

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

M. Gayathri

SRM Institute of Science and Technology

N. Jayaprakssh

Valliammai Engineering College

Sundaravadivel E (✉ sundaravadivelchem@gmail.com)

SRM University <https://orcid.org/0000-0003-0209-6192>

Research Article

Keywords: Binary composite, graphitic carbon nitride, NiO, rhodamine B

DOI: <https://doi.org/10.21203/rs.3.rs-298371/v1>

License:   This work is licensed under a Creative Commons Attribution 4.0 International License.

[Read Full License](#)

Abstract

Heterogeneous binary nanocomposite based photocatalysis is a potential strategy to resolve the worldwide ecological issues. Here, graphitic carbon nitride ($g\text{-C}_3\text{N}_4$) containing NiO nanocomposite (NC) was synthesized by simple mixing. Pure NiO and $g\text{-C}_3\text{N}_4$ was synthesised by hydrothermal and thermal decomposition methods respectively. As synthesized $g\text{-C}_3\text{N}_4$, NiO, and NiO/ $g\text{-C}_3\text{N}_4$, composite were characterized by XRD, FTIR, UV-vis spectroscopy, FE-SEM, and HR-TEM. NiO nanoparticles were uniformly distributed on C_3N_4 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/ C_3N_4 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 (WO_3), strontium titanate (SrTiO_3), and titanium dioxide (TiO_2) 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, $g\text{-C}_3\text{N}_4$ 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, $g\text{-C}_3\text{N}_4$ 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 $g\text{-C}_3\text{N}_4$ [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-C₃N₄ could attain the efficient visible-light-driven photocatalytic activity without any additional oxidant [22]. For example, recently, reported composite like MgO-g-C₃N₄, ZnO/g-C₃N₄, MnO₂-g-C₃N₄ performed outstanding photocatalytic activity than their corresponding single entity [23–27]. In the present study NiO/g-C₃N₄ 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-C₃N₄

The g-C₃N₄ 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-C₃N₄

The 80 mg of NiO was dispersed in 50 mL of distilled water. Then 20 mg of g-C₃N₄ was dispersed in 50 mL of de-ionized water separately. g-C₃N₄ 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-g-C₃N₄ nanocomposite.

2.5. Characterization techniques

The crystalline nature of the synthesized material was studied by powder X-ray diffraction (PAN analytical) with Cu K α 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 BaSO₄ as a reference was used to determine the optical property of samples.

2.6. Dye degradation test

Photocatalytic property of bare NiO, g-C₃N₄, 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).

$$\%of\text{Decolourisation} = \frac{C_0 - C_1}{C_0} \times 100$$

1

Here, C₀ is the initial concentration of the RhB dye solution; C₁ 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-C₃N₄ and NiO/g-C₃N₄. The NiO diffraction peaks at 37.0°, 43.2°, 62.4°, 75.4° and 79.3° 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° and 27.4° which corresponds to (100), and (002) planes of bare g-C₃N₄ (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-C₃N₄ into NiO the diffraction peak intensity of NiO was increased (Fig. 2c). Moreover a minor peak appeared at 27.4° confirms the presence of g-C₃N₄ 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 CO₂ 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-C₃N₄. 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-C₃N₄ nanocomposite exhibits absorption band similar to bare g-C₃N₄. After the doping of NiO with g-C₃N₄, the spectrum does not show any other vibration peak and the corresponding peaks were indicates the pure phases of NiO and g-C₃N₄, confirms the formation of NiO/g-C₃N₄ nanocomposite.

Optical studies

Optical absorption behavior of the synthesized bare NiO, g-C₃N₄ and NiO/ g-C₃N₄, 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-C₃N₄ exhibits the absorbance edge at 470 nm respectively. Then we analyze NiO/g-C₃N₄ composite that exhibits the absorption edge as same as bare NiO due to the highest loading of NiO in NiO/g-C₃N₄ composite. The band gap value of the bare and binary nanocomposite was also evaluated by Tauc's plot method

$$(\alpha h\nu)^2 = C(h\nu - E_g)$$

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-C₃N₄ and NiO/ g-C₃N₄ and were 2.67 eV, 2.86eV and 2.45 eV is given in Fig.4B(a-c) respectively. The band gap energy of NiO/g-C₃N₄ is lower than that of bare NiO and g-C₃N₄ indicate the absorption of NiO/g-C₃N₄ nanocomposite is shifted towards lower energy region[37]

3.2. FESEM and HRTEM

Morphological observations of as prepared bare NiO, bare g-C₃N₄ and NiO/g-C₃N₄ 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-C₃N₄ which exhibits sheet like morphology and contains some wrinkles on it. Further, sheet like morphology of as prepared bare g-C₃N₄ was investigated by high resolution transmission electron microscopic technique (Fig.6 c,d). The observed images were exfoliated g-C₃N₄ nanosheets which has highly light transmittance features might reflects ultrathin layered structure [39].

While the preparation of NiO/g-C₃N₄ composite, NiO NP's was successfully deposited over the surface of g-C₃N₄ nanosheets. Figure 7(a) represents FESEM image of NiO/g-C₃N₄ 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-C₃N₄ shows that spherical NiO nanoparticles are evenly distributed on the surface of g-C₃N₄ nanosheets (Fig. 7b). EDS spectrum of NiO/g-C₃N₄ composite (Fig. 7c) indicates the corresponding peaks confirms C, N, O and Ni elements were present in the NiO/g-C₃N₄ nanocomposite [41].

3.4. Photo catalytic performance

Photo catalytic activity of bare NiO, g-C₃N₄, and NiO/g-C₃N₄ 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-C₃N₄ nanocomposite. While, bare NiO and g-C₃N₄, 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-C₃N₄/NiO nanocomposite was associated with the efficient absorbance of visible light and effective separation of electron-hole pairs. The g-C₃N₄ 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-C₃N₄ 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-C₃N₄ (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-C₃N₄ 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 g-C₃N₄/NiO nanocomposite was associated with the efficient absorbance of visible light and separation of electron hole pairs. After the irradiation of g-C₃N₄ using visible light ($\lambda > 420$ nm), the conduction band potential of $E_{CB}=-0.97$ V vs RHE g-C₃N₄ could easily absorb visible light further generate electrons/pairs. Generally the photo-induced electron-hole pairs again recombined with bare g-C₃N₄. This results of bare g-C₃N₄ is unsatisfied the degradation reaction mechanism. Further improve the efficiency of bare g-C₃N₄ is mixing with NiO NP's and formation of NiO/g-C₃N₄ 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 ($\bullet\text{O}_2^-$) and ($\bullet\text{OH}$) radicles. Therefore holes present in the valence band of NiO and surface of the g-C₃N₄ may also reacts with H₂O to form new $\bullet\text{OH}$ radicals which have a capacity to oxidize dye molecules. From the whole, these radicals and electrons which is remain in the g-C₃N₄ conduction band reacts with oxygen to

generate reactive superoxide radicals ($\cdot\text{O}_2^-$) and they reacts with water through the stepwise reduction of O_2 to produce $\cdot\text{OH}$ 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 $\text{g-C}_3\text{N}_4$ based $\text{NiO/g-C}_3\text{N}_4$ composite was synthesized by facile physical mixing method. The NiO with $\text{g-C}_3\text{N}_4$ offered high active radical ($\cdot\text{O}_2^-$) species and prevented electron/hole pair recombination. As synthesized $\text{NiO/g-C}_3\text{N}_4$ showed excellent photocatalytic activity of RhB under visible light irradiation as compared with bare NiO and $\text{g-C}_3\text{N}_4$. The enhanced photocatalytic activity of $\text{NiO/g-C}_3\text{N}_4$ was achieved due to synergistic effect between NiO and $\text{g-C}_3\text{N}_4$.

Declarations

Acknowledgment

The authors thanks to SRM Institute of Science and Technology for providing major instrumentation facilities (NRC-SRMIST and HRTEM FACILITY at SRMIST set up with support from MNRE (Project No. 31/03/2014-15/PVSE-R&D, Government of India).

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.

References

1. Y.N. Rane, D.A. Shende, M.G. Raghuwanish, R.R. Koli, S.R. .Gosavi, N.G.Deshpande, Visible-light assisted CdO nanowires photocatalyst for toxic dye degradation studies. *Optik*. **179**, 535–544 (2019)
2. M. A.Tadjarodi.H.Kerdari Imani, Experimental design to optimize the synthesis of CdO cauliflower – like nano structure and high performance in photodegradation of various azo dyes. *Mater.Res.Bulletin*. **48**, 935–942 (2013)

3. P. Dhatshanamurthi, B. Subash, M. Shanthi, Investigation on UV-A light photocatalytic degradation of an azo dye in the presence of CdO/TiO₂ coupled semiconductor. *Mater.Sci.Semicond.Process.* **35**, 22–29 (2015)
4. D. D. Battacharya, G. Joshal, D. Mondal, B. K. Paul, N. Bose, S. Das, M. Basu, Visible Light Driven Degradation of Brilliant Green dye using Titanium based ternary metal oxide photocatalyst. *RESULTS.PHYS.* **12**, 1850–1858 (2019)
5. D. C. Debabrata. Shimanta, Visible light induced photocatalytic degradation of organic pollutants. *J.Photochem.Photobiol C.* **6**(2–3), 186–205 (2005)
6. Y. Chen, J. Li, B. Zhai, Y. Liang, Enhanced photocatalytic degradation of RhB by two dimensional composite photocatalyst. *Colloid and surf A.* **568**, 429–435 (2019)
7. S. Vasantharaj, S. S. Mythili, P. Senthilkumar, G. Kavitha, M. Shanmugavel, E. Manikandan, A. Pugazhendi, Synthesis of ecofriendly copper oxide nanoparticles for fabrication over textiles fabrics characterization of antibacterial activity and dye degradation potential. *Journal of photochem& photo biol B.* **192**, 143–149 (2018)
8. M. A. Rauf, S. Ashraf, Fundamental Principle and application of heterogeneous photocatalytic degradation of dyes in solution. *J.Chem.Engg.* **151**, 10–18 (2000)
9. Haneda Hajmine LiD, Morphologies of Zinc oxide nanoparticles and their effects on photocatalysis. *Chemosphere.* **51**(2), 129–137 (2003)
10. A. Fujishima, X. Zhang, D. A. Tryk, TiO₂ photocatalysts and related surface phenomena. *Surf.Sci.Rep.* **63**, 515–582 (2008)
11. I. Atkinson, V. Parvulescu, J. P. Cusu, E. M. Anghel, M. D. Voicescu. S. Culita. C. Somacescu. Munteanu, Influence of preparation method and nitrogen (N) doping on properties and photo-catalytic activity of mesoporous SrTiO₃. *J.photochem&photobiol A. chem.* **368**, 41–51 (2019)
12. S. Ghafoor, A. Inayat, F. Aftab, H. Duran, K. Kirchhof, S. Waseem, S. N. Arshad, TiO₂ nanofibres embedded with g-C₃N₄ nanosheets and decorated with Ag nanoparticles as Z-scheme photocatalysts for environmental Remediation. *J. Environ. Chem. Engg.* **7**(6), 103452 (2019)
13. D. Delgado, R. Sanchis, J. A. Cecilliac, E. Rodriguez-Castelionc, A. Cabalierod, B. Solsonab, J. M. Lopez, Support effects on NiO-based catalysts for the oxidative dehydrogenation (ODH) of ethane. *Catal.Today.* **333**, 10–16 (2019)
14. J. Wang, Y. Liu, Z. Wang, P. Wang, Z. Zheng, X. Qin, X. Zhang, Y. Dal, B. Huang, Enhanced photocatalytic activity towards H₂ evolution over NiO via phosphonic acid surface modification with different functional group. *INT.J.HYDROGEN,ENERG.* **31**(21), 16575–16581 (2019)
15. Y. Bu, Z. Chen, Role of polyaniline on the photocatalytic Degradation and stability performance of the polyaniline/Silver/Silver phosphate composite under Visible Light. *Appl.Mater. interface.* **6**, 17589–17598 (2014)
16. L. Ge, Synthesis and photocatalytic performance of novel metal free g-C₃N₄ photocatalysts. *Mater.Lett.* **65**, 2652–2654 (2011)

17. C. Li, Z. Loua, Y. Yang, Y. Wang, Y. Lu, Z. Ye, L. Zhu, Hollowsphere Nanohetero junction of g-C₃N₄ @ TiO₂ with high visible light photocatalytic property. *Langmuir*. **35**(3), 779–786 (2019)
18. G. Tzvetkov, M. Tzvetkov, T. Spassov, Ammonia-evaporation-induced construction of three dimensional NiO-g-C₃N₄ composite with enhanced adsorption and visible light active driven photocatalyst. *Super Lattices Microstru.* **119**, 122–132 (2018)
19. Difa Xu, B. Cheng, W. Wang, C. Jiang, J. Yu, Ag₂CrO₄/g-C₃N₄/ grapheme oxide ternary composite Z-scheme photocatalyst with enhanced CO₂ reduction activity. *Appl. catal. B: Environ.* **231**, 368–380 (2018)
20. Y. Beibel Wu, K. Li, L. Su, Tan, X. Liu, Z.. Cui, X. Yang, Y. Liang, Z.. Li, S. Zhu, K. W. K. . Yeung, Shulin Wu, The enhanced photovatalytic properties of MnO₂/g-C₃N₄ heterostructure for Rapid sterilization under Visible Light. *J. HAZARD. MATER.* **337**, 227–236 (2019)
21. W. Liu, C. Wang, L. Wang, Photo catalyzed the facile synthesis of #-chloro aryl ketones with PANI/g-C₃N₄/TiO₂ composite under visible ligh. *Ind. Eng. Chem. Res.* **56**(31), 6114–6123 (2017)
22. H. Y. Xu, H. Zhao Li-Cheng, Li-Guolin, Shu-Yan Qi, Synergic effect btrween adsorption and photocatalysis of metal free C₃N₄ derived from different precursors. *PLOS ONE.* (2015)
23. L. Williams, A. R. Prasad, P. Sowmya, A. Joshep, Characterization and Temperature dependence DC Conductivity study of Bio template NiO and their composites using polyaniline. *Mater. Chem. phys.* **242**, 122469 (2019)
24. L. X. Shen. L. Song. Luo, Y. Zhang, J.. Z. Liu, L. Zhang, Preparation of TiO₂/C₃N₄ heterojunctions on carbon-fiber cloth as efficient filter-membrane – shaped photocatalyst for removing various pollutanta from the flowing waste water. *J. Colloid. Interface Sci.* **532**, 798–807 (2018)
25. J. X. N. Mao, . Jiang, MgO/g-C₃N₄ nanocomposites as efficient water splitting photocatalysts under visible light irradiation. *Appl. Surf. Sci.* **476**, 144–150 (2019)
26. V. V. Anusha, S. Thangarajan, K. Anupriya, B. Subramanian, Functionalization of fabric with PANI-CuO nanoparticles by precipitation route for antibacterial applications. *J. Nanopart. Res.* 17–57 (2017)
27. C. X. Yuan. Q. Zhon. Jing, Q. Tang, Y. Mu, A. K. Du, Facile synthesis of g-C₃N₄ nanosheets/Zno nanocomposites with enhanced photocatal photocatalytic activity in reduction of aqueous chromium (VI) under visible light. *Nanomaterials.* **6**, 173 (2016)
28. E. Y. Hang. J. Liu. Shi, X. Lin, L. Sheng, M.. L. Zhang. J. Wang. Chen, A direct one step synthesis of ultrathin g-C₃N₄ nanosheets from thio urea for boosting solar photocatalytic. *Int. J. Hydrogen. Energ.* H₂ evolution. **44**(14), 7194–71204 (2019)
29. S. Mahalingam, Y. Ahn, Improved visible light photocatalytic activity of rGo/Fe₃O₄/NiO hybrid nanocomposites synthesized by insitu facile method for industrial waste water treatment applications. *J. New. Chem.* **42**, 4372–4383 (2018)
30. S. M. Aydoghmish, S. A. Hassanzadeh-Tabrizi, A. Saffar-Teluri, Facile synthesis and investigation of NiO-ZnO-Ag nanocomposites as efficient photocatalysts for degradation of methylenr blue dye.. *Ceram. Int.* **45**(12), 14934–14942 (2019)

31. S. Kulkarni, M. Jadhav, P. Raikar, S. Raiker, U.S.Raiker, Core-shell novel composite metal nanoparticles for hydrogenation and dye degradation applications. *Ind.Eng.Chem.Res.* **58**(9), 3630–3639 (2019)
32. Z. Wei, H. Qiao, H. Yang, C. Zhang, X.Yan, Characterization of NiO nanoparticles by anodic arc method. *J.Alloys.comp.* **479**, 855–858 (2009)
33. L. Tan, J. Xu, X. Zhang, Z. Hang, Y. Jia, S.Wang, Synthesis of g-C₃N₄/ CeO₂ nanocomposites with improved catalytic activity on the thermal decomposition of ammonium perchlorate. *Appl.Surf.Sci.* **356**, 447–453 (2015)
34. J.Y. Tang, R.T. Guo, W.G. Zhou, C.Y. Haung, W.G. Pan, Ball-flower like NiO/g-C₃N₄ heterojunction for efficient visible light photocatalytic CO₂ reduction. *Appl.catal.B.Environ.* **237**, 802–810 (2018)
35. P.E. Saranya, S.Selladurai, Mesoporous 3D network Ce doped NiO nanoflakes as high performance electrodes for supercapacitor applications. *New.J.Chem.* **43**, 7441–7456 (2019)
36. K. X.CWang, A.Thomas Maeda, K. Takanabe, G. Xin, J.M. Carlsson, K. Domen, M.Antonietti, A metal free polymeric photocatalyst for hydrogen production from water under visible light. *Nat.Mater.* **8**(1), 76–80 (2009)
37. P. Singh, C.S. Kushwaha, S.K. Shukula, G.C.Dubey, Synthesis and Humidity sensing properties of NiO intercalated polianiline Nanocomposite. *POLM.PLAS TECH &ENGG.* **58**(2), 139–147 (2018)
38. K.Ngeyun, N.D.Hoa, C.M.Hung, D.T.T..Le, N.V.Duy, N.V. .Hiau, A comparative study on the electrochemical properties of nanoporous nickeloxide nanowire and nano sheets prepared by hydrothermal method.. *RSC Adv.* **8**, 19449–19455 (2018)
39. J.Singh, A.Arora, S.Basu, Synthesis of WO₃/ g-C₃N₄ nanocomposites for the removal of hazardous dyes under visible light. *J.Alloys.Comp.* **808**, 151734 (2019)
40. Y. Zhan, Z. Liu, Q. Liu, D. Huang, Y. Wei, Y. Hu, X.Liana, C. Hu, A facile and one pot synthesis of fluorescent graphitic carbon nitride quantum dots for bio imaging application. *New.J.Chem.* **41**, 3930–3938 (2017)
41. C.J. Mathai, S. Saravanan, M.R. Anantharaman, S. Venkatachalam, S.Jeyalakshmi, Effect of Iodine doping on the bandgap of plasma polymerized aniline thin films. *J.Phys.D.Appl.Phys.* **35**, 2206–2210 (2002)
42. H. Chen, S. Wageh, A.Ahmed Al-Ghamdi, H. Wang, J. Yu, C.Jiang, Hierarchical C/NiO-ZnO nanocomposite fibers with enhanced adsorption capacity for congo red. *J.Colloid Interfac.Sci.* **537**, 736–745 (2019)
43. Y. Fu, C. Liua, C. Zhua, H. Wang, Y. Dou, W. Shia, M. Shao, H. Huang, Y. Liua, Z.Kanga, High-performance NiO/g-C₃N₄ composites for visible-light-driven photocatalytic overall water splitting. *Inorg.* **00**, 1–3 (2013)
44. H.Y. Chen, L.G. Qiu, J.D. Xiao, S. Ye, X. Jing, Y.Peng Yuan, Inorganic-Organic hybrid NiO-g-C₃N₄ photocatalyst for efficient methylene blue degradation using visible light. *RSC Adv.* **4**, 22491–22496 (2014)

45. W. X.Ning.X. Zhen.G. Zhang.Lu, Significant enhancement of stability for visible photocatalytic overall water splitting by assembling ultra-thin layer of NiO over Zn_{1-x}Cd_xS, *chemsuschem*, 12(7),1267–1477 (2019)
46. K.Maniammal, G.Madhu, V.Biju, Nanostructured meoporousNiO as an efficient photocatalyst for degradation of methylene blue: Structure, properties and performance. *J.Nanostructur.* **16**, 266–275 (2018)
47. A. Akhundi, A.H. Yangjeh, A simple large scale method for preparation of g-C₃N₄ / SnO₂ nanocomposite as visible-light driven for degradation of an organic pollutant. *Mater.Express.* **5**(4), 309–317 (2015)
48. M.A. Alenizil, R. Kumar, M. Aslam, A. Alseroury, M.A. Barakat, Construction of ternary g-C₃N₄/TiO₂/PANI nanocomposite for the enhanced photocatalytic activity under solar light. *Sci.Rep.* **9**, 12091 (2019)
49. K.H. Rahman, A.K. Kar, Effect of band gap variation and sensitization process of polyaniline PANI-TiO₂ p-n heterojunction photocatalytic degradation of toxic methylene blue with UV irradiation. *J.Environ chem Engg.* **8**(1), 104181 (2020)
50. K. Yao, Y. Li, H. Yang, J.Yuan, Polyaniline-modified 3D spongy SnS composites for enhanced visible-light photocatalytic degradation of methyl orange. *Colloids.Surf.A.* **603**, 125240 (2020)