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The Effect of Aeration and Hydrogen Peroxide on the Electrochemical Degradation of Methylene Blue Using Carbon Composite Electrodes

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The effect of aeration and hydrogen peroxide to the electrolysis degradation of methylene blue using carbon composite has been done. Methylene blue is a dye which is most commonly used in the textile industry. In the coloring process, methylene blue only used about 5 %, while the remaining 95 % will be disposed of as waste. This compound is quite stable and non-degradable; hence it is very difficult to be degraded in nature and harmful to the environment. The aim of this study is to degrade methylene blue using electrochemical oxidation with the addition of sodium chloride as electrolyte and using carbon electrode. In this study, we use carbon composite in the electrolysis process. The advantages of carbon electrodes are low cost, wide potential window, inert and electrocatalysis activity for a variety of redox reaction. The study was conducted to determine the optimum result degradation of methylene blue by electrolysis method with aeration and hydrogen peroxide in the electrolysis process. The result of the analysis showed the aeration and hydrogen peroxide in the electrolysis process was shorter time electrolysis is 10 min. As a conclusion is the aeration and hydrogen peroxide can be used for improve performance to the electrochemical oxidation of methylene blue using carbon composite. This method is very simple equipment, easy operation and friendly for environmental.

1. Introduction

One of the most disturbing problems of textile industry waste is the dye content. In the textile industry, dye is one of the main raw materials. About 10-15 % of used dye cannot be reused and must be discarded. Substance of dye which is contained textile industry can interfere with health, such as skin irritation and eye irritation to cause cancer. In addition, the dye can also cause mutagen (Guswandi et al., 2007). The disposal of dye textile should be processed prior to disposal into the environment, this is due to the discharged dye belonging to B3 (Hazardous and Toxic) waste. One example of dye that is widely used in the textile industry is methylene blue. In the dyeing of this compound is only used about 5 % while the residue about 95 % will be disposed of as waste. This compound is quite stable and is a non-degradable compound so it is very difficult to be degraded in nature and harmful to the environment. This, of course, can damage the balance of the ecosystem of the environment. In this case it can the death of aquatic organisms around the waste disposal site so that further processing is needed to make this textile waste safe for the environment. The efforts of conventional treatment of textile waste such as adsorption and active sludge has been widely used, but the results are less effective (Ida et al., 2011). The adsorption method is less effective because the adsorbed dye accumulates in the adsorbent so that it can cause new problems (Wijaya et al., 2006).
Methylene blue is a toxic aromatic hydrocarbon compound and a cationic dye with a very strong adsorption power. In general, methylene blue is used as silk dye, wool, textile, paper, office equipment and cosmetics. Treatment of methylene blue dye has been done by electrolysis using carbon electrode (Riyanto and Mawazi, 2015). The results show that methylene blue can be degraded by electrolysis with a current and electrolysis time is 1.5 A and 60 min. In this process, methylene blue which was originally in blue color when it was

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electrolyzed change to colorless. Qualitative analysis after electrolysis methylene blue can be using UV-Vis Spectrophotometer.

Electrochemical techniques are effective method used for treatment of waste water containing contaminants from organic and inorganic compounds (Panizza and Martinez, 2013). Since the end of 1970, electrochemistry has been successfully applied to solve textile waste problems (Nordin et al., 2015), liquid wastes from olive oil (Gosti et al., 2005), tannery waste (Szpykowicz et al., 2005), and phenols (Canizares et al., 2002). Aeration technique is one of the liquid wastes processed by adding oxygen to the liquid waste. The addition of oxygen is one of the efforts of pollutant removal, so the concentration of pollutants will be reduced or even can be eliminated at all. Substances are taken may be gases, liquids, ions, colloids or other mixed materials. The effort of adding oxygen to the waste water can be through two ways that is, entering the air into the waste water and forcing the water upwards to come into contact with oxygen (Sugiharto, 2005). Air serves to consume bacteria so that by actively can eat organic content in waste. The decomposing bacteria consume organic material so that it decomposes into simple material such as CO_2, CO and H_2O. In the end CO_2 flies into the air and H_2O merges with water (Perdana, 2007). According to Riyanto and Mawazi (2015) said that a good electrode has characteristics such as good stability, conductivity, and electro catalysis. The anodes are used for oxidation must have stability in the electrolyzed waste solution, easy to produce and cheap. Carbon electrodes have better effectiveness in the effort to reduce the content of metal ions, stable, easy to get, and cheap (Riyanto and Mawazi, 2015).

Some researchers have conducted several studies with various methods of treatment of dye waste, such as there are using H_2O_2, activating carbon, zeolite, ozone, as well as with microbial methods. But there are some weaknesses in the process of dye waste that require expensive chemicals, the process is difficult to practice, not environmentally friendly, and produce new waste derived from chemicals. Based on some of the weaknesses above so the researchers proposed how to treat the waste of methylene blue dye by electrochemical method with electrolysis method. Carbon electrode can be used for electrolysis method. Carbon electrode can be chosen because carbon has better effective in order to reduce the content of metal ions, stable, inert with electrolysis solution, easy to obtain, and low price. The design of this research is expected to be used as technology development for the treatment of methylene blue using electrolysis method in NaCl solution. This research was also studies the effect of aeration and hydrogen peroxide to electrochemical degradation of methylene blue. Aeration will be produce oxygen so that in the supplied oxygen in the solution. Oxygen will speed up of the electrochemical degradation of methylene blue. Hydrogen peroxide can be used for oxidizing agent to electrochemical degradation of methylene blue.

2. Experimental methods

2.1 Solution

All solutions were prepared by dissolving its analytical grade in deionized distilled water. Sodium chloride (NaCl) from Merck was used as the electrolyte. Methylene blue (MB) was prepared from Merck (Darmstadt, Germany).

2.2 Preparation of a carbon composite electrode (C-PVC)

Carbon powder (< 2 micron in size and 99.9 % purity, Aldrich Chemical Company) and polyvinyl chloride (PVC) in 4 mL tetrahydrofuran (THF) as a solvent and swirled flatly to homogeneous followed by drying in an oven at 100 °C for 3 h. The mixture was placed in stainless steel mould and pressed at 10 t/cm^2. A typical pellet contained approximately amount of carbon powder (95 %), and approximately 5 % of PVC polymer.

2.3 Characterization of a carbon composite electrode (C-PVC) using SEM and EDS

Electrode surface was characterized using SEM from Jeol JSM-6510 LA microscope equipped with a microprobe Voyager Noran system.

2.4 Experiment procedure of the electrochemical degradation of methylene blue

The electrolysis process was performed on a solution of 20 ppm methylene blue with volume 1 L in 30 g NaCl at room temperature with current constant at 1.5 A. Experiments have been conducted in an undivided 1 L capacity electrochemical cell. Carbon composite electrode (C-PVC) was used as anode and cathode. Each electrode was connected to regulated DC power supply. The water pump was used as aeration system. The stirrer was used in electrochemical cell to maintain an unchanged composition. The solutions resulting from electrochemical degradation treatment were analysis using the Spectrophotometer UV-Visible.

3. Results and discussion

3.1 Characterization of C-PVC Using SEM-EDS

Figure 1 show the results of the analysis could be known form of carbon morphology was coarse, irregular, and forming flat particles. Then the carbon electrode after used for electrolysis it had the same morphological form. However, in the electrode after electrolysis, it could be seen that the pore holes formed were larger than the carbon before electrolysis. Electrolysis process that contained of the highest electric could cause the formation of big holes on surface of pore of electrode. The pores of the electrode surface during electrolysis occur an adsorption force which caused the exchange of selective ions during the electrolysis process. Figure 2 and Table 1 show the results of the analysis could be known between the carbon electrode before and after used electrolysis. Base on Table 1 show element composition did not cause change in the electrode in which the content of element C was almost the same between before and after the electrolysis. The percentage of other components was also the same between before and after electrolysis that contained Al, Si, and S with small quantities. Carbon electrode had good stability and capability as electrode in methylen blue electrolysis process.

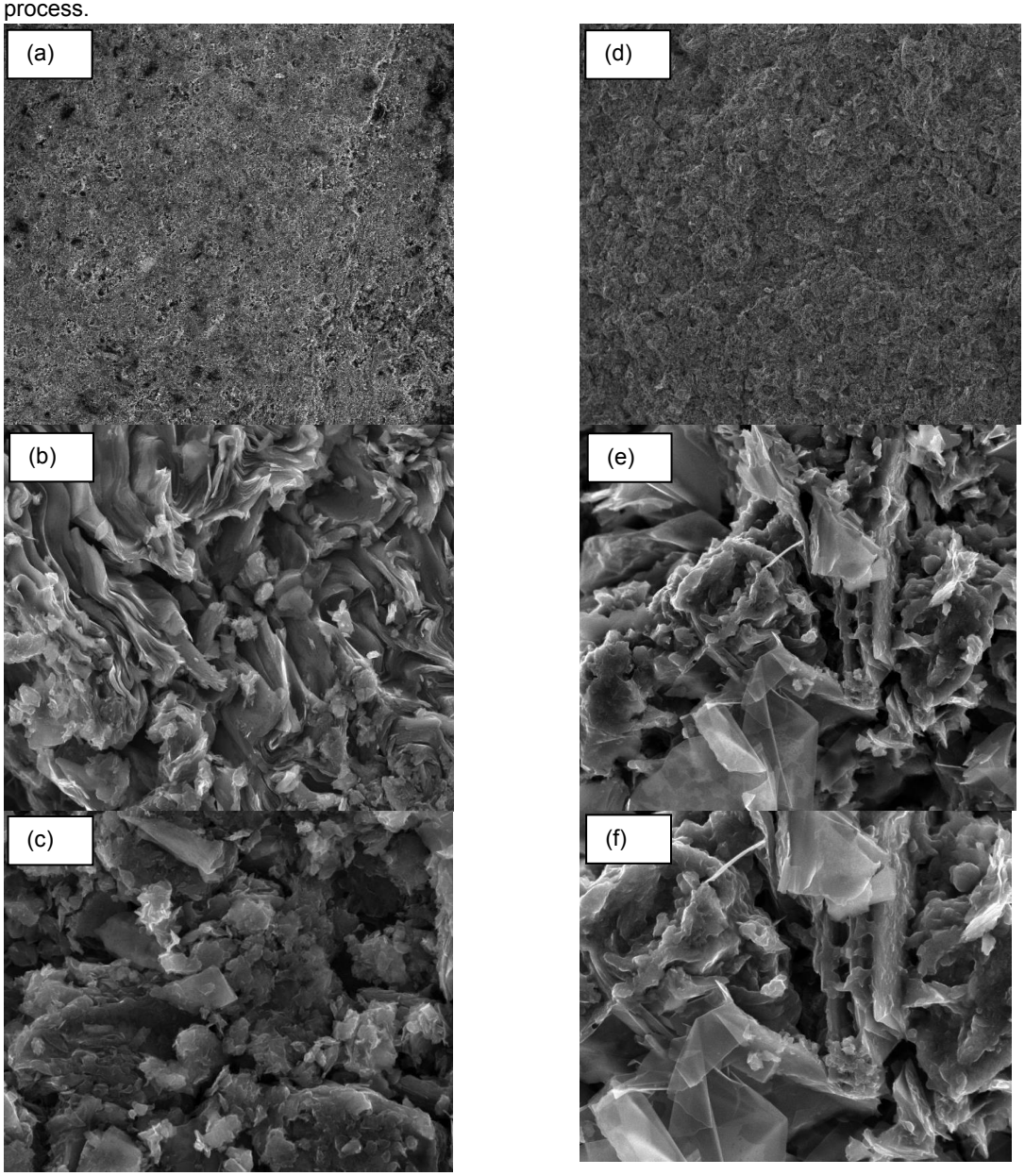

Figure 1: Results of SEM analysis of carbon electrode surface at cross section with magnification (a,d) 50x, (b,e) 3000x, (c,f) 5000x before (a,b,c) and after (d,e,f) used electrolysis of methylene blue 20 ppm (1 L) in 30 g NaCl at 1.5 A

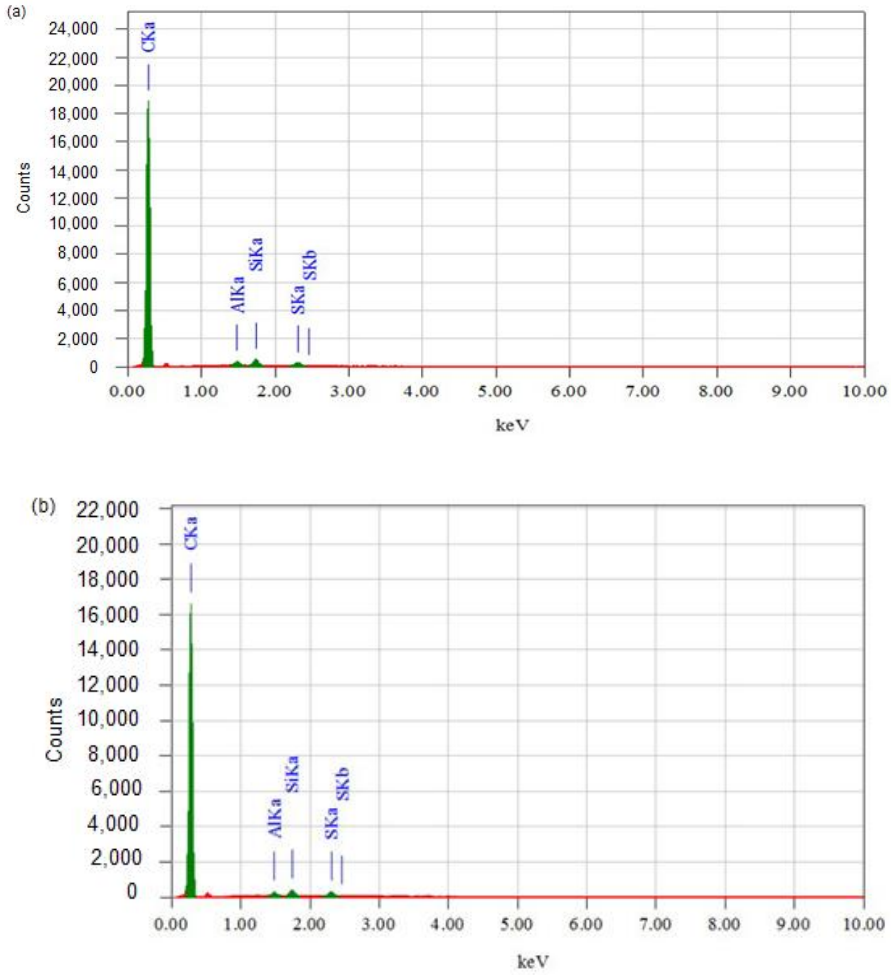

Figure 2: Results of EDS spectra of carbon electrode before electrolysis (A) and after electrolysis (B) of methylene blue 20 ppm (1 Liter) in 30 g NaCl at a current of 1.5 A

Table 1: Components of element on C-PVC electrode before and after used electrolysis

Element	Energy (keV)	Mass (%)		Atom (%)	
		Before	After	Before	After
C	0.277	98.57	98.71	99.40	99.46
Al	1.486	0.38	0.32	0.17	0.14
Si	1.739	0.65	0.54	0.28	0.23
S	2.307	0.39	0.43	0.15	0.16
Total	-	100	100	100	100

3.2 The effect of aeration to electrochemical degradation of Methylene Blue

Aeration technique is one of the liquid waste processes by adding oxygen to the electrolysis. This technique is often performed on large industries with high levels of liquid waste. The addition of oxygen is one of the efforts of pollutant removal, so that the concentration of pollutants will be reduced or even can be eliminated altogether. Oxygen will be oxidizing methylene blue at surface of electrode. The decomposing of methylene blue that breaks down into simple materials such as CO_2, CO and H_2O. Aeration system in this research using water pump so to get this tool is quite easy. Figure 3 shown the effect of aeration to electrochemical degradation of 20 ppm methylene blue 1.0 L without aeration (a) and aeration by water pump (b) using current of 1.5 A in 30 g NaCl. Based on Figure 3 it could be seen that the aeration to the electrolysis process so that it affected the degradation of methylene blue. Based on Figure 3a it could be seen that when used electrolysis aeration system that happened too long cause the formation of new compound by appearance of peak (increase of absorbance value at certain wavelength). The absorbance of the 300 nm wavelength area is high

and the longer the electrolysis process increases. Then the researchers tried to analyze with electrolysis time in the presence of a shorter aeration system presented in Figure 3b, it could be seen that the lower electrolysis time of the better analysis of methylene blue waste which the optimum conditions on electrolysis for 10 min. Degradation process in methylene blue with electrolysis process at current of 1.5 A and NaCl 30 g optimum time was using 60 min has been done, while in this research electrolysis process was added aeration system of electrolysis process which it was done shorter to degrades the methylene blue dye (Riyanto and Mawazi, 2015).

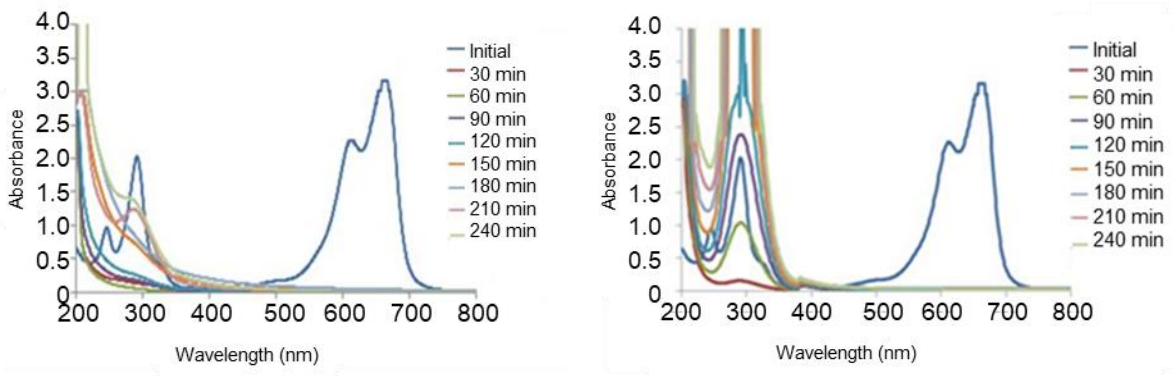

Figure 3: UV-Vis spectra resulted from electrolysis of 20 ppm methylene blue 1.0 L without aeration (a) and aeration by water pump (b) using current of 1.5 A in 30 g NaCl

3.3 The Effect of Hydrogen Peroxide to Electrolysis Degradation of Methylene Blue

Figure 4 shows the effect of H_2O_2 volume 1 mL (a) 2 mL (b) and 3 mL (c) to the electrolysis of 20 ppm methylene blue 1.0 L using current constant of 1.5 A in 30 g NaCl.

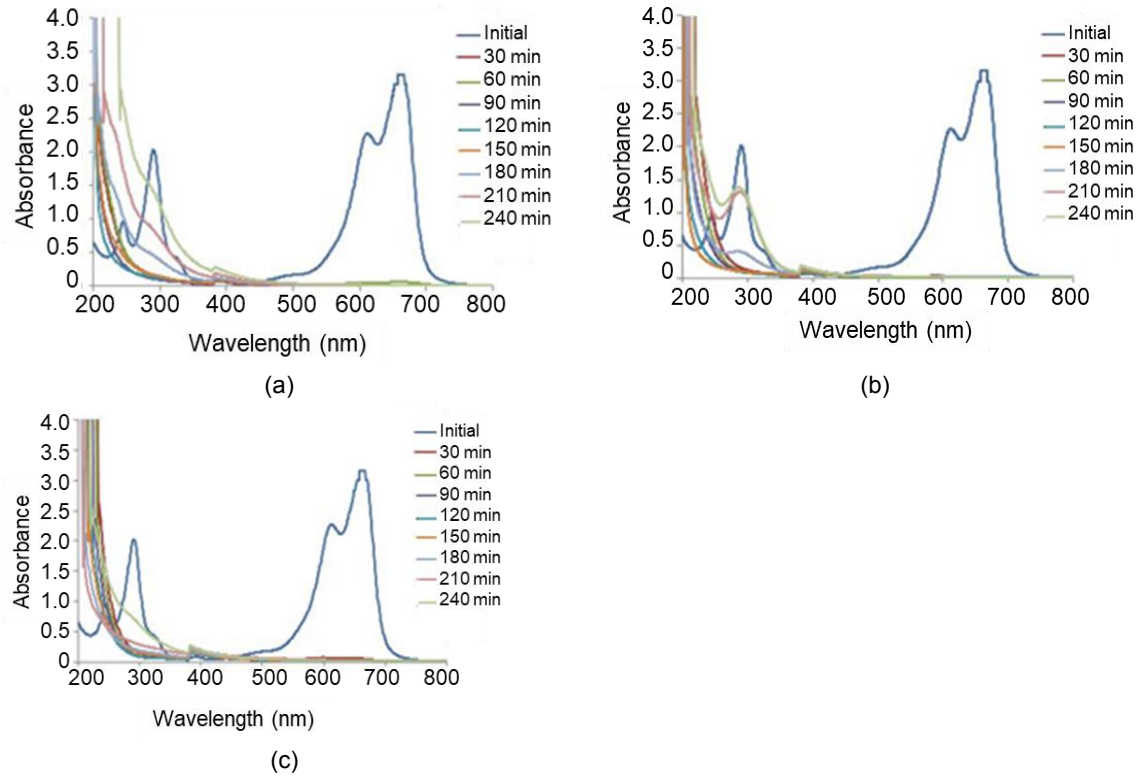

Figure 4: UV-Vis spectra resulted from electrolysis of 20 ppm methylene blue 1.0 L with H_2O_2 volume (a) 1 mL (b) 2 mL and (c) 3 mL

Hydrogen peroxide is a very good solubility compound in water, and stable with very low decomposition rate. At the time of decomposition of this compound decomposes into water and oxygen gas. The reaction that occurs following an exothermic reaction is:

$2H_2O_2 \rightarrow O_2 + 2H_2O$ + heat

The advantage of hydrogen peroxide compared to another oxidation is it's environmentally friendly. This compound leaves no residue, only water and oxygen. Its oxidizing power can be adjusted as needed.
H_2O_2 will be oxidizing of methylene blue solution to colorless this is evidenced by the wavelength range 400 - 800 nm is no longer formed peak. This result show that the use of H_2O_2 causes the time required for faster electrolysis. This study optimum conditions of the electrolysis process when the addition of H_2O_2 as much as 1 mL with electrolysis time of 30 min.

4. Conclusions

Carbon electrodes are very effective used for electrolysis of methylene blue degradation in NaCl solution. The electrolysis process with the addition of the aeration and hydrogen peroxide is good effects to degradation of methylene blue dye with electrolysis time of 10 min. As a conclusion, this method is simple, easy operational and friendly for environmental.

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References

Canizares P, Martinez F, Diaz M, Garcia-Gomez J, Rodrigo MA, 2002, Electrochemical Oxidation of Aqueous Phenol Wastes Using Active and Nonactive Electrodes. Journal of Electrochemical Society, 149, 118-26.

Gosti M, Kalogerakis N, Psillakis E, Samaras P, Mantzavinos D, 2005, Electrochemical Oxidation of Olive Oil Mill Wastewaters. Water Research Journal, 39, 4177-4182.

Guswandi J, Panjaitan S, Suhardi W, Niloperbowo T, Setiadi, 2007, The Removal of Waste Textile Color with Marasmius Sp in Modified Packed Bed. PhD thesis, University of Indonesia, Bandung, Indonesia.

Ida A, Diantarani NP, Yuliana, 2001, Photodegradation of Methylene Blue using UV light and Al_2O_3 catalyst. Jurnal Kimia, 5, 31-42.

Nordin N, Amir SFM, Yusop MR, 2015, Decolorization of C.I Reactive Orange 4 and Textile Effluants by Electrochemical Oxidation Technique Using Silver-Carbon Composite Electrode. Acta Chimica Slovenica, 62, 642-51.

Panizza AM, Martinez H, 2013, Role of Electrode Materials for the Anodic Oxidation of Real Landfill Leachate-Comparison Between Ti-Ru-Sn Ternary Oxide, PbO_2 and Boron-Doped Diamond Anode. Chemosphere, 90, 1455-63.

Perdana G, 2007, Industrial Environmental and Waste Management System. Bandung Press, Bandung, Indonesia.

Riyanto R, Mawazi M, 2015, Electrochemical Degradation of Methylene Blue using Carbon Composite Electrode (C-PVC) in Sodium Chloride. Journal of Applied Chemistry, 8, 31-40.

Sugiharto, 2005, Basics of Wastewater Treatment. University of Indonesia, Jakarta, Indonesia (In Indonesian).

Szpykowicz L, Kaul SN, Neti RN, Satyanaran S, 2005, Influence of Anode Material on Electrochemical Oxidation for Treatment of Tannery Wastewater. Water Research, 39, 1601-1610.

Wijaya K, Sugiharto E, Wijaya K, Tahir I, Kamalia, 2006, Titan Dioxide Dispersions at Natural Zeolite (TiO_2/Zeolite) and Its Application for Congo Red Photo degradation. Indonesia Journal of Chemistry, 6, 38-42.