Preparation And Study Of Photocatalytic Activity Of Graphitic Carbon Nitride For The Removal Of Pharmaceutical Pollutants

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Abstract: Excessive consumption of drugs, as well as wastes from the pharmaceutical industry and hospitals, has increased the concentration of these pollutants in the aquatic environment and even drinking water. These chemicals pose a major danger to human and environmental health due to their degradable nature, even in small quantities. Therefore, the effort to remove them from the water sources is essential. In this regard, graphitic carbon nitride via a cost-efficient method was prepared using thermal polymerization of urea. Its photocatalytic activity was studied for the removal of pharmaceutical pollutants ibuprofen and ofloxacin. The results of the experiments show that the produced graphitic carbon nitride can remove 73% and 33% of ofloxacin and ibuprofen from the aqueous medium, respectively.

Keywords: Graphitic carbon nitride, Photocatalyst, Pollutant, Pharmaceutical, Ofloxacin, Ibuprofen

REFERENCES

[1] A. Tarfiei, H. Eslami, A. A. Ebrahimi, ***Journal of Environmental Health and Sustainable Development***, **vol.** 3, no. 2, **pp.** 492–495, **2018**.
[2] V. K. Gupta, I. Ali, Environmental Water: Advances in Treatment, Remediation and Recycling, Elsevier, 2013.
[3] D. B. Hernández-Uresti, A. Vázquez, D. Sanchez-Martinez, S. Obregón, ***Journal of Photochemistry and Photobiology A: Chemistry,*** **vol**. 324, **pp**. 47–52, **2016**.
[4] B. Zhu, P. Xia, W. Ho, J. Yu, ***Applied Surface Science***, **vol**. 344, **pp**. 188–195, **2015**.
[5] S. Rostami, A. Nakhaei Pour, M. Izadyar**,** ***Journal of Nanostructures***, **vol**. 9, no. 3, **pp**. 498–509, **2019**.

1. Introduction

Nowadays, drugs play an important role in human life and are used to treat various diseases. In this regard, the presence of pharmaceutical pollutants in the environment is one of the most important worldwide issues. The main sources of these pollutants include effluents of pharmaceutical factories, hospital wastes, and disposal by drug users. Pharmaceutical pollutants with a long half-life remain in the environment due to their stability in the wastewater treatment processes. Recently, large amounts of pharmaceutical pollutants have been observed in surface waters, groundwater, and especially in drinking water [1].

Due to the serious problems caused by high concentrations of toxic pollutants for humans, there is an urgent need to develop cost-effective and environmentally friendly processes to remove them from drinking water and protect the health of humans. So far, various methods have been used to control water pollution, among them, advanced oxidation processes have been identified as an attractive option for water treatment, especially in cases where it is difficult to remove pollutants using conventional biological or physicochemical methods [2].

Heterogeneous photocatalysts have been used as an appropriate technology to solve a wide range of environmental and energy problems. In this regard, in recent years graphitic carbon nitride due to significant photocatalytic activity under visible light, metal-free structure, high chemical stability and also easy preparation through polymerization of nitrogen-rich organic compounds such as melamine, cyanamide, and urea have received much attention [3].

The aim of this study was to prepare the graphitic carbon nitride via an easy and cost-effective method and also to study its photocatalytic activity to remove the pharmaceutical pollutants ibuprofen and ofloxacin.

2. Experimental section

2.1. Synthesis of g-C_3N_4 photocatalyst

Graphitic carbon nitride was prepared using thermal polymerization of urea. In detail, 10 g urea was placed firstly in a crucible with a cover, and subsequently transferred into a furnace. Finally, the crucible was heated to 550°C with a heating rate of 4.6°C/min and then kept at this temperature for 1 h.

2.2. Photocatalystic activity test

In order to perform photocatalytic removal experiments, 50 mL of pharmaceutical pollutants ibuprofen and ofloxacin with an initial concentration of 5 ppm was poured into the photocatalytic reactor. Then 0.05 g of graphitic carbon nitride was added. To achieve adsorptiondesorption equilibrium, the resulting suspension was stirred in the dark for 60 min. Then, it was exposed to light (xenon lamp) for 120 min to perform the photocatalytic removal process. During light irradiation and at 30 min intervals, 5 mL samples were taken from the suspension and using a centrifuge, the photocatalyst in the sample was removed from the aqueous medium. It should be noted that a circulator was used to keep the temperature constant during the experiment. A schematic of the system used in these experiments can be seen in Figure 1.

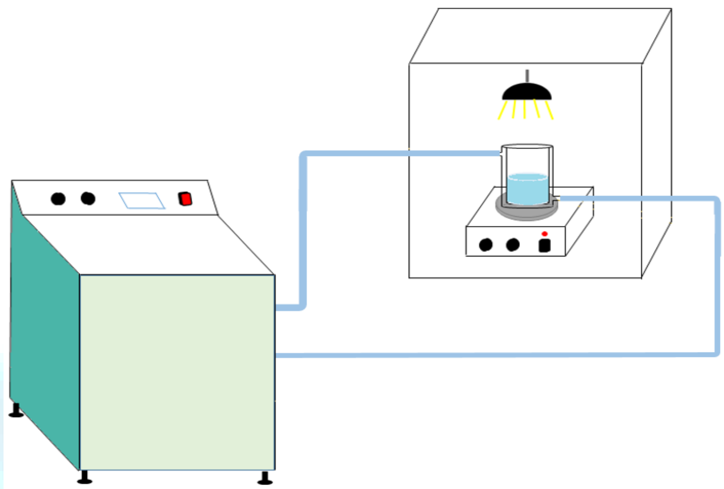

Figure 1: Schematic of the system used to perform photocatalytic removal experiments

3. RESULTS AND DISCUSSION

3.1. Characterization of g-C_3N_4

Figure 2 shows the XRD pattern for the graphitic carbon nitride. The X-ray diffraction pattern for graphitic carbon nitride has two distinct diffraction peaks. The strong peak at 2θ = 27.5°, which is the characteristic peak of aromatic systems, is related to the crystal plane (002). The weak peak at 2θ = 13° is related to the crystal plane (100) and is attributed to the in-plane structure of the heptazine repeating units. The sharpness and high intensity of the peak at 2θ = 27.5° indicates that the resulting graphitic carbon nitride has a high crystallinity [4].

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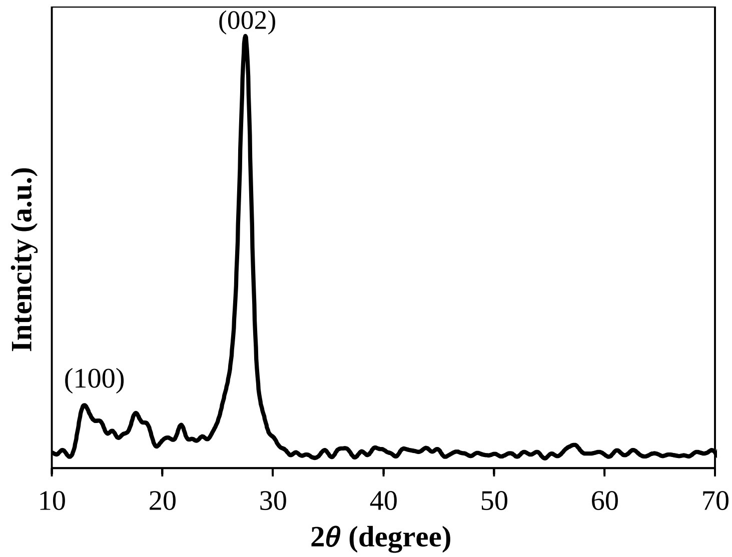

Figure 2: The X-ray diffraction pattern for graphitic carbon nitride

FTIR spectrum of graphitic carbon nitride (Figure 3) has a sharp peak at 815 cm^{-1}, which is related to the bending vibrations of heptazine units. The peak at 891 cm^{-1} is related to N-H deformation and shows the partial condensation of amine groups. In addition, several strong peaks are seen in the 1250-1650 cm^{-1}, which belong to the stretching modes of CN heterocycles. The wide peak in the region of 3000-3300 cm^{-1} is related to N-H stretching vibrations, which indicates that the structure of the resulting graphitic carbon nitride has uncondensed amine groups [5].

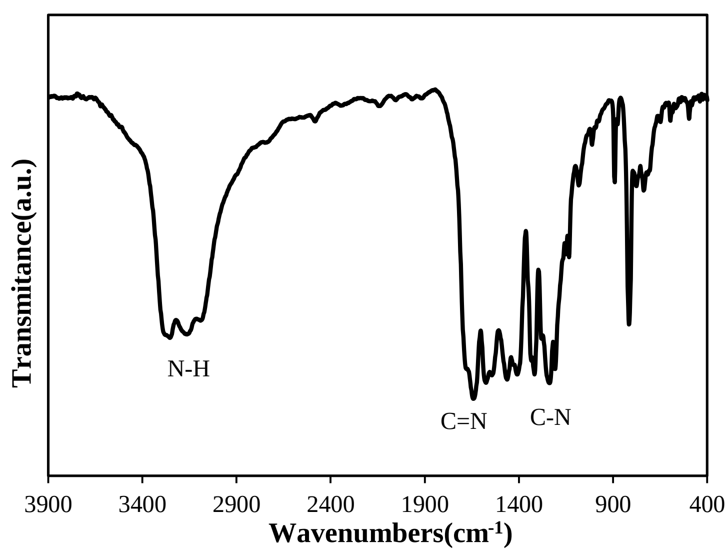

Figure 3: FTIR spectra of the prepared graphitic carbon nitride

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The FESEM image of graphitic carbon nitride is shown in Figure 4. As can be seen, this material has a flake-like structure that consists of the accumulation of nanosheets with a thickness of 40 nm. Also, the mesoporous structure (probably due to the release of gas during the thermal polymerization process) is well seen in the prepared graphitic carbon nitride.

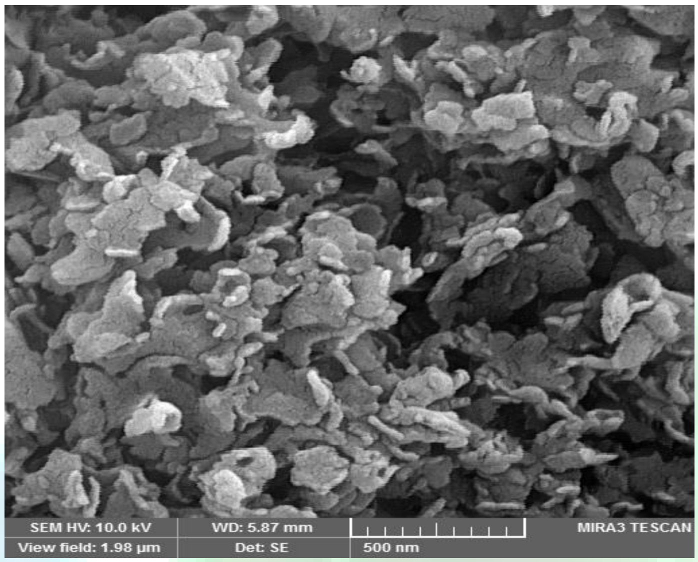

Figure 4: FESEM image of graphitic carbon nitride

The results of photocatalytic removal experiments of pharmaceutical pollutants using graphitic carbon nitride as photocatalyst are shown in Figure 5. As seenin the figure, according to the removal amount of ibuprofen in the first 60 min (dark conditions), the adsorption of this pollutant by graphitic carbon nitride is negligible. For ofloxacin in dark conditions, 10% reduction in the concentration was achieved, due to adsorption by graphitic carbon nitride. Finally, after 120 min of radiation, the removal amount of ofloxacin and ibuprofen reached 73 and 33%, respectively. The concentration of these pollutants was determined using UV-visible spectroscopy at maximum wavelengths of 220 and 287 nm for ibuprofen and ofloxacin, respectively.

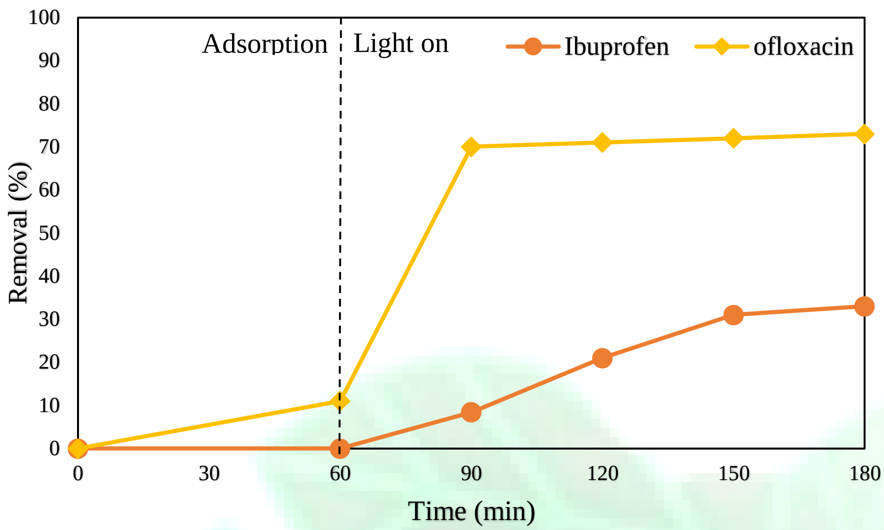

Figure 5: Ibuprofen and ofloxacin degradation using graphitic carbon nitride

4. Conclusion

In summary, graphitic carbon nitride was prepared by thermal polymerization of urea. Then, using characterization analysis of XRD, FTIR, and FESEM, the production of this material was successfully proved. The photocatalytic activity of graphitic carbon nitride was investigated in order to eliminate pharmaceutical pollutants. According to the results, graphitic carbon nitride showed a good capability to remove ofloxacin. For ibuprofen, the properties of graphitic carbon nitride can be improved and higher removal amount can be achieved by several modification approaches such as composite formation with other semiconductors or carbonaceous materials.

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