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MAGNETIC SEPARATOR FOR REMOVAL OF ORGANIC POLLUTANTS

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ABSTRACT

Magnetic composite of magnetite (Fe_3O_4) and graphitic carbon nitride (g-C_3N_4) was synthesized using a co-precipitation method starting from $Fe^{2+}$and Fe^{3+} salts and g-C_3N_4, which was obtained by different starting nitrogen-rich organic precursors urea and melamine by thermal polymerization technique. The composites were tested for methylene blue (MB) degradation in an aqueous solution under visible light irradiation. The physicochemical properties of the composites were characterized by XRD and FTIR. The XRD pattern shows the existence of sharp peaks, indicating that all the samples are in the crystalline phase. In addition, FTIR results revealed both bonds of C=N and C-N were at the range of 1200 cm^{-1} to 1600 $cm^{-1,}$ confirming the existence of g-C_3N_4. The magnetic composite for S4 and S5 enables 68.9% and 90.9% degradation of MB within 5 hours, respectively. This study demonstrates that the photocatalytic methylene blue under visible light is approximately two times greater when a mixture of urea and melamine is used as the g-C_3N_4 precursor than urea alone. Furthermore, the composite's high saturation magnetization suggests that the photocatalyst can be easily separated from the treated solution using a magnetic field.

Keyword: *graphitic carbon nitride, iron oxide, photocatalyst, methylene blue, magnetic, degradation*

1. INTRODUCTION

Huge quantities of pollutants, including organic dyes, antibiotics, and pesticides, are routinely discharged into various water bodies, eventually contaminating raw water. (Luo et al., 2021). Organic dye such as methylene blue (MB) is one of the threats of dye wastewater to the environment and human beings which needs to be handled carefully. It harms the ecosystem and may limit the photosynthesis of aquatic plants due to the water-dispersed RhB molecules that prevent light from penetrating aquatic habitats. Therefore, environmentally friendly technology should be created to efficiently reduce dye pollution contamination (Zhang et al., 2021). Due to its advantageous physical and chemical characteristics, such as its thermal, mechanical, and electrical properties, nontoxicity, low cost, metal-free nature, optical response in the visible light region, ease of preparation, environmental friendliness, biocompatibility, and chemical inertness, g-C_3N_4 is an efficient visiblelight-driven material photocatalyst (Luo et al., 2021). g-C_3N_4 is easily prepared by thermal polymerization of low-cost nitrogen rich precursors such as urea and melamine. Despite all the properties, there are a few shortcomings in applying g-C_3N_4 to be used in an industrial scale, such as it is difficult to recycle from the treated solutions using an external magnetic field and it creates secondary contamination (Mousavi & Habibi-Yangjeh, 2016). However, due to its high thermal and chemical stability, g-C_3N_4 can be modified by doping magnetic materials such as iron oxide (Fe_3O_4) to the g-C_3N_4, enhancing the photocatalytic performances to remove the dye pollutants (Mousavi et al., 2016). The resulting photocatalyst has a few advantages, such as being stable and easy to reuse and recycle. The effectiveness of the photocatalyst obtained was investigated using UV-Vis characterization and its photodegradation activity of methylene blue. In this study, magnetic carrier photocatalysts driven by visible light were prepared through ultrasonic and in situ deposition method denoted as g-C_3N_4 (CN)/Fe_3O_4. In addition, Fe_3O_4 was prepared via the co-precipitation method from the resulting mixture of iron (II) chloride tetrahydrate ($FeCl_2.4H_20$) and iron (III) chloride hexahydrate ($FeCl_2.6H_20$) with a neem leaf extract and ammonium hydroxide (NH_4OH) under the flow of nitrogen atmosphere and vigorous stirring (Taib et al., 2018). Thus, the first objective of this study is to prepare graphitic carbon nitride g-C_3N_4 using different precursors and g-C_3N_4/iron oxide (Fe_3O_4) composite. The microstructure, purity, morphology, and optical, textural, and magnetic properties of the resultant samples were characterized using X-ray diffraction (XRD) and Fourier transform-infrared spectroscopy (FTIR), which is the second objective. This study aims to identify the photocatalytic behavior of different precursors of the g-C_3N_4 composite under visible light radiation.

2. METHODOLOGY

2.1 Sample Preparation

Azadirachta indica aqueous leaf extract was prepared by mixing 5 g of neem leaf powder with 100 mL of distilled water in a conical flask. The mixture was then heated for 30 minutes at a constant temperature of 80 °C. The mixture was heated on a hot plate and continuously stirred with a magnetic stirrer to ensure that the mixture was homogeneous. The mixture was allowed to cool at room temperature after 30 minutes. Using Whatman No. 1 filter paper in a vial, the mixture was vacuum filtered, and the extract was then chilled at 4 °C.

Urea and melamine were used as the initial precursor materials in the thermal polymerization method to prepare graphitic carbon nitride (g-C_3N_4). 3 g of urea and 7 g of melamine powder were weighed separately, and both samples were mixed in a crucible. The sample was then heated up to 550 °C at a rate of 5 °C/min for 3 hours in a furnace inside the fume hood. Next, the sample was cooled down at room temperature. A pestle and mortar were used to grind the resulting yellowish powder. The same procedures were repeated using 10 g of urea as the precursor.

Iron Oxide Nanoparticles (Fe_30_4-NPs) were prepared by using the Co-Precipitation Method.,1.1 g of iron (III) chloride hexahydrate and 0.4 g of iron (II) chloride tetrahydrate were weighed and dissolved in 100 mL of distilled water. The solvent was transferred into the round bottom flask. Then the mixture was heated up for 10 minutes at 80 °C with nitrogen gas flowing through it. A measuring cylinder was used to measure 5 mL of aqueous neem leaf extract, which was then added to the resulting solution. By using a dropper, 20 mL of 25% ammonium hydroxide (NH_4OH) was gradually added drop by drop into the solution for 30 minutes while vigorously stirring. The colour of the mixture instantly changed to the black color solution, which is the formation of Fe_3O_4-NPs. The mixture was allowed to cool at room temperature for 30 minutes until the dark precipitate settled to the bottom of the flask. After the supernatant was discarded, the remaining black precipitate was rinsed with 15mL of distilled water. The washed precipitate was centrifuged for 5 minutes at 8500 rpm. Then the pellet was dried in the oven at 60 °C for 8 hours after removing the supernatant. Afterward, the sample was ground using a pestle and mortar and kept at room temperature.

CN/Fe_3O_4 Nanocomposite was prepared by using the ultrasonic deposition method. First, in a conical flask, 0.3 g CN of urea and 0.08g of iron oxide (Fe_3O_4) were weighed and dissolved with 40 mL of methanol and distilled water. The mixture was then constantly stirred for 3 hours at room temperature using a magnetic stirrer. The mixture underwent ultrasonication for 45 minutes to ensure the mixture is homogeneous. Then, the solution was stirred for another 1 hour and centrifuged for 5 minutes at a rate of 8500 rpm. The supernatant was decanted, and all the resulting pellets were dried using an oven for 8 hours at 60 °C. All the procedures were repeated using a different precursor—CN urea and melamine.

3. FINDINGS

All synthesized results were labeled as S1: g-C_3N_4 (urea), S2: g-C_3N_4 (urea+ melamine), S3: Fe_3O_4, S4: Fe_3O_4/g-C_3N_4 (urea), S5: Fe_3O_4/g-C_3N_4 (urea+ melamine). Based on Fourier-transform infrared spectroscopy (FTIR) characterization, two main functional groups are assigned to C-N and C=N bonds for the g-C_3N_4, g-C_3N_4/Fe_3O_4, and its precursors. Furthermore, two main functional groups are assigned to C-N and C=N bonds for g-C_3N_4. From the spectrum, the peaks of S1, S2, S4, and S5 at 805cm^{-1} corresponded to the breathing mode of C-N stretching in triazine rings, a specific characteristic peak of g-C_3N_4 (Nguyen et al., 2021). On the other hand, the broadbands around 1200 cm^{-1} to 1600 cm^{-1} are the typical stretching vibration of CN heterocyclic consisting of C-N and C=N bonds (Zhang et al., 2021). In addition, the broadbands around 3000 cm^{-1} to 3400 cm^{-1} were present due to the stretching mode of NH groups and OH stretching vibration, which may have developed from unreacted amino groups or may have been a residual hydrogen atom bonded to the margins of the graphitic nitride structure (Nguyen et al., 2021). Thus, it can be said that S1, S2, S4, and S5 contain g-C_3N_4 because all the samples consist of the main functional groups of g-C_3N_4 which are C-N and C=N bonds that appear at its theoretical range, which are near 1100 cm^{-1} for C-N and near 1660 cm^{-1} for C=N ("The Nature", n.d.). Meanwhile, for $Fe_3O_{4,}$ only the Fe-O bond exists as the main functional group. The IR band of Fe_3O_4 for Fe-O stretching should be observed around 421 cm^1 to 572 cm^{-1}, which revealed and further confirmed their phase to be Fe_3O_4 (Gupta et al., 2014). However, restriction due to apparatus limit range starting minimum of 500 cm^{-1} wavenumbers made the peaks undetectable in the FTIR spectrum.

Next, the X-Ray diffraction (XRD) results confirmed the crystalline phase of g-$C_3N_{4.}$ Both S1 and S2 sample at $2\theta = 13.1^{o}$ and $2\theta = 13.4^{o,}$ respectively ascribed to the (100) plane, indicating a small angle reflection of g-C_3N_4. According to Mitra and co-researchers, these weak peaks correlate with interlayer stacking (Mousavi et al., 2016). In the XRD pattern, the apparent strong peak appeared in all four samples, S1, S2, S4, and S5**.** The located peaks are at $2\theta = 27.7^{o}$, 27.8^{o},27.9^{o}, and 27.9^{o} for each S1, S2, S4, and S5, respectively, which corresponded to plane (002) for each sample. This angle is attributed to the stacking of the conjugate aromatic system, which indicates a bigger angle reflection of g-C_3N_4. The weak and strong peaks obtained from the results are similar to the values from previous research, $2\theta = 13.0^{o}$ for the weak peak and $2\theta = 27.4^{o}$ for the strong peak (Bao & Chen, 2016). In addition, the XRD pattern confirmed the crystalline phase of Fe_3O_4. Four distinctive peaks from the XRD pattern corresponded to the result for each sample containing Fe_3O_4. Three samples showed the appearance of Fe_3O_4. Four distinctive peaks are observed in S3, S4, and S5 at $2\theta = 35^{o}$- 36^{o}, 43^{o}- 44^{o}, 57^{o} - $56^{o,}$ and 63^{o}- 64^{o}. The samples show a similar degree value of the peaks, and all the four series peaks corresponded to planes (311), (400), (511), and (440), respectively. These results were proven based on previous studies (Taib et al., 2018) that showed the crystalline phase of Fe_3O_4 from the peaks $2\theta = 36^{o}$, 43^{o}, $57^{o,}$ and 63^{o}.

The UV-Vis characterization method evaluated the photodegradation activity of S4 and S5 composites. No photodegradation of MB was observed when the experiments were conducted under dark conditions. However, under visible light radiation, the magnetic composite S4 and S5 enabled 68.9% and 90.9% degradation of MB within 6 hours, respectively. This reaction successfully followed the pseudo-first-order reaction and confirmed that no absorption process occurred. The degradation reaction rate constant over the S4 and S5 composites is 4.0×10^{-3} min^{-1} and 8.3×10^{-3} min^{1}, respectively. This study demonstrates that the photocatalytic MB is approximately two times greater when urea is used as the g-C_3N_4 precursor than a mixture of urea and melamine.

4. CONCLUSION

The thermal decomposition approach for producing graphitic carbon nitride, g-C_3N_4, from two readily accessible precursors, urea and melamine, was conducted successfully. Highly enhanced visible-light-driven photocatalysts, g-C_3N_4/Fe_3O_4 nanocomposite obtained was the improvised version of g-C_3N_4 composite photocatalyst, which gives higher photocatalytic performance and is more ecofriendly. The obtained photocatalyst sample has a lot of potential as a versatile material with myriad applications, such as the photodegradation of organic pollutants (Mittal & Dutta, 2021). The photocatalytic activity of the composites was evaluated by degradation of dye pollutants methylene blue, MB under visible-light radiation. In addition, FTIR should be conducted at a minimum wavenumber 400 cm^{-1} so that the existence of Fe_3O_4 can be investigated at the range of 421 cm^{-1} to 572 $cm^{-1,}$ and due to the limitation of apparatus limit range, the existence of Fe_3O_4 cannot be confirmed. Thus, for future studies, we suggest analyzing the sample at UiTM Shah Alam, which has a more suitable apparatus to analyze Fe_3O_4.

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