Role of The Concentration of Fe/C Catalysts on Heterogeneous Fenton Degradation Remazol Yellow FG

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Abstract. Remazol Yellow FG is a thiazine dye widely used in textile industries. This compound is difficult to degrade naturally. One method that can be used for wastewater treatment is the Advanced Oxidation Process with heterogeneous Fenton reaction. The heterogeneous Fenton reaction mechanism uses \( \text{H}_2\text{O}_2 \) as an oxidizer and iron nanoparticles as a catalyst. One material that can be used as catalyst support for iron nanoparticles is activated carbon. Activated carbon can be modified as a catalyst support because it has a large surface area. Iron oxide (\( \text{Fe}_2\text{O}_3 \)) is embedded in activated carbon through the process of impregnation and calcination at a temperature of 300°C. \( \text{Fe}_2\text{O}_3 \) loading varies 2%, 4%, and 6% of the total carbon mass. Fe/C catalysts were characterized by SEM and BET-BJH. The catalytic degradation reaction of Remazol Yellow FG was carried out by dissolving 200ml of Remazol Yellow FG at a concentration of 20ppm and adding 5ml of \( \text{H}_2\text{O}_2 \). The degradation results using 96 hours Fe/C catalyst for variations of concentration were analyzed using a UV-VIS spectrophotometer. Then the measurement of degradation for \( \text{Fe}_2\text{O}_3 \) concentration of 2%, 4%, 6% using the heterogeneous Fenton method resulted in the percent removal of 16.82%, 40.46%, 38.32%. Whereas using physical adsorption on each variable \( \text{Fe}_2\text{O}_3 \) concentration did not result in percent removal. Data shows that the degradation capacity of Remazol Yellow FG using heterogeneous Fenton reaction increases with increasing \( \text{Fe}_2\text{O}_3 \) concentration. This also proves that the heterogeneous Fenton method using the Fe/C catalyst is effective for the degradation of Remazol Yellow FG.

Keywords: Catalytic reaction; Heterogeneous Fenton; Iron Oxide Nanoparticles; Modified Activated Carbon; Remazol Yellow FG

1. Introduction

Dyes are the main component used in the textile industry processes. So far, the textile industry is the main user of dyes. However, the textile dye then disappeared into waste and was found to accumulate in industrial waste streams [1]. Textile industrial waste contains dyes that are difficult to degrade naturally and cause disruption of aquatic ecosystems [2]. The dyes produced from the textile industry are generally non-biodegradable compounds that cause environmental pollution, especially the aquatic environment [3]. The dyes used in the textile industry are generally made of azo compounds and their derivatives which are benzene groups. The benzene group is very difficult to degrade because it is a stable compound. Remazol Yellow FG is a thiazine dye widely used in textile industries. This compound...
is difficult to degrade naturally because of its low biodegradability, so it can damage the balance of the environmental ecosystem. Adsorption methods, for example using porous carbon as adsorbent, is one popular technique in dye removal [4-8]. However, the adsorption method is only transferring pollutant compounds to other media or phases. Thus, an additional process is needed to degrade the dye by including the active substance in the adsorbent for degradation. In this context, the application of the Fenton oxidation is heterogeneous appears as a promising alternative because it allows to completely degrade organic pollutants [9,10]. This process is based on the breakdown of H$_2$O$_2$ by a solid catalyst leading to formation of hydroxyl radicals, highly reactive species attacking mostly non-selective organic pollutants. Iron represents the main active phase for the heterogeneous Fenton system [11]. Regarding catalytic supports, alumina [12], pillared clay [13] or silica [14] have been evaluated but carbon materials are usually considered the most promising solids because of them outstanding properties such as good physical and chemical stability and high specific surface area (500-2000 m$^2$/g) [15–17].

Therefore, in this research, it can be seen that the degradation capacity of Remazol Yellow FG using Fe / C catalyst at various concentrations. The degradation process uses the Fenton Heterogeneous method with the addition of H$_2$O$_2$ as a producer of hydroxyl ions.

2. Material and Method

2.1. Experiment Material
The materials used in this study were activated carbon (Bratachem). The precursors for the iron catalyst used were Fe(NO$_3$)$_3$. 9H$_2$O (99%, Merck), and 0.09 M isopropyl alcohol (analytical grade, Bratacem) as solvents. Synthetic dye FG Remazol Yellow was used to simulate waste in the Fenton reaction. Then H$_2$O$_2$ (50%, Merck) was used as a hydroxyl radical producer.

2.2. Fe/C Catalyst Preparation
Fe(NO$_3$)$_3$.9H$_2$O with a concentration of 2%, 4%, and 6% were dissolved in 10 ml of 0.09 M isopropyl alcohol. Then the Fe(NO$_3$)$_3$.9H$_2$O solution was added to 5 grams of dried activated carbon. After that, the stirring was carried out for 1 hour using an ultrasonicator and dried naturally at room temperature for 24 hours. The next process is the mixture is calcined in nitrogen flow at a temperature of 300 ℃ for 3 hours in order to remove NO$_2$. The resulting iron oxide-loaded porous carbons were denoted as X wt% Fe/C.

2.3. Degradation Process of Remazol Yellow FG
A total of 5 ml H$_2$O$_2$ was added to 200 ml of Remazol Yellow FG solution with a concentration of 20 ppm then stirred until homogeneous. After that the solution was added with Fe / C catalyst with a concentration of wt % are 2%, 4%, and 6%. Furthermore, 5 ml of solution was taken at intervals of 24, 48, 72, and 96 hours. Then the sample solution was analyzed using a UV spectrophotometer at a wavelength of 414 nm.

3. Results and Discussion

3.1. Characterization of Fe/C catalyst
The degradation process of Remazol Yellow FG is influenced by several factors, one of which is the distribution of iron oxide on the carbon surface. Even distribution on the carbon surface can maximize the degradation process. Scanning Electron Microscope (SEM) is a tool that can be used to determine the distribution of iron oxide on the carbon surface. SEM analysis was performed on carbon for each variable Fe 2%, 4%, and 6%. The results of the analysis of the distribution of iron oxide on the carbon surface are shown in Figure 1. Based on Figure 1, the area with pores is activated carbon, while the white clumps that stick are iron oxide particles.
In Figure 1 it can be seen that the iron oxide on the carbon surface has been evenly distributed. The even distribution of iron oxide can increase the degradation capacity due to the wider contact area for the pollutants to the catalyst.

![Figure 1](image1.jpg)

(a) 
(b) 
(c) 

Figure 1. The SEM image and elemental mapping analysis results for (a) 2% Fe/C, (b) 4% Fe/C, and (c) 6%Fe/C

3.2. Catalytic Performance

One way to degrade dye waste is the heterogeneous Fenton method. Oxidation with Fenton's reagent uses hydrogen peroxide (H$_2$O$_2$) as an oxidizer and iron nanoparticles as a catalyst. In this study, a heterogeneous catalyst made by impregnation of Fe into carbon pores is used so that it can facilitate the catalyst separation process. In research conducted by [18], stated that in its function as a catalyst carrier, carbon itself has affinity for dyes. Therefore, actually the physical adsorption process and Fenton's degradation reaction occur simultaneously in the heterogeneous catalyst system.

The study was conducted using 200 ml of Remazol Yellow FG solution with a concentration of 20 ppm added with 25 mg of Fe / C catalyst and 5ml H$_2$O$_2$. In this process, a reaction occurs between Fe$^{3+}$ and H$_2$O$_2$ ions which produce hydroxyl radicals (·OH). The hydroxyl radical (·OH) produced from hydrogen peroxide has very strong oxidative properties so that it can decompose Remazol Yellow FG. The degradation of Remazol Yellow FG by Fe / C catalyst with variable concentrations of iron oxide (Fe$_2$O$_3$) has been analyzed by UV-VIS spectrophotometer and can be seen in Figure 2.
Figure 2. Concentration profile of Remazol Yellow FG at different contents of iron oxide on carbon support.

From Figure 2 it can be seen that the Fe/C catalyst with a concentration of 4% is the catalyst with the highest degradation rate with a removal percentage of 40.47% from the initial level of 20 ppm. This is because the concentration of iron oxide used affects the degradation capacity. The greater the iron oxide concentration, the more the degradation capacity increases. However, at large concentrations of iron oxide, the possibility of agglomeration is higher. This actually inhibits the Fenton reaction so that the degradation capacity will decrease.

Figure 3. Percentage removal of Remazol Yellow FG with and without H$_2$O$_2$

Meanwhile, to determine the difference in degradation capacity between heterogeneous Fenton method and physical adsorption, degradation was carried out using Fe / C at a concentration of 2% and 4% with the addition of H$_2$O$_2$ for heterogeneous Fenton and without the addition of H$_2$O$_2$ for physical adsorption. In Figure 3(a) it is shown that the 2% concentration using the heterogeneous Fenton method resulted in a removal percentage of 16.82% while the physical adsorption method did not produce a percentage of removal. The 4% concentration in Figure 3(b) using the heterogeneous Fenton method
resulted in a removal percentage of 40.46% while the physical adsorption method did not produce a removal percentage, which means that no Remazol Yellow FG has been degraded. This shows that the addition of H$_2$O$_2$ to the heterogeneous Fenton method affects the degradation process of Remazol Yellow FG dye. H$_2$O$_2$ functions as an oxidizer that generates hydroxyl radicals (OH-) so that it can accelerate the degradation process. As shown in Figure 3, the degradation yield by the fenton method is greater than using physical adsorption. This is because Remazol Yellow FG has azo bonds so it is difficult to degrade naturally.

4. Conclusion

Based on the research results discussed above, it can be concluded that the difference in the concentration of iron oxide has an effect on the degradation capacity. The highest degradation capacity was achieved at 4% iron oxide concentration with a contact time of 96 hours. In addition, the degradation process can only occur with the addition of H$_2$O$_2$ as a producer of hydroxyl ions.

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