

## **Abstract**

Non-precious transition metals are introduced for doping, utilizing visible light and thus eliminating the costs associated with ultraviolet light generation. In this investigation, we synthesized graphitic carbon nitride doped with three metals (Iron, Nickel, and Cobalt) and evaluated their effects on various parameters compared to pristine graphitic carbon nitride (g-C<sub>3</sub>N<sub>4</sub>). Among these metals, iron exhibited the most significant changes in dye removal properties. Both pristine g-C<sub>3</sub>N<sub>4</sub> and doped g-C<sub>3</sub>N<sub>4</sub> were prepared using the thermal polymerization technique, and the results were compared among Iron (Fe) doped g-C<sub>3</sub>N<sub>4</sub>, Cobalt (Co) doped g-C<sub>3</sub>N<sub>4</sub>, Nickel (Ni) doped g-C<sub>3</sub>N<sub>4</sub> and pristine g-C<sub>3</sub>N<sub>4</sub>. Their optical properties under sunlight were further examined using techniques such as UV-Vis diffuse reflectance spectroscopy (UV-Vis DRS), and elemental changes were analysed using Energy Dispersive X-ray (EDX) spectroscopy for all four materials. Surface areas, adsorbate volumes, and pore volumes of g-C<sub>3</sub>N<sub>4</sub> and iron-doped g-C<sub>3</sub>N<sub>4</sub> (g-C<sub>3</sub>N<sub>4</sub>/Fe) were determined using BET studies. Additionally, alterations in properties were assessed using Fourier-transform infrared spectroscopy (FTIR) and X-ray diffraction (XRD). The photocatalytic activity of the aforementioned catalysts was evaluated by degrading the hazardous dye Congo Red (CR) under direct sunlight. Fe-doped g-C<sub>3</sub>N<sub>4</sub> exhibited the most promising results at a catalyst dose of 2 mg/l. Furthermore, the rate order was calculated, confirming it to be pseudo first order (PFO). All the materials and chemicals used during the experiments were melamine for the synthesis of g-C<sub>3</sub>N<sub>4</sub>; FeSO<sub>4</sub>, CoSO<sub>4</sub> and C<sub>12</sub>H<sub>10</sub>Ni<sub>3</sub>O<sub>14</sub> (Nickel citrate), as source of metal, for fabrication of corresponding metal doped g-C<sub>3</sub>N<sub>4</sub> and Congo red (CR) (C<sub>32</sub>H<sub>22</sub>N<sub>6</sub>O<sub>6</sub>S<sub>2</sub>Na).

Synthesis of all photocatalysts was by thermal condensation method with melamine of fixed quantity as a precursor at 600°C for 6h. The resulting yellow powder was washed with ethanol followed by distilled water. The same process was repeated with the addition of a metal compound (5% wt. of melamine) with melamine. To ensure proper mixing, 10 ml water was added to form a slurry. The photocatalytic activity of g-C<sub>3</sub>N<sub>4</sub> was monitored using photoreactor equipped with visible light by choosing a common hazardous pollutant, solution of congo red (CR) dye. Before the start of the photocatalytic reaction, a photocatalyst is added to the solution to meet the adsorption-desorption equilibrium for 30 minutes. After 30 minutes light is turned on for completion of photocatalytic reaction After completion of the reaction, the solution is centrifuged at a constant speed for 5 minutes to separate the particles of the photocatalyst. The concentration of CR was metered by UV-Vis spectrophotometer at 497nm. g-C<sub>3</sub>N<sub>4</sub> and other

catalysts with metallic doping are characterized by XRD, FTIR, UV-DRS, EDX, and BET surface area analyser. XRD results revealed that the peak at  $27.3^\circ$  and  $13.3^\circ$  which is showing the presence of hexagonal rings and layered structure of material respectively which is in accordance with JCPDS 87-1526. In doped catalyst, peak at  $13.3^\circ$  (100) is having more layer are formed. FTIR results show the various peaks vibrations associated with the tri s-triazine aromatic repeating units are affirmed by absorption peaks at wavenumbers of  $885\text{ cm}^{-1}$  and  $800\text{ cm}^{-1}$ . Before metal doping, BG was  $2.7\text{eV}$  which is reduced after metal doping and in case on iron doping is least  $2.43\text{eV}$  as compared to  $2.50\text{eV}$  in case of Nickel and  $2.47\text{ eV}$  in case of Cobalt.

Doping also doubled the surface area from  $18.30\text{ m}^2/\text{g}$  to  $35.00\text{ m}^2/\text{g}$  and pore volume from  $0.16$  to  $0.30\text{ cm}^3/\text{g}$ . Also doping of iron led to gas adsorption volume  $110$  to  $200\text{ cm}^3/\text{g}$ .

Degradation reaction is found to be first order with pristine  $\text{g-C}_3\text{N}_4$  and  $\text{g-C}_3\text{N}_4/\text{Fe}$  with rate constants rose to  $0.0055$  to  $0.0077\text{ min}^{-1}$

We fabricated iron doped  $\text{g-C}_3\text{N}_4$ , nickel doped  $\text{g-C}_3\text{N}_4$  and cobalt doped  $\text{g-C}_3\text{N}_4$  and a compared with  $\text{g-C}_3\text{N}_4$ . Among all, iron doped  $\text{g-C}_3\text{N}_4$  was good as compared to all other.

During the study, catalyst dose was kept constant at  $2\text{ mg/l}$ ,  $20\text{W}$  light bulb was used and during the process pH of the solution was kept constant at  $6.5-7$

Holes are playing the leading role in degradation process.

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