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# Synthesis of Novel CuO/ZnFe<sub>2</sub>O<sub>4</sub>/NaA Zeolite Nanocomposite Adsorbent for the Decontamination of Nerve Agent Sarin Simulant

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#### **Abstract**

The aim of present research is to evaluate the decontamination of nerve agent sarin simulant dimethyl methyl phosphonate (DMMP) in water media using the  $CuO/ZnFe_2O_4/NaA$  zeolite as a novel ternary nanocomposite adsorbent. In this regard, the CuO and  $ZnFe_2O_4$  nanoparticles were successfully synthesized within the NaA zeolite applying the ultrasound-assisted hydrothermal route and characterized via XRD, FESEM, FTIR, EDAX, VSM and BET analyses. Then, the  $CuO/ZnFe_2O_4/NaA$  nanocomposite activity was investigated for the decontamination of DMMP molecule and monitored by a GC-FID and GC-MS analyses. Plus, the impacts of several parameters, including contact time, adsorbent dose and adsorbent type on the decontamination of DMMP were studied. The gained data from GC-FID analysis confirmed the maximum decontamination more than 98.4% for DMMP. The parameters including: adsorbent amount of 50 mg and contact time of 40 min were achieved as the optimized values for the decontamination reaction. In addition, the non-toxic methyl phosphoric acid (MPA) as the DMMP degradation product in the presence of  $CuO/ZnFe_2O_4/NaA$  adsorbent was characterized.

**Keywords:** CuO/ZnFe<sub>2</sub>O<sub>4</sub>/NaA, zeolite, nerve agent sarin simulant, decontamination, dimethyl methyl phosphonate (DMMP), methyl phosphoric acid (MPA).

## Introduction

Today, organic dye sewages are a main source of water contamination (Gusmão et al., 2013; Turki et al., 2015). Every year, the estimation demonstrates that up to 15% of dye sewages are evacuated into the surface and ground water supplies (Rai et al., 2005; Gupta et al., 2009). The exposure with these dye contaminants even at very low concentrations can be harmful to human and the environment (Sadettin et al., 2006). 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 (Behnajady et al., 2008; Zhang et al., 2019; Qin et al., 2014; Sboui et al., 2017; Ayyappan et al., 2018; Hassan, 2012). Among the aforementioned methods, the sonochemical process is considered as an effective route for the removal of these types of organic pollutants (Ghodbane & Oualid, 2009). 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 1000 atm

and 5000 K, respectively and subsequently the production of hot spots (Mason & Phillip, 2002; Rajoriya et al., 2017). Eventually, the produced hot spots can in turn leading to the hydroxyl radical (.OH) and superoxide anion radical (.O<sub>2</sub>-) originated from the breakdown of H2O and O2 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, InVO4/TiO2, 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 (Sadeghi et al., 2019; Min et al., 2012; Wang et al., 2020; Alwash et al., 2013; Farhadi & Firouzeh, 2016). 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(Sadeghi et al., 2019; Díaz-Ramírez et al., 2019; Sadeghi et al., 2020). On the other hand, the zeolites are introduced as a group of hydrated crystalline aluminosilicate involving

alkali and alkaline earth metals (Sadeghi et al., 2017). 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 (Dehghani et al., 2019; Sadeghi et al., 2019; Ramezani et al., 2017). 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<sub>2</sub>O<sub>4</sub>/US/H<sub>2</sub>O<sub>2</sub> system in before.

#### **Conclusion**

In this research, the ultrasound-assisted hydrothermal method was used to fabricate magnetic CuO/ZnFe<sub>2</sub>O<sub>4</sub>/NaA zeolite as a novel nanocomposite. The noteworthy activity of the CuO/ZnFe<sub>2</sub>O<sub>4</sub>/NaA nanocomposite for the decontamination of DMMP in aqueous solution was investigated and monitored via GC-FID analysis. Besides, the influences of several parameters such as contact time, adsorbent amount and adsorbent type on the decontamination efficiency of DMMP were evaluated. On the basis of the GC-FID analysis outcomes, the decontamination (98.4%) of DMMP in the presence of the CuO/ZnFe<sub>2</sub>O<sub>4</sub>/NaA under the optimized conditions of contact time 40 min and adsorbent amount of 50 mg was proved. Alternatively, the high activity of above-mentioned nanocomposite makes it a superior candidate to utilize in the decontamination applications of chemical warfare agents.

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