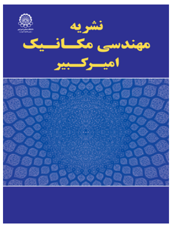

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Investigation of Effective Parameters on Dye Removal from Petrochemical Industry Effluents by Fenton and Electro-Fenton Methods

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ABSTRACT: Petrochemical industry effluents are one of the most hazardous effluents available, which contain toxic and biodegradable compounds, and their release into nature carries serious environmental risks. In this way, it is necessary to purify them. In this research, the Fenton method was used as one of the efficient advanced oxidation methods for Abadan petrochemical wastewater treatment. The effect of three parameters of pH, H_2O_2 concentration and Fe/H_2O_2 ratio, and their interference on the dye removal efficiency of this effluent was performed using the design method of the Box-Bonken statistical test with the help of Design Expert software. In order to achieve the maximum amount of dye removal as an indicator of pollution, the optimal conditions were Selected by the software as $\mathrm{pH}=3.06$, the amount of oxidizing $H_2O_2=8.88(\mathrm{ml})$, and the molar ratio $Fe/H_2O_2=0.1$. In these conditions, the highest rate of dye removal was predicted to be 79.5%, which in real conditions showed a yield of 72.5%, which is acceptable due to laboratory error ($|R^2-R^2_{adj}|<0.004$). The electro-Fenton method was also used to increase the removal efficiency. In the electro-Fenton method, under optimal conditions of Fenton process ($\mathrm{pH}=3.06$ and $H_2O_2=8.88(\mathrm{ml})$) and current density of , the maximum dye removal efficiency of 100% was obtained.

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1- Introduction

With the growth of the petrochemical industry, the release of large amounts of petrochemical wastewater contaminated with toxic, hazardous, and biodegradable substances, as well as heavy metals into the environment, take place, which causes many environmental problems and diseases, while their treatment can be Preventing multiple hazards paved the way for the reuse of these effluents for a variety of uses, including agricultural and industrial uses in drought and water crisis conditions [1, 2]. Physical and chemical processes are commonly used to remove dye from effluents [3]. Advanced oxidation processes such as photocatalysts, ozonation, ultrasonics, and Fenton are among the chemical methods that have been successful as the main pretreatment method in removing dyes from effluents [1-3]. Fenton process is one of the advanced oxidation processes that due to its ease of implementation, the possibility of using it on different scales without any need to use energy, and being cheaper, is a well-known method that is able to completely decompose organic pollutants in a short time, and convert them into harmless or harmless compounds such as carbon dioxide, water, and mineral salts. In the Fenton method, the reaction of iron (II) with H_2O_2 in aqueous solution leads to the formation of OH° and OH_2° radicals as active mediators which Hydroxyl radicals play an effective role in the oxidation of pollutants [4].

2- Methodology

H_2O_2 with a density of $1.13(\mathrm{gr/ml})$ and purity of 35% (Dr. Mojallali Co.), seven-aqueous iron sulfate (merk Co.), sulfuric acid, and one molar of Sodium hydroxide (Merck Co.) to adjust the pH and sodium chloride salt (Merck Co.) to increase conductivity Electric are used in the electro-Fenton method in the experiment. The actual effluent was received from a petrochemical plant in southern Iran. The effluent was well homogenized after transfer to the laboratory and then packaged in plastic bottles and stored in the refrigerator at 4°C. The amount of Chemical Oxygen Demand (COD), total dissolved solids, and initial pH of the effluent were measured at $15000(\mathrm{mg/l})$, $21(\mathrm{mg/l})$ and 9, respectively. The operating volume of the reactor in the Fenton process is $25(\mathrm{ml})$. The reaction time was considered to be one hour. After adding iron as a catalyst to the effluent, H_2O_2 was added to the effluent in 4 steps with an interval of fifteen minutes. Due to the foaming of the solution, the stirrer speed was adjusted between $100(\mathrm{r/min})$ and $200(\mathrm{r/min})$ after the reaction and separation of the produced sludge, and the effluent was collected.

3- Results and Discussion

3- 1- Analysis

To measure the amount of dye removed, an ultraviolet/

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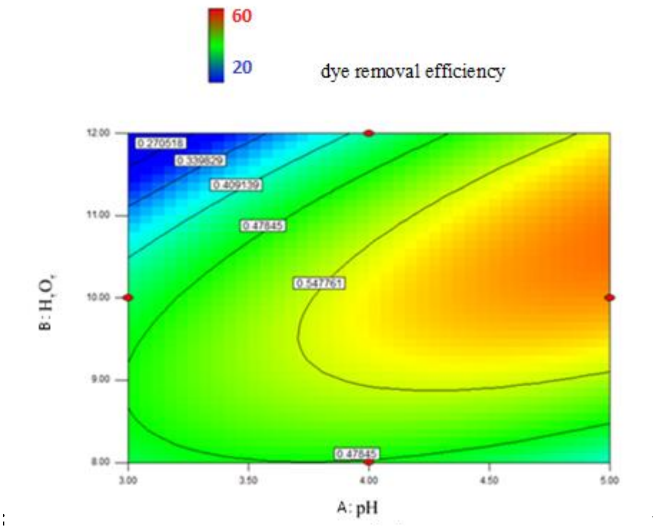

Fig. 1. Phase velocity dispersion curves for a steel pipe with outer diameter of 220 mm and wall thickness of 4.8 mm

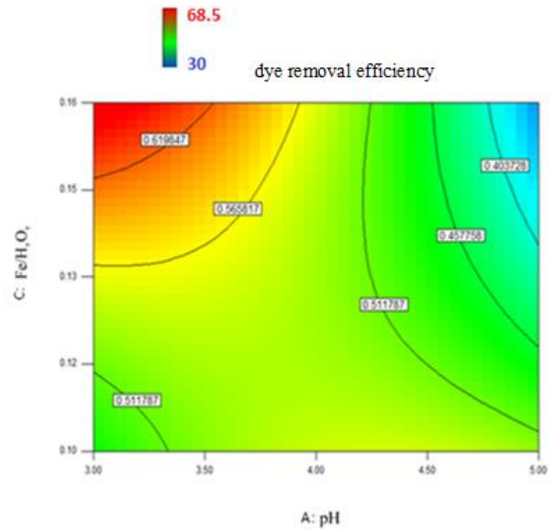

Fig. 2. Contour diagram of the interaction of pH and Fe/H2O2 concentration

visible spectrophotometer (UV-visible absorption spectrometer / double-beam UV-670 Jasco) was used. Each sample was diluted with a ratio of 1 to 10 so that the amount of light absorption in the range could be measured and then the amount of surface area below the light absorption diagram was measured in terms of wavelength in the range of $700(\mathrm{nm})$-$400(\mathrm{nm})$. The values are R^2=0.9970, R^2_{adj}=0.9930 and R^2_{Pred}=0.9654, which indicate that the predicted amount and the actual amount are correct, and this amount of error is acceptable with respect to laboratory errors.

3- 2- Interaction of operating parameters pH, H_2O_2, Fe/H_2O_2

In the low range of pH, the percentage of dye removal at low concentrations is higher than at high concentrations. In Fig. 1, at different concentrations of H_2O_2, with an increase of pH, the percentage of dye removal increased, which confirms the non-interference of these two parameters. The reason for the increase in dye removal efficiency is that at lower pH, due to the formation of $Fe(H_2O)^{2+}$, there is less reduction in dye. $Fe(H_2O)^{2+}$ reacts with H_2O_2 to reduce hydroxyl radicals [5].

At low ratios of Fe/H_2O_2, the percentage of dye removal in the low range of pH is higher than at high ratios but at high ratios of Fe/H_2O_2, the percentage of dye removal in the high range of pH is less than low range. As shown in Fig. 2, the slope of the dye removal diagram at different Fe/H_2O_2 ratios is not similar to the increase of pH, which causes interference between these two parameters. At low pH, the percentage of dye removal increases. With the increase of Fe^{2+} due to the tendency of hydroxyl radicals to perform the oxidation-reduction reaction with Fe^{2+} and

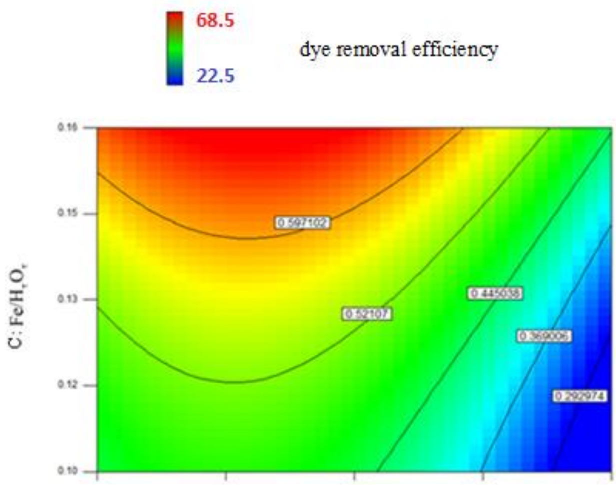

Fig. 3. Contour diagram of the interaction of and concentration

H_2O_2, the removal efficiency increases and the production of hydroxyl radicals reaches its highest value. At high pH, the removal percentage decreases. In this pH range, H_2O_2 decomposition is limited by the disappearance of H^+, and less OH is produced [2].

At low ratios of Fe/H_2O_2, the percentage of dye removal in a low range of H_2O_2 is higher than at high ratios, but at high ratios of Fe/H_2O_2, the percentage of dye removal at the high range H_2O_2 is higher than at the low range. In Fig. 3, at the low range of H_2O_2, the dye removal percentage increases with increasing of Fe/H_2O_2, because the excess iron reacts optimally with hydroxyl radicals to produce $Fe(OH)_3$. In the high amounts of H_2O_2 and with increasing of Fe/H_2O_2, it also acts as a scavenger of hydroxyl radicals and reduces the rate of adsorption of hydroxyl radicals, which ultimately leads to increased dye removal [5].

4- Conclusion

Examination of the effect of parameters showed that the most important parameter is H_2O_2 concentration. On the other hand, the study of the interaction of the parameters showed that the two parameters H_2O_2 and Fe/H_2O_2 as well as pH and Fe/H_2O_2 have the most interference, respectively. Based on the design results, optimal conditions ($pH=3.06$, $H_2O_2=8.88(ml)$ and $Fe/H_2O_2=0.1$) were determined. In these conditions, the highest amount of dye removal was predicted to be 79.5%, and performing the experiment in real conditions showed a yield of 72.5%, which is acceptable due to the laboratory error ($|R^2-R^2_{adj}|<0.004$).

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