyond this optimum dosage reduced the percentage removal of color from leachate. Removal of color from leachate decreased with the increasing of ZnO dosage because the excellent result showed at lower dosage which at 0.2g. This could be attributed by the restabilization of colloidal particulates when catalysts were used at dosage in excess of the optimum limit.

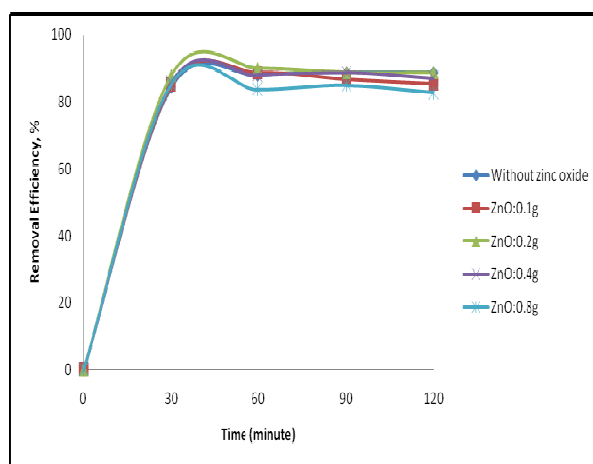


Fig. 1: Determination of ZnO dosage and optimum contact time in removal of color

## 4. Conclusions

This project was conducted to examine the percentage removal of color from landfill leachate by using coagulation as a pretreatment followed by the effect of ZnO as a final treatment. Based on observation towards the colours of the samples during the experiment run, sample with 0.2g of ZnO has the lightest yellow colour, almost colourless. Based on the result, it has been proved that 0.2g of ZnO gives the highest

percentage removal of color which is 90.1% at 60 minutes. Any further addition of ZnO beyond this optimum dosage reduced the percentage removal of color from leachate. Removal of color from leachate decreased with the increasing of ZnO dosage because the excellent result showed at lower dosage which at 0.2g. This could be attributed by the restabilization of colloidal particulates when catalyst was used at dosage in excess of the optimum limit.

## 5. References

- [1] Ahmedna et al., Ahmedna, M., Marshall, W.E., and Rao, R.M. (2000). Production of granular activated carbons from select agricultural by-products and evaluation of their physical, chemical and adsorption properties, *Bioresource Technology*, 71, pp.113–123.
- [2] Katayon, S., Megat Mohd Noor, M.J., Asma, M., Abdul Ghani, L.A., Thamer, A.M., Azni, I., Ahmad, J., Khor, B.C., and Suleyman, A.M. (2005). Effects of storage conditions of moringa oleifera seeds on its performance in coagulation, *Bioresources Technology*, 13, pp. 1455-1460.
- [3] Kargi, F., and Pamukoglu, M.Y. (2004). Adsorbent supplemented biological treatment of pre-treated landfill leachate by fed-batch operation, *Bioresource Technology*, 94, pp. 285–291.
- [4] Zouboulis, A.L., Chai, X.-L., and Katsoyiannis, I.A. (2004). The application of bioflocculant for the removal of humic acids from stabilized landfill leachates, *Journal of Environmental Management*, 70, pp. 35–41.
- [5] Amokrane, A., Comel, C., & Veron, J. (1997). Landfill leachates pretreatment by coagulation-flocculation. *Wat. Res.* 31(11), 2775-2782.
- [6] APHA., AWWA., & WPCF. (1992). *Standard Methods for Examination of Water and Wastewater*. 16<sup>th</sup> Edition. American Public Health Association, Washington.
- [7] Aziz, H. A., Alias, S., Adlan, M. N., Asaari, F. A. H., & Zahari, M. S. (2005). Colour removal from landfill leachate by coagulation and flocculation processes. *Bioresource Technology*. Article in press.
- [8] Aziz, H. A., Yusoff, M. S., Adlan, M. N., Adnan, N. H., & Alias, S. (2004). Physico-chemical removal of iron from semi-aerobic landfill leachate by limestone filter. *Waste Management*. 24, 353-358.
- [9] Bekbolet, M., Lindner, M., Weichgrebe, D., & Bahnemann, D. W. (1996). Photocatalytic detoxification with the thin-film fixed-bed reactor (TFFBR): Clean-up of highly polluted landfill effluents using a novel TiO<sub>2</sub>-photocatalyst. *Solar Energy*. 56(5), 455-469.
- [10] Bogner, J., Spokas, K., Burton, E., Sweeney, R., & Corona, V. (1995). Landfills as atmospheric methane sources and sinks. *Chemosphere*. 31(9), 4119-4130.
- [11] Chakrabarti, S., & Dutta, B. K. (2004). Photocatalytic degradation of model textile dyes in wastewater using ZnO as semiconductor catalyst. *Journal of Hazardous Materials*. B112, 269-278.
- [12] Cho, S. P., Hong, S. C., & Hong, S. (2002). Photocatalytic degradation of the landfill leachate containing refractory matters and nitrogen compounds. *Applied Catalysis B: Environment*. 39, 125-133.
- [13] Chun, C. S. (2001). Sanitary Landfills: Toward Sustainable Development. [online] [Accessed 16<sup>th</sup> June 2006]. Available at: [www.lumes.lu.se/database/Alumni/00.01/theses/choong\\_shu\\_chun.pdf](http://www.lumes.lu.se/database/Alumni/00.01/theses/choong_shu_chun.pdf)
- [14] Davis, M. L., & Cornwell, D. A. (1998). *Introduction to Environmental Engineering*. 3<sup>rd</sup> Edition. Singapore : McGraw-Hill Companies, Inc.