

# Effective Sonodegradation of Methylene Blue (MB) Organic Dye by Mil-88(Fe)/NaY/ MnFe<sub>2</sub>O<sub>4</sub> Nanocomposite Sonocatalyst in Aqueous Solution

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#### **Abstract**

This research examined sonocatalytic degradation of Methylene Blue (MB) dye in the presence of MIL-88(Fe)/NaY/MnFe $_2$ O $_4$  nanocomposite synthesized using the ultrasound assisted-hydrothermal route. Multiple identification techniques were utilized to investigate the MIL-88(Fe)/NaY/MnFe $_2$ O $_4$  nanocomposite sonocatalyst involving FESEM, EDAX, FTIR, XRD and BET. The influences of various parameters like contact time, H $_2$ O $_2$  concentration, initial MB concentration and sonocatalyst dosage were precisely studied. About 98.1% of MB dye degradation was achieved under the optimum parameter conditions i.e. at pH of 7, 25mg/L of initial MB concentration, H $_2$ O $_2$  concentration of 4mM and 0.5g/L of MIL-88(Fe)/NaY/MnFe $_2$ O $_4$  dosage within 60min. The enhancement of sonocatalytic activities can be related to the function of NaY zeolite as trap state for the electron. The scavenger tests outcomes demonstrated that the sono-generated hydroxyl radical (.OH) would play an important role in the MB degradation. Additionally, the MIL-88(Fe)/NaY/MnFe $_2$ O $_4$  was quite stable since the efficiency of MB degradation gained in the four run was 93.7%.

**Keywords:** Sonocatalytic; Degradation; Methylene blue; MIL-88(Fe)/NaY/MnFe<sub>2</sub>O<sub>4</sub>; Nanocomposite

## **Graphical Abstract**

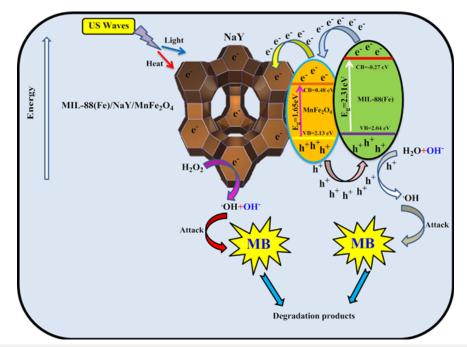


Figure 1

## Introduction

Today, organic dye sewages are a main source of water contamination [1,2]. Every year, the estimation demonstrates that up to 15% of dye sewages are evacuated into the surface and ground water supplies [3,4]. The exposure with these dye contaminants even at very low concentrations can be harmful to human and the environment [5]. For this reason, these toxic compounds must be removed from the environment. Recently, the various beneficial methodologies like adsorption, membrane filtration, electrochemical treatment, oxidation, ozonation, photocatalytic and sonocatalytic processes have been implemented for the removal of toxic organic dyes in the polluted aqueous solutions [6-11]. Among the aforementioned methods, the sonochemical process is considered as an effective route for the removal of these types of organic pollutants [12]. Plus, applying the ultrasound irradiation causes the production of the bubbles inside the reaction solution leading to the formation of high pressure and temperature approximately 1000atm and 5000K, respectively and subsequently the production of hot spots [13,14]. Eventually, the produced hot spots can in turn leading to the hydroxyl radical (OH) and superoxide anion radical (0, ) originated from the breakdown of H<sub>2</sub>O and O<sub>2</sub> molecules, respectively. Furthermore, it has been specified that the use of semiconductors catalysts along with the US/H2O2 system provides higher degradation performance in comparison to the H<sub>2</sub>O<sub>2</sub> only and US only. Additionally, different types of catalysts, involving Cr-MIL-101@NiO/13X, InVO<sub>4</sub>/TiO<sub>2</sub>, NiGa<sub>2</sub>O<sub>4</sub>/CeO<sub>2</sub>, CoFe<sub>2</sub>O<sub>4</sub>/CdS, La/TiO<sub>2</sub>/Y have been used for the removal of organic dye pollutants via the sonodegradation processes [15-19]. Among these types of reported catalysts, Metal-Organic Frameworks (MOFs) are considered as new classes of porous compounds that have various applications due to their different properties such as catalysis, nanofluids, chemical sensing and so on [20-22]. On the other hand, the zeolites are introduced as a group of hydrated crystalline aluminosilicate involving alkali and alkaline earth metals [23]. These materials possesses unique properties such as high hydrothermal stability and biocompatibility, large pore volume and high surface area, which is suitable for its use as catalysis, adsorbents and so on [24-26]. Regarding the aforementioned explanations, it is explicitly known that combining the spinel MnFe<sub>2</sub>O<sub>4</sub> nanoparticles with MIL-88(Fe) metal-organic framework in the NaY zeolite framework will produce a magnetically separable and superior nanocomposite catalyst that remarkably facilitate the hole and electron separation. Hence, in the presence work, the MIL-88(Fe)/NaY/MnFe<sub>2</sub>O<sub>4</sub>, nanocomposite sonocatalyst was prepared using the ultrasound assisted-hydrothermal route. The as-prepared nanocomposite was fully characterized via the FESEM, EDAX, FTIR, XRD and BET analyses. Then, the above-mentioned nanocomposite catalyst was assessed for the effective sonodegradation of MB dye from aqueous solution in the presence of the ultrasound (US)/H<sub>2</sub>O<sub>2</sub> system. Besides, the impacts of several parameters like contact time, H<sub>2</sub>O<sub>2</sub> concentration, initial MB concentration and sonocatalyst dosage on the MB degradation were studied. Nevertheless, as far as we know, no study has been done on the sonodegradation reaction

in the presence of the MIL-88(Fe)/NaY/MnFe $_2\mathrm{O}_4/\mathrm{US/H}_2\mathrm{O}_2$  system in before.

#### Conclusion

To sum up, MIL-88(Fe)/NaY/MnFe $_2$ O $_4$  was successfully synthesized by the ultrasound assisted-hydrothermal method. The results revealed that the sonocatalytic degradation of MB was obviously affected by contact time,  $\rm H_2O_2$  concentration, initial MB concentration and sonocatalyst dosage. The highest 60min at a pH of 7, 4mM of  $\rm H_2O_2$  concentration, 25mg/L of initial MB concentration and 0.5g/L of MIL-88(Fe)/NaY/MnFe $_2$ O $_4$  dosage. The sonocatalytic process applying the MIL-88(Fe)/NaY/MnFe $_2$ O $_4$ , US (ultrasound) and  $\rm H_2O_2$  displayed great potential in treatment of MB. In addition, MIL-88(Fe)/NaY/MnFe $_2$ O $_4$  could be applicable to the practical application since it showed high stability and recyclability. By this test, MIL-88(Fe)/NaY/MnFe $_2$ O $_4$  nanocomposite illustrates a promising candidate for the sonocatalytic degradation in environmental remediation.

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