Synthesis, Characterization and Enhanced visible light induced photocatalytic activity of NiO/g-C3N4 nanocomposite

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Abstract

Heterogeneous binary nanocomposite based photocatalysis is a potential strategy to resolve the worldwide ecological issues. Here, graphitic carbon nitride (g-C₃N₄) containing NiO nanocomposite (NC) was synthesized by simple mixing. Pure NiO and g-C₃N₄ was synthesised by hydrothermal and thermal decomposition methods respectively. As synthesized g-C₃N₄, NiO, and NiO/g-C₃N₄, composite were characterized by XRD, FTIR, UV-vis spectroscopy, FE-SEM, and HR-TEM. NiO nanoparticles were uniformly distributed on C₃N₄ matrix. Photocatalytic degradation of Rhodamine B (RhB) was investigated under visible light irradiation. The results confirm that 81% of RhB was degraded within 60 min under NiO/g-C₃N₄ visible light system. This study suggests that potential in environmental applications.

1. Introduction

Recently most of the chemical industries using several toxic substances such as chlorophenols, heavy metals, dioxins, polychlorinated biphenyls, asbestos, and dyes as in the form of solvents, and active ingredients for preparation of products [1]. After completion of chemical process, they are let out through our environment [2]. These toxic chemicals cause severe health issues in human beings such as skin infection, cancer, nervous problem, and waterborne disease [3]. Even aquatic and terrestrial animals also affected and it reflects in ecological imbalance. Dyes are unavoidable necessary materials for our lifestyle. Nowadays, rhodamine B (RhB) is a widely used dye in medicine, biological staining, painting, printing, food, and leather industries [4]. It has highly colorant which leads to the carcinogenic/mutagenic to the living organisms [5]. RhB has a diminution of the light diffusion in water which reduces aquatic species photosynthesis and natural water purification [6]. Hence, RhB removal is very important from the environment water/industrial wastewater before deteriorating in the ecosystem.

Several methods such as adsorption, reverse osmosis, ion exchange, and biological methods were employed to the removal of RhB [7]. Photocatalytic degradation of dye molecules is one of the best methods compare to other techniques [8]. In photocatalysis, many of the semiconductors like zinc oxide (ZnO), tungsten oxide (WO₃), strontium titanate (SrTiO₃), and titanium dioxide (TiO₂) are acted as dynamic photocatalysts [9–12]. Among others, NiO is one of the semiconducting materials. It has a better chemical stability and adsorption property [13]. However, NiO has less absorption in the visible region and also limits the photocatalytic efficiency [14, 15].

On the other hand, g-C₃N₄ is a chemically stable and non-hazardous metal free semiconductor photocatalyst. Because, it has a band gap of 2.8 eV [16], which is responsible to absorb visible light region and it was suggested to be the best option to produce nano heterostructure. Moreover, g-C₃N₄ can be easily synthesized by using economically available precursors such as urea, melamine, cyanamide, and thiourea [17]. Though, NiO acts as a photocatalyst, the efficiency is low as compared with the g-C₃N₄ [18]. Generally, bare metal oxides has some limitations regarding the charge separation and difficult to transfer and participate redox reaction [19]. To overcome this limitation, it is necessary to prepare new
composites to further improve the photocatalytic efficiency of dye degradation [20–21]. Hence, combination of NiO and g-C\(_3\)N\(_4\) could attain the efficient visible-light-driven photocatalytic activity without any additional oxidant [22]. For example, recently, reported composite like MgO-g-C\(_3\)N\(_4\), ZnO/g-C\(_3\)N\(_4\), MnO\(_2\)-g-C\(_3\)N\(_4\) performed outstanding photocatalytic activity than their corresponding single entity [23–27]. In the present study NiO/g-C\(_3\)N\(_4\) nanocomposite was synthesized by the simple mixing method and applied as photocatalyst to the RhB photo degradation reaction assisted with visible light illumination.

2. Materials And Methods

2.1. Reagents required

Nickel acetate tetrahydrate, sodium hydroxide, rhodamine B and urea were purchased from Sigma-Aldrich (India) and used as received. Distilled water was used as solvent for the whole experiments.

2.2. Synthesis of g-C\(_3\)N\(_4\)

The g-C\(_3\)N\(_4\) sheets were synthesized by the direct decomposition method [28]. By this method, 5 g of urea were taken in a silica crucible covered with a lid and it was kept into the muffle furnace (JOTHI) up to 550 °C for about 4 h.

2.3. Synthesis of NiO-NP

NiO was synthesized by the hydrothermal method. 20 ml of 1 M nickel acetate tetrahydrate aqueous solution was taken and stirred till dissolved and then 20 ml of 0.5 M of aqueous sodium hydroxide solution was added dropwise under vigorous stirring which forms green color nickel hydroxide precipitate. Further, the suspension was transferred into a 50 ml capacity of Teflon-lined autoclave and maintained a temperature of 120 °C over 6 h. Then the obtained precipitate was washed with water and ethanol. Finally, the dried powder was calcined at 400 °C up to 4 h thus forming nickel oxide nanoparticles.

2.4. Preparation of NiO-g-C\(_3\)N\(_4\)

The 80 mg of NiO was dispersed in 50 mL of distilled water. Then 20 mg of g-C\(_3\)N\(_4\) was dispersed in 50 mL of de-ionized water separately. g-C\(_3\)N\(_4\) dispersion was added to NiO aqueous dispersion under constant stirring up to 2 hours. Afterward, it was washed with ethanol and water several times and dried at 70 °C over night to obtain NiO-gC\(_3\)N\(_4\) nanocomposite.

2.5. Characterization techniques

The crystalline nature of the synthesized material was studied by powder X-ray diffraction (PAN analytical) with Cu K\(\alpha\) radiation at a voltage of 40 kV. Fourier transform infrared spectra were recorded by IR tracer 500, the surface morphology and elemental composition were identified by field emission scanning electron microscopy (FESEM, FEI Quanta FEG 200 with an accelerating voltage of 20 kV).
coupled with EDS. The transmission electron microscopic (TEM) images were captured by using a JEOL, JEM, Fb-2000 instrument at an accelerating voltage of 200 kV. The UV–Vis (AGINITY 5000) diffuse reflectance spectroscopy using BaSO$_4$ as a reference was used to determine the optical property of samples.

2.6. Dye degradation test

Photocatalytic property of bare NiO, g-C$_3$N$_4$, and their binary nanocomposites was studied by using the model of RhB pollutant under visible light irradiation. First, 10 mg of RhB was dissolved in 100 mL of distilled water in addition to that 100 mg of the synthesized sample were added and kept in constant stirring up to 1 h to get an equilibrium solution. Finally, the solution was placed under visible light irradiation to get the solution at every 10 minutes intervals. Each sample was filtered and analyzed through UV Visible absorption spectroscopy. The decolorization efficiency was calculated by using the equation (s).

$$%ofDecolourisation = \frac{C_0 - C_1}{C_0} \times 100$$

1

Here, $C_0$ is the initial concentration of the RhB dye solution; $C_1$ is the concentration of the dye solution at various time intervals under visible-light irradiation [29-30].

3. Results And Discussion

3.1. Physicochemical studies

Figure 2 (a-c) shows the XRD pattern of NiO, g-C$_3$N$_4$ and NiO/g-C$_3$N$_4$. The NiO diffraction peaks at 37.0$^\circ$, 43.2$^\circ$, 62.4$^\circ$, 75.4$^\circ$ and 79.3$^\circ$ corresponding to the reflective planes of (111), (200), (220), (311) and (222), respectively. This result is well-matched with the JCPDS card no-04-0835. These diffraction peaks are perfectly indexed with face-centered cubic phase crystalline structure of NiO (Fig. 2a) [31]. The diffraction peaks exist at 13.4$^\circ$ and 27.4$^\circ$ which corresponds to (100), and (002) planes of bare g-C$_3$N$_4$ (JCPDS card No-65-287), These peaks are attributed to interplanar staking of aromatic systems along with inter-layer structural packing represented in Fig. 2(b) [32]. After the addition of g-C$_3$N$_4$ into NiO the diffraction peak intensity of NiO was increased (Fig. 2c). Moreover a minor peak appeared at 27.4$^\circ$ confirms the presence of g-C$_3$N$_4$ in the composite material. It may occur due to dominant of diffraction peak of NiO[33].

FTIR spectrum (Fig. 3a) of bare NiO exhibits the peak at 833 cm$^{-1}$ which is assigned to Ni-O [34], The peak, 1352 cm$^{-1}$ is assigned to symmetric and asymmetric stretching vibration of O–C = O, 1636 cm$^{-1}$ assigned to H-O-H bending vibration, 2342 cm$^{-1}$ assigned to CO$_2$ mode, peak formed around 3459 cm$^{-1}$ indicates the presence of water molecule even after calcination of NiO [35]. Figure 3b shows the FTIR spectrum of bare g-C$_3$N$_4$. It shows a peak at 805 cm$^{-1}$ due to bending vibration mode of triazine units,
and numerous band appeared at 1277 cm$^{-1}$, 1321, 1558, 1634 could be assigned to C-N, 3184 cm$^{-1}$ indicates O-H bond [36]. Finally we compare with binary NiO/g-C$_3$N$_4$ nanocomposite exhibits absorption band similar to bare g-C$_3$N$_4$. After the doping of NiO with g-C$_3$N$_4$, the spectrum does not show any other vibration peak and the corresponding peaks were indicates the pure phases of NiO and g-C$_3$N$_4$, confirms the formation of NiO/g-C$_3$N$_4$ nanocomposite.

### Optical studies

Optical absorption behavior of the synthesized bare NiO, g-C$_3$N$_4$ and NiO/ g-C$_3$N$_4$, was evaluated by DRS UV-Vis spectroscopy (Fig. 4A (a-c). Bare NiO exhibits a strong absorption in Uv and visible region and bare g-C$_3$N$_4$ exhibits the absorbance edge at 470 nm respectively. Then we analyze NiO/g-C$_3$N$_4$ composite that exhibits the absorption edge as same as bare NiO due to the highest loading of NiO in NiO/g-C$_3$N$_4$ composite. The band gap value of the bare and binary nanocomposite was also evaluated by Tauc's plot method

\[(αhv)2 = C(hν − E_g)\]

Here $α$ is a light absorption coefficient, $h$ is a plank constant, $ν$ is a light frequency, $C$ is a velocity of light, $E_g$ is band gap energy. The band gap values of bare NiO, g-C$_3$N$_4$ and NiO/ g-C$_3$N$_4$ andwere 2.67 eV, 2.86eV and 2.45 eVis given in Fig.4B(a-c) respectively. The band gap energy of NiO/g-C$_3$N$_4$ is lower than that of bare NiO and g-C$_3$N$_4$ indicate the absorption of NiO/g-C$_3$N$_4$ nanocomposite is shifted towards lower energy region[37]

### 3.2. FESEM and HRTEM

Morphological observations of as prepared bare NiO, bare g-C$_3$N$_4$ and NiO/g-C$_3$N$_4$ was done by scanning electron microscope coupled with EDS. FESEM images (Fig. 5a-c) of bare NiO exhibit spherical shape which results in enough rough surface and large specific surface area and active adsorption sites.[38] Furthermore bare NiO was characterized by HRTEM and the obtained images are given in Fig. 5.(d & e). It could observe an agglomerated spherical morphology which was consistent with SEM results. Figure 5(f) shows the SAED pattern of bare NiO.

Fig. 6(a, b) shows the FESEM images of bare g-C$_3$N$_4$ which exhibits sheet like morphology and contains some wrinkles on it. Further, sheet like morphology of as prepared bare g-C$_3$N$_4$ was investigated by high resolution transmission electron microscopic technique (Fig.6 c,d). The observed images were exfoliated g-C$_3$N$_4$ nanosheets which has highly light transmittance features might reflects ultrathin layered structure [39].

While the preparation of NiO/g-C$_3$N$_4$ composite, NiO NP's was successfully deposited over the surface of g-C$_3$N$_4$ nanosheets. Figure 7(a) represents FESEM image of NiO/g-C$_3$N$_4$ composite. It exhibits an aggregated and rough surface of the structure which enhances the absorption capacity of the dye. It
could further increase the photocatalytic activity of the composite material [40]. Moreover, the HRTEM image of NiO/g-C\textsubscript{3}N\textsubscript{4} shows that spherical NiO nanoparticles are evenly distributed on the surface of g-C\textsubscript{3}N\textsubscript{4} nanosheets (Fig. 7b). EDS spectrum of NiO/g-C\textsubscript{3}N\textsubscript{4} composite (Fig. 7c) indicates the corresponding peaks confirms C, N, O and Ni elements were present in the NiO/g-C\textsubscript{3}N\textsubscript{4} nanocomposite [41].

3.4. Photo catalytic performance

Photo catalytic activity of bare NiO, g-C\textsubscript{3}N\textsubscript{4}, and NiO/g-C\textsubscript{3}N\textsubscript{4} nanocomposites was carried out by visible light irradiation. It is noteworthy that the RhB was degraded up to 81% after 60 min in presence of NiO/g-C\textsubscript{3}N\textsubscript{4} nanocomposite. While, bare NiO and g-C\textsubscript{3}N\textsubscript{4}, photocatalyst exhibits 73% and 75% of efficiency (Fig. 8 (a-c)). A photocatalytic activity of these materials was elucidated by kinetic studies and decomposition rate is calculated by pseudo first order kinetic equation [42–43]. In the present work, a feasible mechanism of the enhanced photocatalytic activity of g-C\textsubscript{3}N\textsubscript{4}/NiO nanocomposite was associated with the efficient absorbance of visible light and effective separation of electron-hole pairs. The g-C\textsubscript{3}N\textsubscript{4} and NiO band edge positions are calculated using the below equations

\[ E_{VB} = X - E_c + 0.5E_g \]

\[ E_{CB} = E_{VB} - E_g \]

Where, \( E_{VB} \), \( E_{CB} \), \( X \), and \( E_g \) have represented the valance band (VB), conduction band (CB) potentials, Mulliken's electronegativity of semiconductor, and energy band gap respectively. The \( X \) values for g-C\textsubscript{3}N\textsubscript{4} and NiO are calculated to be 4.73 and 5.76 respectively [44-47]. As per the above equations the calculated VB, CB values for g-C\textsubscript{3}N\textsubscript{4} (\( E_{VB} \) 1.43eV, \( E_{CB} \) -0.97 eV), NiO (\( E_{VB} \) 2.79 eV, \( E_{CB} \) -0.27 eV), respectively, and the band gap energies were calculated by Tauc's-plot. According to the calculated band edge value positions of g-C\textsubscript{3}N\textsubscript{4} and NiO, the band arrangement of composite with a charge transfer mechanism were also evaluated.

The kinetic studies were elucidated in Fig. 9A feasible mechanism of the enhanced photo catalytic activity of gC\textsubscript{3}N\textsubscript{4}/NiO nanocomposite was associated with the efficient absorbance of visible light and separation of electron hole pairs. After the irradiation of g-C\textsubscript{3}N\textsubscript{4} using visible light (\( \lambda > 420 \) nm), the conduction band potential of \( E_{CB} \)=-0.97 V vs RHE g-C\textsubscript{3}N\textsubscript{4} could easily absorb visible light further generate electrons/pairs. Generally the photo-induced electron-hole pairs again recombined with bare g-C\textsubscript{3}N\textsubscript{4}. This results of bare g-C\textsubscript{3}N\textsubscript{4} is unsatisfied the degradation reaction mechanism. Further improve the efficiency of bare g-C\textsubscript{3}N\textsubscript{4} is mixing with NiO NP's and formation of NiO/g-C\textsubscript{3}N\textsubscript{4} nanocomposites. This nanocomposite prevents the recombination of electrons and holes. The interaction of photogenerated electrons and holes would react with oxygen and water to generate superoxide radicals (\( \cdot O_2^- \)) and (\( \cdot OH \)) radicles. Therefore holes present in the valence band of NiOand surface of the g-C\textsubscript{3}N\textsubscript{4} may also reacts with H\textsubscript{2}O to form new \( \cdot OH \) radicals which have a capacity to oxidize dye molecules. From the whole, these radicals and electrons which is remain in the g-C\textsubscript{3}N\textsubscript{4} conduction band reacts with oxygen to
generate reactive superoxide radicals ($\cdot O_2^-$) and they react with water through the stepwise reduction of $O_2$ to produce $\cdot OH$ radicals. Thus increases the spatial separation of electron hole pair reduces the possibility of recombination and increased hydroxyl and super oxide ions which are essential oxidizing element used for the improvement of the photocatalytic activity. [48–50].

4. Conclusions

In summary g-C$_3$N$_4$ based NiO/g-C$_3$N$_4$ composite was synthesized by facile physical mixing method. The NiO with g-C$_3$N$_4$ offered high active radical ($\cdot O_2^-$) species and prevented electron/hole pair recombination. As synthesized NiO/g-C$_3$N$_4$ showed excellent photocatalytic activity of RhB under visible light irradiation as compared with bare NiO and g-C$_3$N$_4$. The enhanced photocatalytic activity of NiO/g-C$_3$N$_4$ was achieved due to synergistic effect between NiO and g-C$_3$N$_4$.

Declarations

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Conflict of Interest

No conflict of interest exists.

We wish to confirm that there are no known conflicts of interest associated with this publication and there has been no significant financial support for this work that could have influenced its outcome.

Credit Author Statement

M.Gayathri: Conceptualization, Data curation, Investigation, Visualization, Writing - original draft.

N.Jayaprakash: Image editing, interpretations, and writing.

E.Sundaravadivel: Project administration and Supervision.

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