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Green synthesis of nickel oxide nanoparticles using plant extracts: an overview of their antibacterial, catalytic, and photocatalytic efficiency in the degradation of organic pollutants

Farzaneh Moradnia[1], Saeid Taghavi Fardood[2,*], Armin Zarei[1], Siamak Heidarzadeh[3], Ali Ramazani[1], Mika Sillanpää[4,5,*]

[1]*Department of Chemistry, Faculty of Science, University of Zanjan, Zanjan, Iran.*
[2]*Department of Chemistry, Faculty of Science, Ilam University, Ilam, Iran.*
[3]*Department of Microbiology and Virology, School of Medicine, Zanjan University of Medical Sciences, Zanjan, Iran.*
[4]*Department of Chemical Engineering, School of Mining, Metallurgy and Chemical Engineering, University of Johannesburg, South Africa.*
[5]*Department of Civil Engineering, University Centre for Research & Development, Chandigarh University, Gharuan, Mohali, Punjab, India.*

[*]**Corresponding authors:** Saeid Taghavi Fardood: s.taghavi@ilam.ac.ir, saeidt64@gmail.com
Mika Sillanpää: mikaesillanpaa@gmail.com

Review Paper

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Abstract:

In the past decade, numerous longitudinal studies have explored green chemistry and its applications in nanoparticle synthesis due to the toxicity associated with traditional methods. Among the various techniques for nanoparticle synthesis, the use of plant extracts in green synthesis has recently gained significant popularity. Green methods are particularly suitable for large-scale nanoparticle synthesis, offering faster preparation rates compared to microorganisms and the ability to produce nanoparticles in diverse sizes and shapes. Nickel oxide nanoparticles (NiO NPs) have been extensively utilized in catalysis, photocatalysis, optics, magnetism, and antibacterial applications. This review focuses on the preparation of NiO NPs using plant extracts, emphasizing their advantageous features such as the absence of contaminant release, environmental friendliness, and cost-effectiveness. Additionally, we delve into the catalytic, photocatalytic, and antibacterial applications of NiO NPs.

Keywords: Nickel oxide; Green synthesis; Catalytic activity; Antibacterial activity; Photocatalytic ability

1. Introduction

Nanotechnology is ascribed as a novel branch of science related to the synthesis of NPs and their utilizations in a wide range of areas such as health, food, space, environmental, and chemical industries, so the demand for eco-friendly and biodegradable methods for the synthesis of nanoparticles is indisputable [1–14]. Nanoparticles are of important interest owing to their very small size and large surface-to-volume ratio [15–20]. Nanoparticles are particles in the size range of 1-100 nm, and materials at the nanometer sizing represent different properties to those of bulk materials and even the isolated atom. Nanomaterials indicate fortified features such as high catalytic reactivity, thermal conductivity, and chemical steadiness [21–24]. Metal NPs and metal oxide NPs have lately received considerable attention among all the other nanomaterials due to their outstanding properties [25–31]. Generally, there are two approaches for the synthe-

sis of NPs: Bottom-up and top-down (Fig. 1). The former way of NPs synthesis involves the arrangement of smaller molecules atom-by-atom into more complex assemblies and the latter way needs bulk material. In this way, macroscopic particles are first prepared and then they are turned into nanosized materials through a process named plastic deformation. This method is less likely to be applied on a large scale as it is less cost-effective [33–36]. One of the most popular and usual techniques for the preparation of nano substances, which engaged in the top-down method, could be Interferometric Litho- graphic (IL) [37–40]. In the mentioned technique, the self-assembling of the minimized particles plays a vital role in the synthesis of nanomaterials. It is more likely to be an efficient and cost-effective approach [41–43].

In the applied physical procedures for the synthesis of nanoparticles are physical factors needed in the creation of stable and well-structured nano-based materials. The colloidal dispersion method is the best example. The other physical techniques are vapor condensation, amorphous crystallization, and physical fragmentation. Nanoparticle synthesis is gone through physical, chemical, and environment-friendly processes [44–47]. The physical methods are less cost-efficient because of using utilizing expensive facilities, and requiring high temperature and pressure. However, chemical approaches for NPs synthesis, such as chemical microemulsion, wet chemical, direct precipitation and etc. [48–55], involve toxic chemicals that could pose a threat for the environment and people who are in touch with them. Although a considerable number of studies focused on NPs synthesis from chemical and physical approaches, some terrible side effects of the mentioned two approaches, such as using noxious and carcinogenic chemicals, high vacuum usage needed, and less cost-efficacy, have been reported. [56–61] Therefore, it is undeniable that a cost-efficient and green method is required for the preparation of NPs [62–72]. A green method for the synthesis of NPs could be plant extract usage. This method possesses conving advantages such as cost-efficiency, safety, simple usage, and non-poison [73–75]. Thus, in this review, we have focused on NiO NPs synthesis by means of plant extract usage.

1.1 Nickel oxide nanoparticles

NiO NPs stand as one of the most abundant metal oxides on Earth. In the past decade, numerous studies have been conducted to synthesize NiO NPs due to their diverse applications in fields such as electronics, optics, and biomedical systems [76–81]. The preparation of metallic oxides, like TiO_2, CuO, ZnO, and NiO, has become one of the important subjects in recent studies [82–89]. Among all the mentioned inorganic metal oxides, the NiO NPs have been especially interesting among scientists because these nanoparticles are cost-efficient, easy to prepare, and safe [90]. A. Angel Ezhilarasi et al. presented the NiO NPs by means of the Aegle marmelos plant to treat hazardous diseases in India. High-resolution transmission electron microscopy (HR-TEM) and High-resolution scanning electron microscopy (HR-SEM) analysis indicated the average size of NPs between 8-10

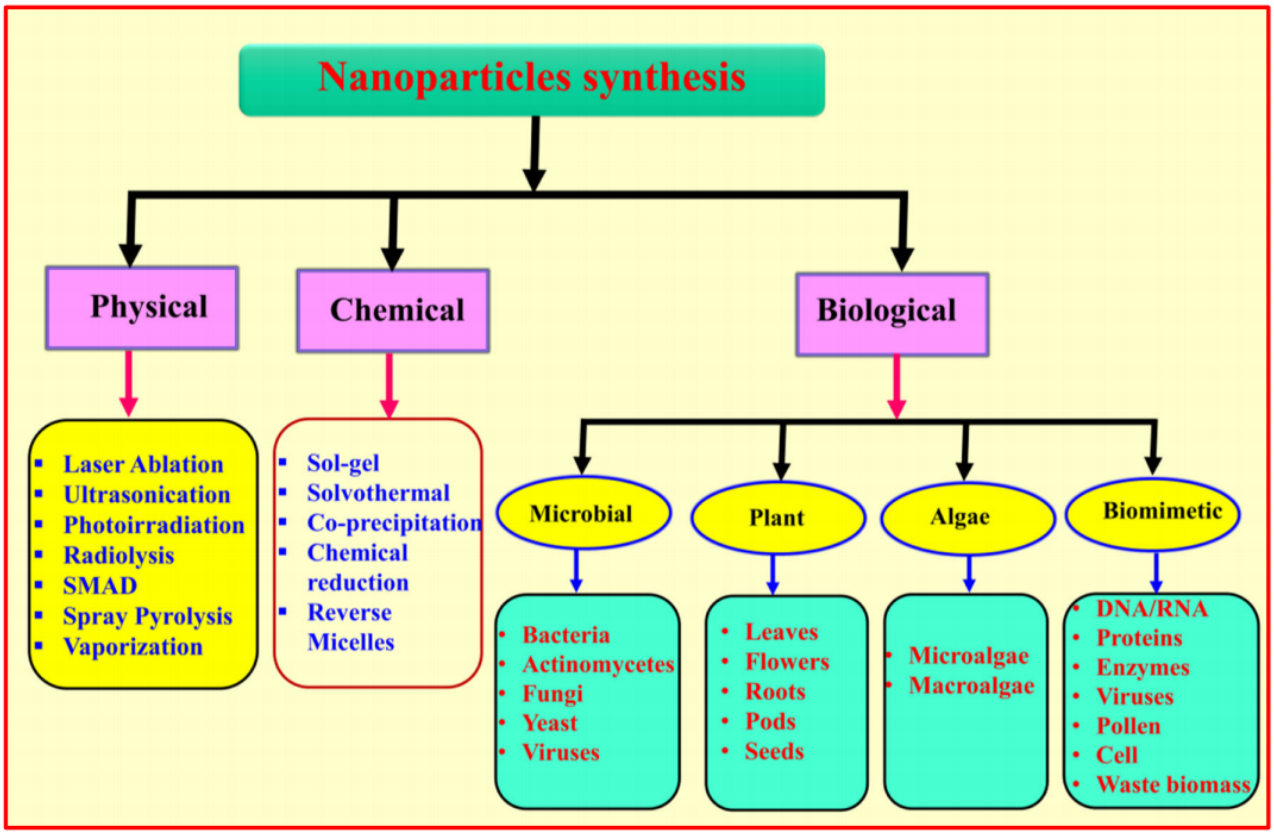

Figure 1. Various methods for the nanoparticle synthesis [32].

nm and the spherical shape of NPs. The band gap of NPs was at 3.5 eV which shows that NiO NPs are active in the visible area, so this is a priceless benefit in comparison to the other photocatalysts like TiO_2 [91]. Diallo et al. reported the NiO NPs synthesis by using Aspalathus linearis. In this research, the optical, structural, and photocatalytic activity of the NPs have been considered. The optical analysis showed that NiO NPs had absorption in UV-visible (518.98 nm), in which the more the temperature increased, the less absorption was observed. For example, the bandgap energies fell to below 4 eV when the sample was heated to around 300-400 °C. Furthermore, the final results depicted that NiO NPs have noticeable photocatalytic activity for the degradation of methylene blue [92].

1.2 Eco-friendly synthesis the NiO NPs

Recently, researchers have shown an increased interest in nanoparticles synthesis using microorganisms. The advantages of such a method far outweigh the disadvantages. Some [93]. merits of this approach are its cost-efficiency, safety, and being ecologically-friendly [63, 95, 96]. Green synthesis is an approach in which the NPs are produced using plants, bacteria, fungi, algae, etc. [63, 65, 97–99]. Ayesha Mariam et al. synthesized NiO NPs using a billing approach by means of Azadirachta indica and Psidium guajava with the aim of considering the anti-cancer activities of these NPs. They figured out that the NiO NPs were spherical with a size range of 17-70 nm [100]. B.T. Sone et al. prepared NiO nanopowders by the use of a green approach of aqueous extracts usage from the red flowers of the plant. Callistemon Viminal's X-ray diffraction analysis delineated the average size of NPs to be 20-35 nm with a crystalline shape. The band gap of these NPs was 3.35 eV. Initially controlled redox processes at ambient temperature were proved by electrochemical impedance spectroscopy. This research finally suggested that plant extract is an eco-friendly, safe, harmless, and economical friendly candidate for NiO NPs preparation which could be utilized in energy storage applications [101].

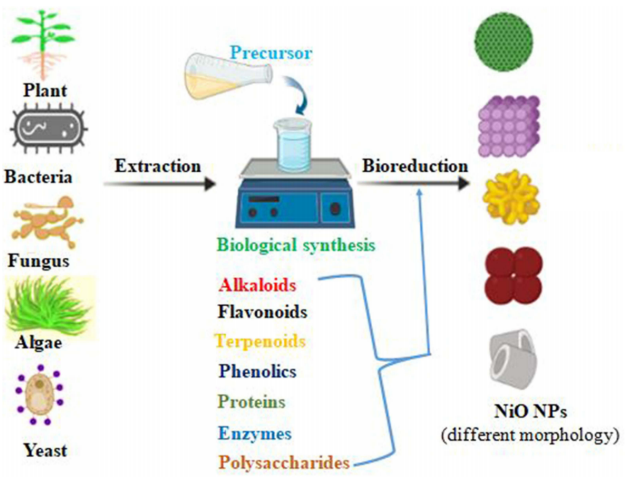

Figure 2. Some examples of the nanoparticle synthesis using plant extracts [94].

2. Green synthesis and different applications of NiO NPs

2.1 Green synthesis of NiO NPs using plant extract

Several parts of the plant like leaf, stem, root, fruit, and seed have been applied for the NPs preparation. Figure 2 depicts a number of nanoparticle synthesis using plant extracts, and Figure 3 showcases synthetic methods of NPs along with their characterizations and applications. NPs synthesis by plant extracts usage also has some merits such as lowcost, biodegradability, being green, lack of intermediate base groups needed and costly equipment usage required, less time-consuming, and it results in products with lesser impurities [102]. One of the most applicable NPs synthesis methods is the NPs fabrication with plant sources because this way leads to nano-based materials with a variety in shape and size [103]. A useful approach in terms of NiO NPs synthesis is a process in which leaves or flowers are added at a basic pH of 12. After shaking the mixture for about 10 min and putting it in a hot air oven as long as 90 min, the mixture color shifted from light to dark green as the reduction process supplemented. Finally, NiO NPs dried and stored for the following usage. The final results of the research clearly showed that NiO NPs, which have been synthesized by using a synthetic nature-friendly method of Calotropis gigantea leave usage, have a significant antibiotic potential against *Escherichia coli* and *Bacillus subtilus* Furthermore, NiO NPs are a convenient choice in a variety of applications because of their proper features such as their magnetic and electrical features. P. Kganyago et al. have recently investigated the innovative and green synthesis of NiO NPs by the use of Monsonia Burkenea's leaves. XRD and HR-TEM analysis proved the preparation of NiO NPs with a 20 nm average size in a spherical shape. The final findings of the research revealed that the NPs have an antibacterial property against gram-negative strains, such as *E.*

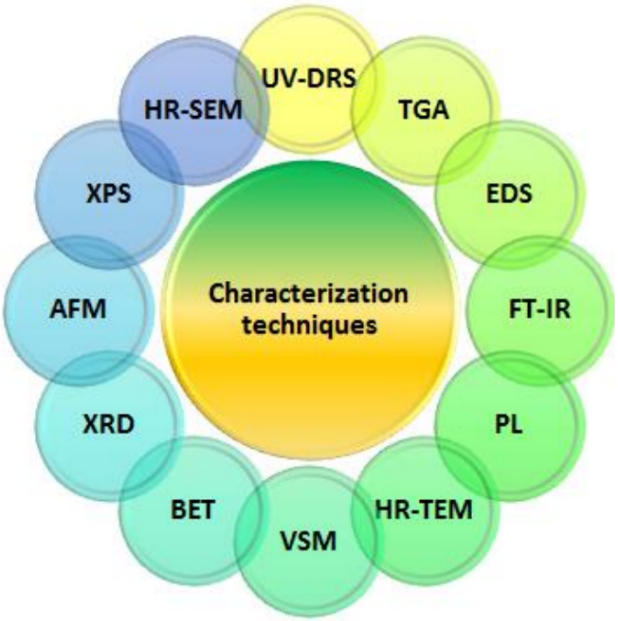

Figure 3. Characterization of nanoparticles [104].

coli and *Pseudomonas aeruginosa*. The researchers also found that although these NiO NPs are less likely to have a very positive proliferation impact on carcinogenic cells, they could be preferably utilized as drug carriers for cancer treatment in the human body [106]. Ali Talha Khalil et al. prepared the NiO NPs by using sageretia tea (Osbeck.) leaves and considered their biological effects. The size of NPs observed by XRD and HR-SEM/TEM analysis was 18 nm and their shape was also spherical. This study has investigated the effects of these NPs on six various gram-positive and gram-negative bacterial strains and finding their toxicity, biocompatibility, antioxidant, and enzyme inhibition features. The results showed that NiO NPs revealed improved antibacterial activities and moderate antioxidant properties. The synthesized NiO NPs also indicated modest enzyme inhibition activities and relatively non-toxicity to human cells [107]. To determine the supercapacitor electrodes applications, Manab Kundu et al. prepared NiO NPs by a green and novel approach to Hydrangea paniculata flower extracts usage. The average size of the NPs was 33 nm. The electrode-based NPs depicted high capacitance, and, interestingly, the NiO NPs electrodes showed remarkable cycling stability. The mentioned excellent electrochemical proficiency was related to the nano-scale of particles, which eases migration over the rapid charge-discharge procedure, reduces the diffusion path length of ions and electrons and finally enhances the efficient electrochemical application of electroactive material [108].

Singh et al. present the structural and crystallinity information of NiO NPs, as depicted in Fig. 4a. The XRD pattern confirms NiO nanoparticle formation, displaying intense reflection peaks at $2\theta = 37.3°$, 43.3°, 62.9°, 75.6°, and 79.6°. The pattern aligns with JCPDS card number 47–1049, indicating a face-centered cubic structure with an average crystallite size of 27.7 nm. BET analysis in Fig. 4b reveals a specific surface area of 16.71 m^2g^{-1}, an average pore size of 1.2 nm, and a total pore volume of 0.32 cm^3g^{-1}, suggesting mesoporous characteristics with a uniform pore size distribution [105].

Subsequently, Singh et al. utilized HRTEM analysis to examine the morphology and structure of NiO NPs. Fig. 5 (a-c) confirms the formation of agglomerated quasi-spherical particles. In Fig. 5d, sharp diffraction spots in the SAED pattern indicate a polycrystalline nature, consistent with XRD results.

Behera et al. demonstrate the synthesis of NiO NPs, confirmed through XRD. The XRD pattern in Fig. 6(a) shows no additional impurity peaks, indicating the crystalline nature of the NiO NPs. The intense diffraction peaks observed at 37°, 43°, 63°, 75°, and 79° correspond to crystal planes (111), (200), (220), (311), and (222), respectively, consistent with the diffraction patterns of NiO NPs. Additionally, Scherrer's equation was used to determine that the average crystallite size of the NiO NPs was 22.8 nm. The FE-SEM image in Fig. 6(b) illustrates the surface morphology of the NiO. Most of the particles appear spherical, with some exhibiting a rod-like shape. The particle size ranges from 40 to 100 nm. Fig. 6(c) presents the EDX spectrum, confirming the elemental composition of the NiO nanoparticles. Lastly, the TEM image in Fig. 6(d) depicts spherical and rod-shaped NiO nanoparticles with diameters ranging from 10 to 80 nm [109].

As the green synthesis methods are now well-liked and popular among scientists, a considerable amount of literature has been published on the synthesis of NiO NPs by the use of different plants, and their details are given in Table 1.

2.2 Antibacterial applications of NiO NPs

Metallic oxide nanoparticles can be a suitable antibacterial tool. Biological features of metallic oxides NPs are related to their nano-based size and the high possibility of the NPs to interact with bio-objects [130, 131]. Over the last few years, more researchers have studied the antibacterial impacts of different metallic oxides on varied bacterial strains. As there have been more researches that focused on antibacterial applications of NiO NPs, we have considered reported antibacterial applications of NiO NPs on different bacterial strains such as *Streptococcus pneumoniae*, *Staphylococcus aureus*, *E. coli*, *E. hermannii* [132], *P. aeruginosa* [133], *B. anthracis*, *B. subtilus*, *Klebsiella pneumoniae*, *Enterobacter*

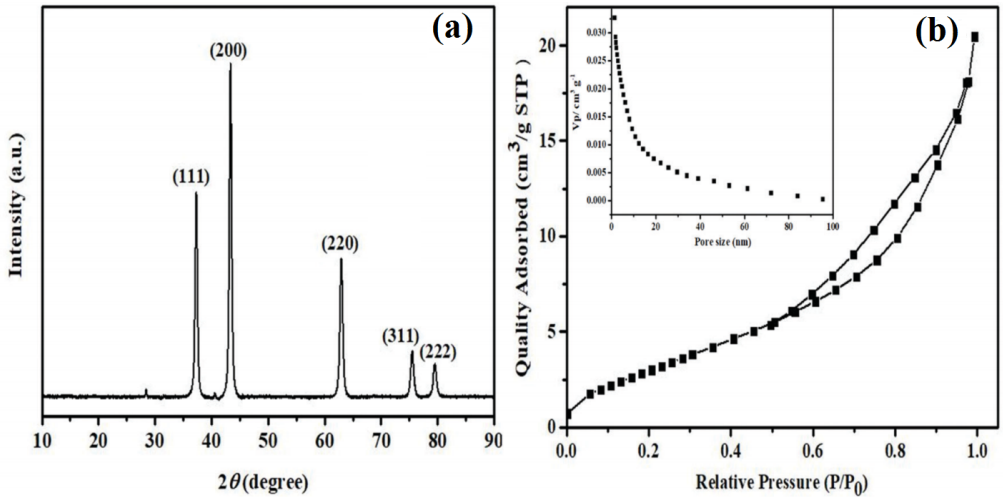

Figure 4. (a) XRD pattern, **(b)** BET (inset: pore size distribution) of the NiO NPs [105].

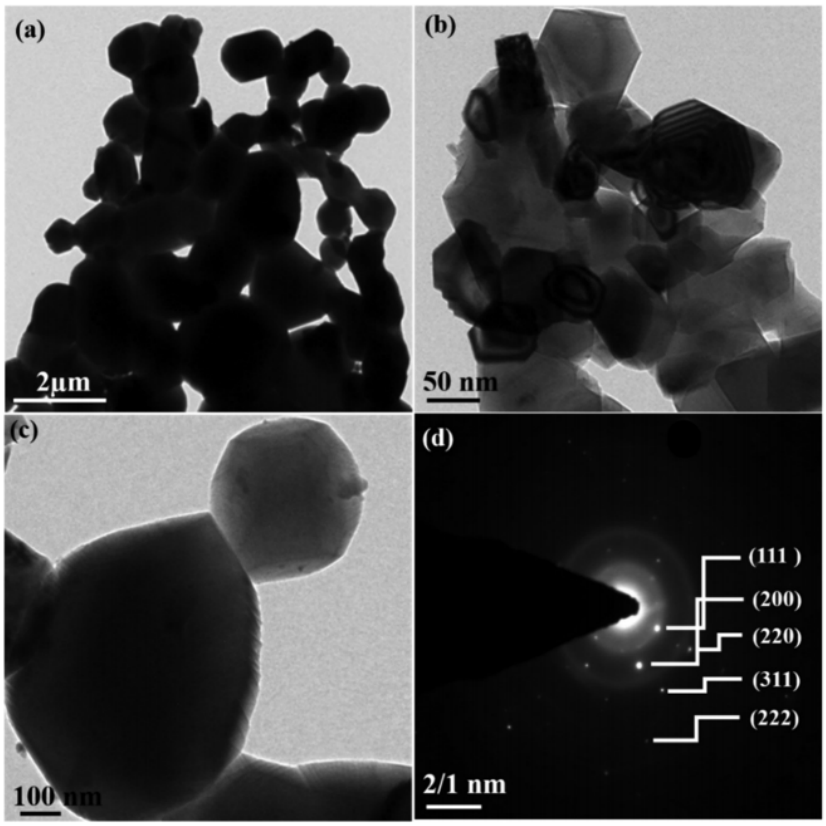

Figure 5. HRTEM images **(a–c)** and **(d)** SAED pattern of NiO NPs [105].

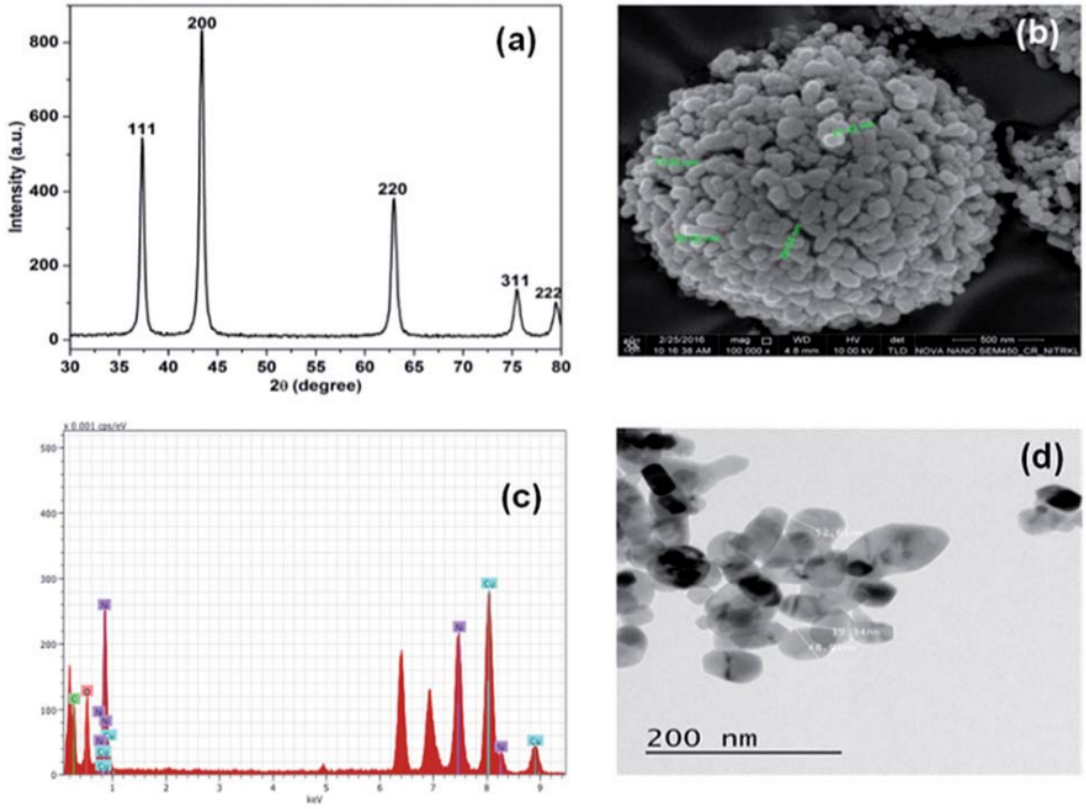

Figure 6. **(a)** XRD Pattern, **(b)** FESEM Image, **(c)** EDX and **(d)** TEM Image of NiO NPs [109].

Table 1. Plant mediated synthesis of NiO NPs.

No.	Plant(family)	Part taken for extraction	Size (nm)	Shape	Ref.
1	Mytaceae	Red flower	20–35	Quasi reversible with films	[101]
2	Rutaceae	Leaf	8–10	Spherical	[91]
3	Acacia tree	Sap	34	Cubic-spherical	[110]
4	Fabaceae	Flower	6–10	Crystalline	[92]
5	Apocynaceae	Leaf	20–40	Spherical	[93]
6	Moringaceae	Leaf	5–10	Spherical and slightly agglomerated	[111]
7	Lythraceae	Peel	3	Spherical and face-centered cubic	[112]
8	Geraniceae	Leaf	20–25	Crystalline and spherical	[106]
9	Rhamnaceae	Leaf	18	Spherical	[107]
10	Hydrangeaceae	Flower	33	Cubic crystalline	[108]
11	Tamaricaceae	Flower	10–14	Crystalline	[113]
12	Euphorbiaceae	Cassav waste	5–10	Hexagonal and sphere	[114]
13	Mytraceae	Leaf	10–20	Cubic	[115]
14	Apocynaceae	Leaf	20	Crystalline	[116]
15	Fabaceae	Root	10–15	Face-centered cubical structure	[117]
16	Rutaceae	Leaf	15–23	Cubic crystalline	[118]
17	Sapindaceae	Rambutan peel waste	50–100	Crystalline	[119]
18	Mytraceae	Leaf	44	Cubic and crystalline	[100]
19	Limonia accidissima	Fruit	23	Spherical	[120]
20	Plectranthus amboinicus	Leaf	100	Spherical	[121]
21	Jatropha gossypiifolia	Leaf	30–40	Rod shape	[122]
22	Ageratum conyzoides	Leaf	40–60	Nanoflakes	[122]
23	Senna auriculata	Flower	20	Spherical	[123]
24	Nigella sativa	Seed	10–50	Spherical and oval	[124]
24	Olive	Leaf	30–65	Spherical	[125]
25	Lantana camara	Flower	40–50	Oval	[126]
26	Orange	Leaf	26–64	Spherical and like oval	[127]
27	Piper betle	Leaf	20–40	Spherical	[128]
28	Aloe vera	Leaf	–	–	[129]

aerogenes [134], *S. pyogenes*, and *Proteus vulgaris* [135]. The other synthesized NiO NPs with different antibacterial capabilities are listed in Table 2.

2.3 Photocatalytic applications of NiO NPs

Organic pollutants, which are released from industrial sectors, might be the biggest threat to our environment as they are highly poisonous and harmful to a variable microorganism [148–151]. The hazardous pollutants degradation has gained more attention, so photocatalytic degradations in the presence of different light sources is said to be an applicable factor for organic pollutant degradation [60, 152–158]. Nano-based semiconductors like CuO, ZnO, TiO_2, etc. have been the most sufficient photocatalysts for organic pollutants degradation [159–165].

Ahmad Khan and colleagues elucidate semiconductor photocatalysis, wherein photons with energy equal to or greater than the bandgap energy of the photocatalyst are required. Upon light irradiation on the nanoparticles (NPs), electrons (e^-) in the valence band (VB) are excited to the conduction band (CB), creating positively charged holes (h^+) in the VB. The conduction band electrons (e^-) react with O_2 species as electron acceptors, forming the strong oxidizing agent superoxide anion radical ($\cdot O_2^-$). Similarly, h^+ in the VB reacts with H_2O molecules, generating hydroxyl radicals ($\cdot$OH). Both radicals are responsible for dye degradation [166]. The degradation mechanism is illustrated in Fig 7.

Among different nanoparticles which are used as photocatalysts, NiO NPs have been utilized for the degradation of various dyes, such as Methylene blue [136], Methyl orange [167], Evans blue [168], Rhodamine B [169], Rose Bengal [134], Reactive black [170], Congo Red [171], Violet dye [172], Acid scarlet dye [173], and Trypan Blue [144], as well as different organic pollutant degradations

Table 2. Antibacterial applications of NiO nanoparticles.

Synthetic approach	Bacterial strain	Zone of Inhibition	Morphology	Crystallite Size (based on XRD)	Reference
Hot plate combustion method	S. pneumoniae	12 mm	Cubic and spherical shape	8–10 nm	[132]
	S. aureus	16 mm			
	E. coli	11 mm			
	E. hermannii	6 mm			
Hydrothermal	S. aureus E. coli	–	Disk like	NiO-300 = 5nm	[136]
				NiO-400 = 6.8 nm	
				NiO-500 = 12.7 nm	
Thermal decomposition	E. coli	12 mm	Rod like	60 nm	[133]
	P. aeruginosa	11 mm			
	S. aureus	10 mm			
Hot combustion reaction	S. pneumoniae	22.4 mm	Stick like	50 nm	[134]
	B. anthracis	25.4 mm			
	K. pneumoniae	23.2 mm			
	E. aerogenes	27.5 mm			
	B. subtilus	–			
Biosynthetic method	S. aureus	NiO: 10 mm	Hexagonal crystal phase And oblong shape	12 nm	[137]
		NiO-350: 11 mm			
		NiO-450:15 mm			
	E. coli	NiO: 11 mm			
		NiO-350: 13 mm			
		NiO-450: 15 mm			
Pulsed laser ablation	S. aureus	12.6 ± 0.57 mm	Spherical	2–21 nm	[138]
	E. coli	14.3 ± 1.15 mm			
Green method using Eucalyptus globulus leaf extract	E. coli-60	17 mm	–	10–20 nm	[115]
	E. coli-52	17 mm			
	P. aeruginosa-48	15 mm			
	P. aeruginosa-64	14 mm			
	Methicillin sensitive S. aureus-06	15 mm			
	Methicillin sensitive S. aureus-02	13 mm			
	Methicillin resistant S. aureus-10	15 mm			
	Methicillin resistant S. aureus-31	14 mm			
Sol-gel	E. coli	10 mm	Cubic and irregular shape	30 nm	[139]
	K. pneumoniae	0			
	P. vulgaris	0			
	S. mutans	8 mm			
	B. subtilus	12 mm			
	S. aureus	11 mm			

Continued on next page

Table 2. Antibacterial applications of NiO nanoparticles. (Continued)

Synthetic approach	Bacterial strain	Zone of Inhibition	Morphology	Crystallite Size (based on XRD)	Reference
Co-precipitation	E. coli	Maximum zone of inhibition = 15 mm	Cubic and spherical shape	30 nm	[140]
	K. pneumoniae				
	B. subtilus				
	S. aureus				
Green synthetic routes using Calotropis gigantea	E. coli	–	–	< 60 nm	[93]
	B. subtilus	–			
Green method using Monsonia burkenea leaf extract	E. coli	–	Spherical shape	25 nm	[106]
	P. aeruginosa				
	Enterococcus faecalis				
	S. aureus				
Microwave-assisted route	P. aeruginosa	13.67 ± 0.29 mm	Spherical shape	20 nm	[141]
	E. coli	15.37 ± 0.17 mm			
	K. pneumoniae	13.80 ± 0.29 mm			
	S. aureus	14.67 ± 0.17 mm			
Co-percipitation method	E. coli (5 mg/ml) RT	14 mm	–	–	[142]
	E. coli (3 mg/ml) RT	12 mm			
	E. coli (5 mg/ml) RT 60	16mm			
	E. coli (3 mg/ml) RT 60	15 mm			
	S. aureus (5 mg/ml) RT	11 mm			
	S. aureus (5 mg/ml) RT	7mm			
	S. aureus (5 mg/ml) RT 60	15 mm			
	S. aureus (3 mg/ml) RT 60	18 mm			
Sol-gel	E. coli	–	Grainy	30–80 nm	[143]
	S. aureus				
Co-precipitation	S. aureus	9.8 mm	Spherical and cubical shape	40 nm	[135]
	S. pyogenes	9.0 mm			
	B. subtilus	8.5 mm			
	P. aeruginosa	8.3 mm			
	K. pneumoniae	8.1 mm			
Combustion method	K. aerogenes	–	Bunsenite form of simple cubic system	28 nm	[144]
	E. coli	–			
	P. aeruginosa	4.12 ± 0.43			
	S. aureus	–			
Microwave-Assisted	S. aureus	24 ± 0.9	Spherical	15–16 nm	[135]
	P. vulgaris	19 ± 0.9			
	E. coli	25 ± 0.3			
	P. aeruginosa	24 ± 0.6			

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Table 2. Antibacterial applications of NiO nanoparticles. (Continued)

Synthetic approach	Bacterial strain	Zone of Inhibition	Morphology	Crystallite Size (based on XRD)	Reference
Green method using Rhamnus virgata extract	S. aureus	20 mm	Spherical	24 nm	[145]
	B. subtilus	29 mm			
	P. aeruginosa	16 mm			
	K. pneumoniae	14 mm			
	E. coli	27 mm			
Co-precipitation	E. coli	–	Spherical and cubic	12–16 nm	[146]
	S. aureus	–			
Electrospray method	E. coli	–	–	20 nm	[147]
Biogenic synthesis	Proteus	1.1 cm	Spherical	–	[121]
Green method using Jatropha gossypiifolia	B. subtilus	4.1 mm	Rod	30–40 nm	[122]
	B.cereus	3.6 mm			
	S. aureus	6.2 mm			
	Klebsiella	3.5 mm			
Green synthesis	B. subtilus	21 mm	Spherical	3.92 nm	[123]
	S. aureus	20 mm			
	P.aeruginosa	21 mm			
	E. coli	23 mm			
Solution combustion	E. coli	Nio-1: –	Oval	Nio-1: 14.3 nm	[126]
		Nio-2: –			
		Nio-3: 12.33 mm			
	M. luteus	Nio-1: 10.67 mm		Nio-2: 20 nm	
		Nio-2: 11.67 mm			
		Nio-3: 10.33mm			
	S. aureus	Nio-1: 11.67 mm		Nio-3: 26 nm	
		Nio-2: 15 mm			
		Nio-3: 13.33 mm			
Green synthesis	E. coli	25 mm	Spherical and like oval	20.34–27.04 nm	[127]
	S. aureus	32 mm			
Green synthesis	E. coli	20 mm	Spherical	26.27 nm	[128]
	B. subtilis	25 mm			
Green synthesis	E. coli	18 mm	–	22 nm	[129]
	P. multocida	19 mm			
	B. subtilis	23 mm			
	S. aureus	25 mm			

like NO destruction [174], 4-chlorophenol [132], Phenol [175], Acetaldehyde [176], Acid fuchsine [177], Whole of the reported photocatalytic applications of NiO NPs in organic pollutants degradation listed in Table 3.

2.4 Catalytic applications of NiO NPs

Biodegradable, green, and costless metallic oxides NPs, such as ZnO, CuO, NiO, MgO, and CdO, have been comprehensively surfed as selective catalysts in C-C and C-heteroatom reactions [179]. These nanoparticles and their performance in multicomponent reactions (MCRs) have gained considerable attention in the last decade [180–187]. A Proficient and suitable property of MCRs over the conventional synthetic approaches is that the MCRs are one spot, cheaper as well as being energy and time-sufficient [188, 189]. Some articles have presented the procedure

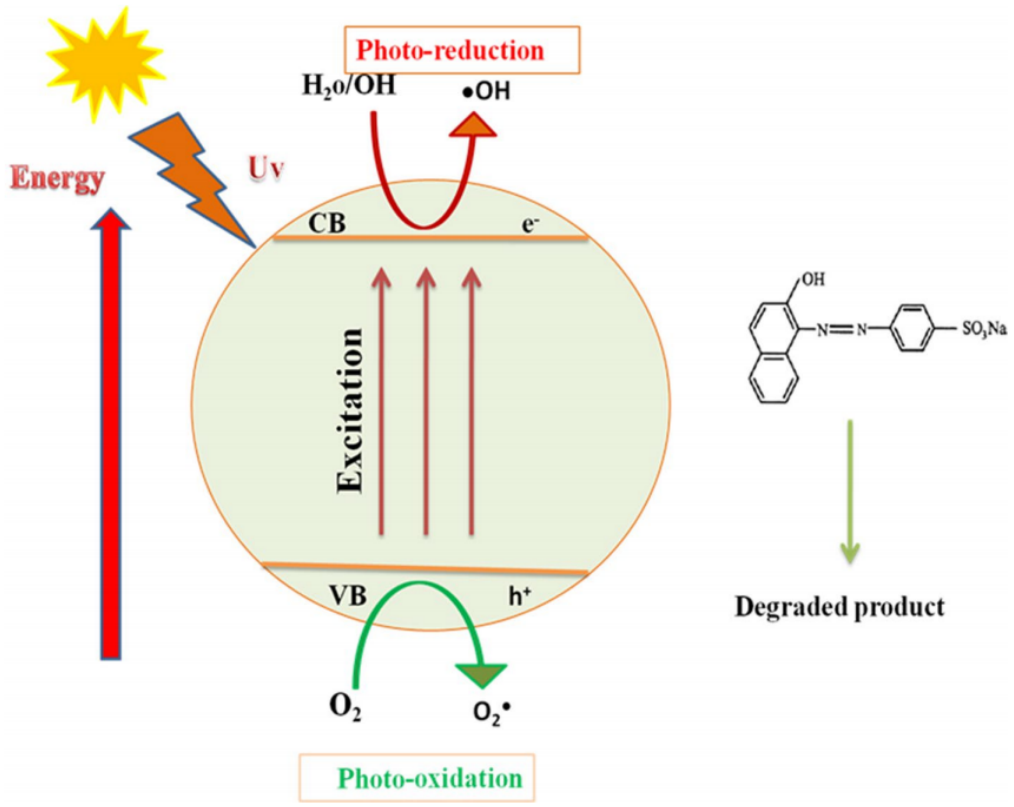
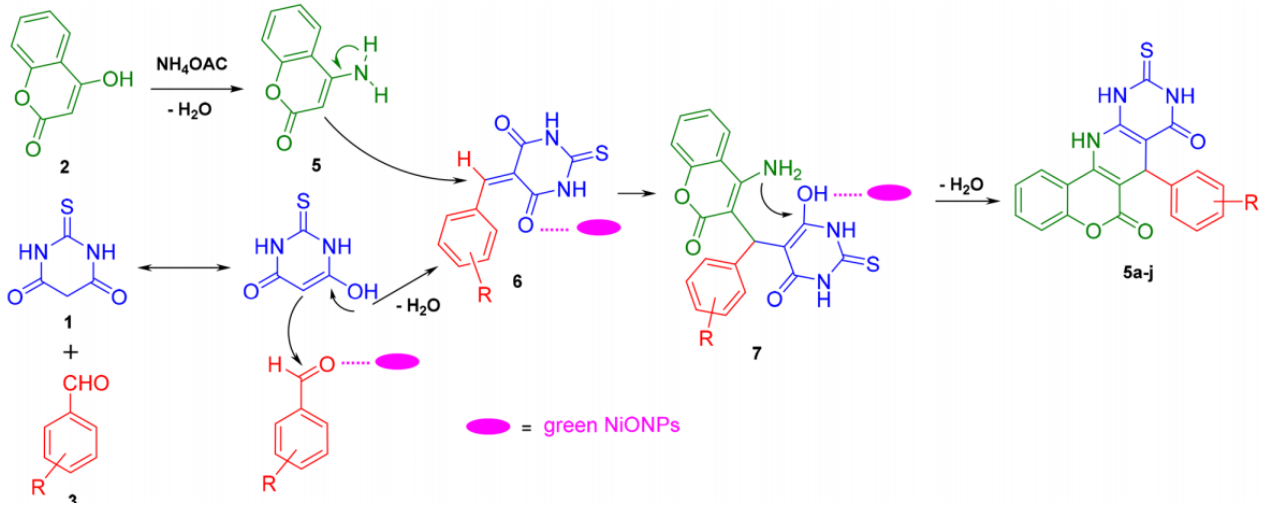


Figure 7. Illustrative mechanism of photocatalytic degradation using NiO NPs [166].

of various reported metallic oxides nanoparticles catalyzed by different MCRs for vital organic compounds synthesis. Herein, among the mentioned metallic oxide NPs, NiO NPs catalytic applications in MCRs have been considered.

Scheme 1 demonstrates a catalyzed sequential multicomponent reaction route proposed by Moavi and colleagues. The enol form of thiobarbituric acid (1) undergoes Knoevenagel condensation with the catalyst-activated carbonyl of an aryl aldehyde (3) to give the α,β-unsaturated molecule 6, followed by dehydration. Subsequently, 4-hydroxycoumarin (2) reacts at 100 °C with in situ generated ammonia from ammonium acetate (4) to form 4-aminocoumarin (5). Michael's addition of the enamine (5) to the α,β-unsaturated carbonyl acceptor of 6 yields the intermediate 7. After the elimination of H_2O, an amino group attacks the carbonyl group with NiO catalyst to cyclize the intramolecular ring to give the desired products 5 in an aqueous medium. The addition of algal NiO NPs to the process enhances the electrophilicity and facilitates the attack of nucleophilic groups due to their acidity. In addition, the Lewis acid NiO nanocatalyst improves the reactivity of the organic material and the stability of the intermediates [178].



Scheme 1. Proposed mechanism for pyridopyrimidine derivatives synthesis using the NiO NPs [178].

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Table 3. Photocatalytic efficiency of NiO nanoparticles in the degradation of organic pollutants.

Synthetic approach	Pollutant type	Pollutant concentration	Morphology	Time	Light source	Catalyst concentration	Crystallite Size based on XRD	Degradation efficency	Ref.
Thermal decomposition	NO destruction	1 mg/L	Hexagonal Quasi-hexagonal	30 min	High pressure Mercury lamp	–	400 nm	21%	[174]
hot plate combustion method using Aegle marmelos plant	4-Chlorophenol	50–250 mg/L 100 mg/L	pure face centered cubic phase and single crystalline, spherical	30 min	Low pressure Mercury lamp	30 mg	8–10 nm	87% over 120 min	[132]
Hydrothermal	Methylene blue	15 mg/L	Disk shaped and spherical	20 min	UV-Visible	1 mg	5.1 nm	98.7% (Blue dye) 94–97.6% (other mixture)	[136]
Thermal decomposition	Methylene blue	10 mg/L	Crystalline	30 min for NiO-400 and 90 min for NiO-300	UV-Visible Green light	5 mg	14 nm 6–10 nm at 300–400 °C	46% for NiO-300 and NiO-400 was 3 times as that of NiO-300	[92]
Thermal-decomposition (A), sol-gel (B), hydrothermal (C), and emulsion (D) nano-reactors	Methyl orange	40 mg/L	Spherical A: flower like shape B: shape of regular nano-woods C: plate-like D: spherical	120 min	UV-Visible Green light	20 mg	100 nm	86%	[167]
Sol-gel method	Phenol	100 mg/L	Spherical	60 min	UV laser	100 mg	6–10 nm	97%	[175]
Chemical precipitation method	Methylene Blue	1×10^{-5} M	Oval shape	150 min	Solar light	0.031 g	64 nm	–	[190]
Combustion method	Methyl orange	0.1 mM	Octahedral shape	–	UV	0.0125 mM	50 nm	–	[191]
Microwave-assisted method	Evans blue	–	Spherical	120 min	Solar light	20 mg	24 nm	88%	[168]
Thermal decomposition	Methylene Blue	5 mg	Coral-like	90 min	Solar light	1 mg	2–3 nm	$\geq$ 97%	[192]
Thermal decomposition	Rhodamine B		Sheet-like	120 min	Osram ultraviolet lamp	–	10 nm	80%	[169]

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Table 3. Photocatalytic efficiency of NiO nanoparticles in the degradation of organic pollutants. (Continued)

Synthetic approach	Pollutant type	Pollutant concentration	Morphology	Time	Light source	Catalyst concentration	Crystallite Size based on XRD	Degradation efficency	Ref.
Thermal decomposition	Methylene Blue	25 mg/L	Rod-like	135 min	250 W highpressure mercury lamp (GYZ-250)	15 mg/L	20 nm	99%	[193]
Co-percipitation	Acetaldehyde	8 M	Rime-like	120 min	UV	6.5 ± 0.1 mg	–	98%	[176]
Solvothermal	Methylene Blue	20 mg/L	Rocksalt cubic face	140 min	300 W Xe arc Lamp	0.1 g	5–10 nm	100%	[194]
Hot combustion reaction	Rose Bengal	10 mg/L	Stick like	100 min	UV	50 mg	50 nm	–	[134]
Calcination method	Phenol	0.02 mg/L	Spherical NiO Hexagonal NiO	100 min	UV	0.22 mg	5–10 nm 22–27 nm	82% $\geq$ 69%	[195]
Thermal decomposition	Reactive black 5 Methylene Blue	3×10^{-5} M	Simple cubic structure	60 min 5 h	Tungsten halogen lamps	30 mg	–	RB-5 = 87.2% MB = 70.2%	[170]
solid-state thermal decomposition	Methylene Blue (MB)	5 mg/L	Spherical	120 min	UV	20 mg	20–50 nm	98%	[196]
Co-percipitation	Methylene Blue	–	Face cubic	270 min	UV	–	10–28 nm	80%	[197]
Solvothermal	Acid fuchsine	20.0 mg/L	Rose-like	30 min	300 W medium-pressure mercury lamp	1.0 g L^{-1}	3–4 nm	100%	[177]
Hydrothermal	Congo Red	10 mg/L	Cube shape	30 min	Mercury lamp	0.1 g	20 nm	98%	[171]
Percipitation	Violet dye	–	Sphere like	50 min	UV	50 mg	20 nm	98%	[172]
Hydrothermal	Methylene Blue	10 mg/L	Face centered cubic	60 min	UV	1.0 g	$\leq$ 100 nm	$\leq$ 47%	[198]
Thermal decomposition	DFB	75 mg/L	Spherical and face-centered cubic	150 min	Mercury lamp	10 mg	50 nm	96.4%	[199]
Pulsed laser deposition	Rhodamine B	10 mg/L	Thin films	60 min	Xe lamp	–	8–16 nm	–	[200]
Thermal decomposition	Acid scarlet	20 mg/L	Wire like	60 min	15 W ultraviolet (UV) lamps	10 mg	8–12 nm	–	[173]
Green synthetic routes using Calotropis gigantea	Methylene Blue	0.08 mM	–	–	Sun light	0.01 g	$<$ 60 nm	98.6%	[93]

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Table 3. Photocatalytic efficiency of NiO nanoparticles in the degradation of organic pollutants. (Continued)

Synthetic approach	Pollutant type	Pollutant concentration	Morphology	Time	Light source	Catalyst concentration	Crystallite Size based on XRD	Degradation efficency	Ref.
Co-precipitation	Congo red dye	20 mg/L	Cubic and spherical shape	30 min	UV	5 mg	30 nm	84%	[140]
Green method using Monsonia burkenea leaf extract	Methylene Blue	20 mg/L	Spherical shape	75 min	15 W LED	20 mg	25 nm	50%	[106]
Microwave-assisted method	Evans blue	–	Spherical shape	150 min	Sunlight	20 mg	20 nm	91%	[141]
Combustion method	Trypan Blue	100 mg/L	Bunsenite form of simple cubic system	150 min	UV	10 mg	40 nm	98%	[144]
Green combustion	Methylene Blue	1 mM	Spherical	90 min	UV	40 mg	23 nm	99.6%	[120]
Green synthesis	Methylene Blue	10 g/L	Spherical	90 min	sunlight	0.2 g/L	3.92 nm	97%	[123]
solution combustion	Methylene Blue	10 mg/L	oval	60 min	UV	30 mg	14.3–26 nm	96–98%	[126]
Green synthesis	Reactive Red 141	50 mg/L	Spherical	100 min	Visible Light	15 mg	26.27 nm	99%	[128]
Green synthesis	methylene blue	50 mg/L	triangular	13 min	UV	10 mg	7–20 nm	98%	[201]
Green sonochemical	Safranin	1 mg/L	Spherical	100 min	UV	5 mg	9.38 nm	92.75%	[202]
Green sonochemical	Methyl orange	1 mg/L	Spherical	100 min	UV	5 mg	9.38 nm	98%	[202]
Biosynthesis	Rhodamine-B	20 mg/L	Spherical	90 min	Visible Light	20 mg	12 nm	85%	[203]
Precipitation	Rhodamine B	0.025 mg/L	Spherical	16 min	Solar light	0.5 mg	–	98	[204]
Biosynthesis	Methylene Blue	20 mg/L	flowerlike	270 min	UV	20 mg	18.4 nm	86	[205]
Biosynthesis	Malachite Green	20 mg/L	flowerlike	210 min	UV	20 mg	18.4 nm	98	[205]

2.4.1 Synthesis of pyrroles

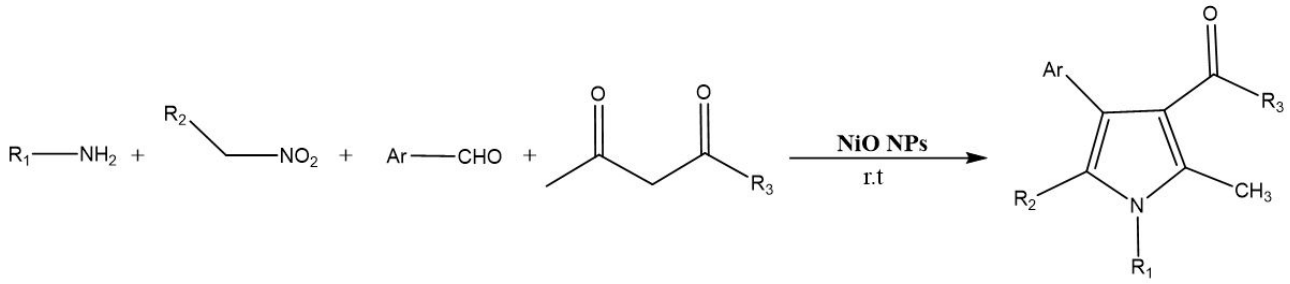
Bhalchandra M. Bhanage et al. reported the synthesis of substituted pyrroles via amines, aldehydes, nitroalkanes, and 1,3-dicarbonyl compounds in one pot synthesis reaction which catalyzed by NiO NPs at ambient temperature. They figured out that NiO NPs could be a trustable catalyst, due to having suitable activity and features, for a broad range of different substrates with desirable yields of focused products along with catalyst reusability [206] (Scheme 2).

2.4.2 Synthesis of amidoalkyl napthol derivatives

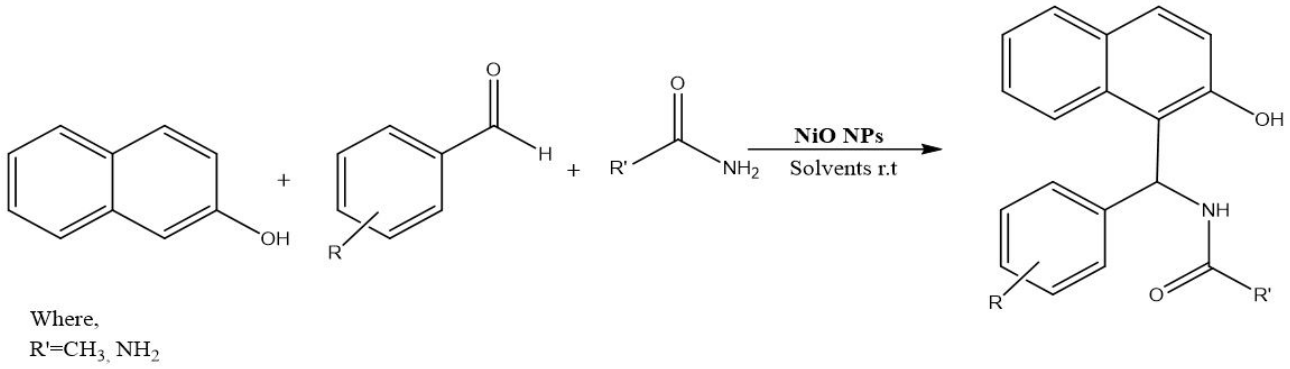
Ratiram G. et al. reported the synthesis of amidoalkyl napthol derivatives by means of applicable and appropriate NiO NPs catalyst and using condensation reaction of urea, β-naphthol, and aldehydes. The advantages of this synthetic method were simple workup, solvent free usage, and highly reusability of its catalyst [207] (Scheme 3).

2.4.3 Synthesis of varied spiro and condensed indole derivatives

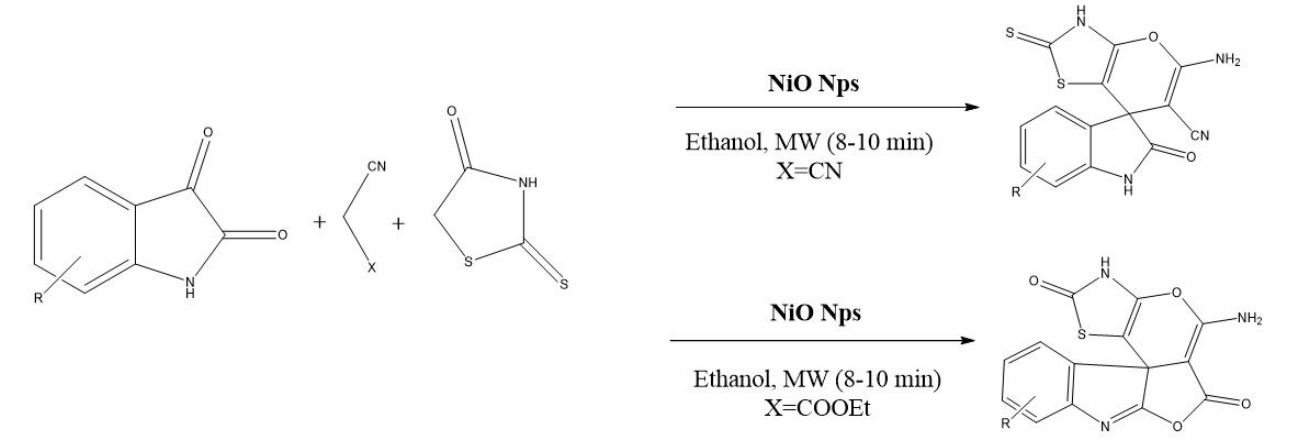
Another study investigated the synthesis of different spiro and condensed indole derivatives by means of substituted 1H-indole-2,3-diones, 2-thioxo-4-thiazolidinone, and methylene as a reagent as well as highly efficient NiO NPs as a catalyst. The reaction was engaged in Michael and Knoevenagel condensation (Scheme 4). Work simplicity,



Scheme 2. Synthesis of pyrroles by using NiO NPs.



Scheme 3. Synthesis of amidoalkyl naphthols derivatives by using NiO NPs.



Scheme 4. Synthesis of spiro and condensed indole derivatives.

easy-to-handle, and convincing yields make the catalytic approach more applicable [208].

2.4.4 Synthesis of 2-substituted-4, 6- diarylpyrimidines

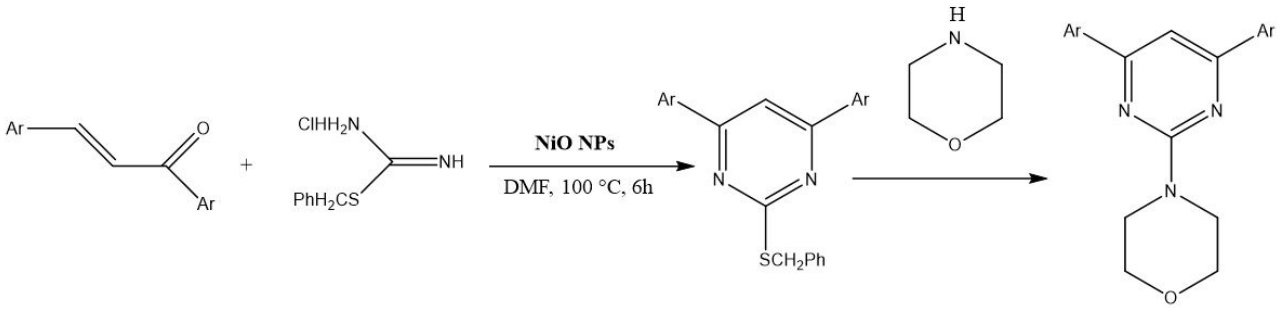
A trusted and economical-friendly approach has recently been proposed for the synthesis of 2-substituted-4, 6-diarylpyrimidines in the one-pot reaction between S-benzylthiouronium chloride, morpholine, and varied substituted chalcones by means of NiO NPs as a reproducible and costless catalyst. The reaction could improve the catalytic synthesis of 2-(4-morpholinyl)-4,6-diarylpyrimidines with a proper yield and short reaction time [209] (Scheme 5).

2.4.5 Synthesis of 2-(1H-tetrazol-5-yl) acrylonitrile derivatives

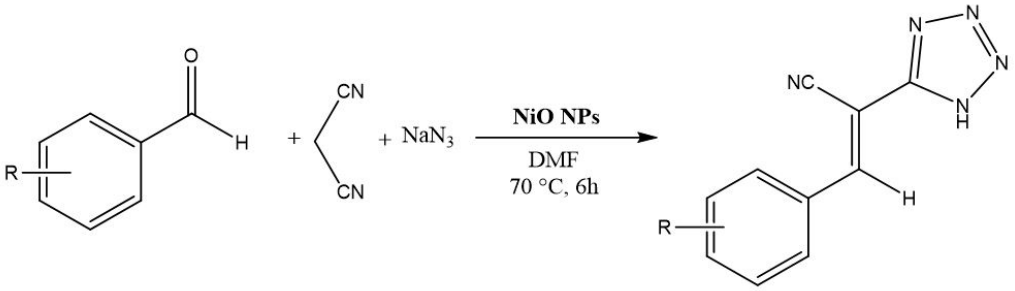
A novel and rapid procedure of 2-(1H-tetrazol-5-yl) acrylonitrile derivatives synthesis has recently been fulfilled using one pot MCRs of malononitrile, aldehydes, and sodium azide which required NiO nanoparticles as a reproducible catalyst (Scheme 6). Safaei-Ghomi et al. stated that this method has a suitable features such as easy to work, shorter reaction time in comparison with traditional methods, and high yields as well as reusability of catalyst [210].

2.4.6 Synthesis of 3,4-dihydropyrimidin-2(1H)-ones

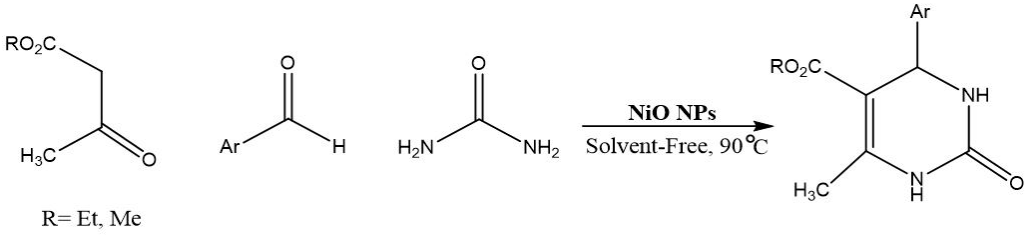
Khashaei and coworkers discovered a method for producing 3,4-dihydropyrimidin-2(1H)-ones (DHPMs) using a NiO NPs catalyst. This involved a condensation reaction between urea, ethyl/methyl acetoacetate, and an arylaldehyde. The technology offers several benefits, including the production of pure NiO nanoparticles through a unique route and their utilization as a low-toxic, affordable, and efficient catalyst for DHPM synthesis [211] (Scheme 7).
Scheme 8 illustrates the reaction catalyzed by NiO nanoparticles to produce the target product. The process initiates by activating arylaldehydes and β-ketoester, leading to the formation of enol-tautomeric forms. Subsequent Knoevenagel condensation results in the formation of intermediate α,β-unsaturated ketones. Activated urea molecules undergo Michael-type addition, yielding an open-chain ureide intermediate, which cyclizes to form the target DHPMs, six-membered heterocyclic compounds. NiO accelerates



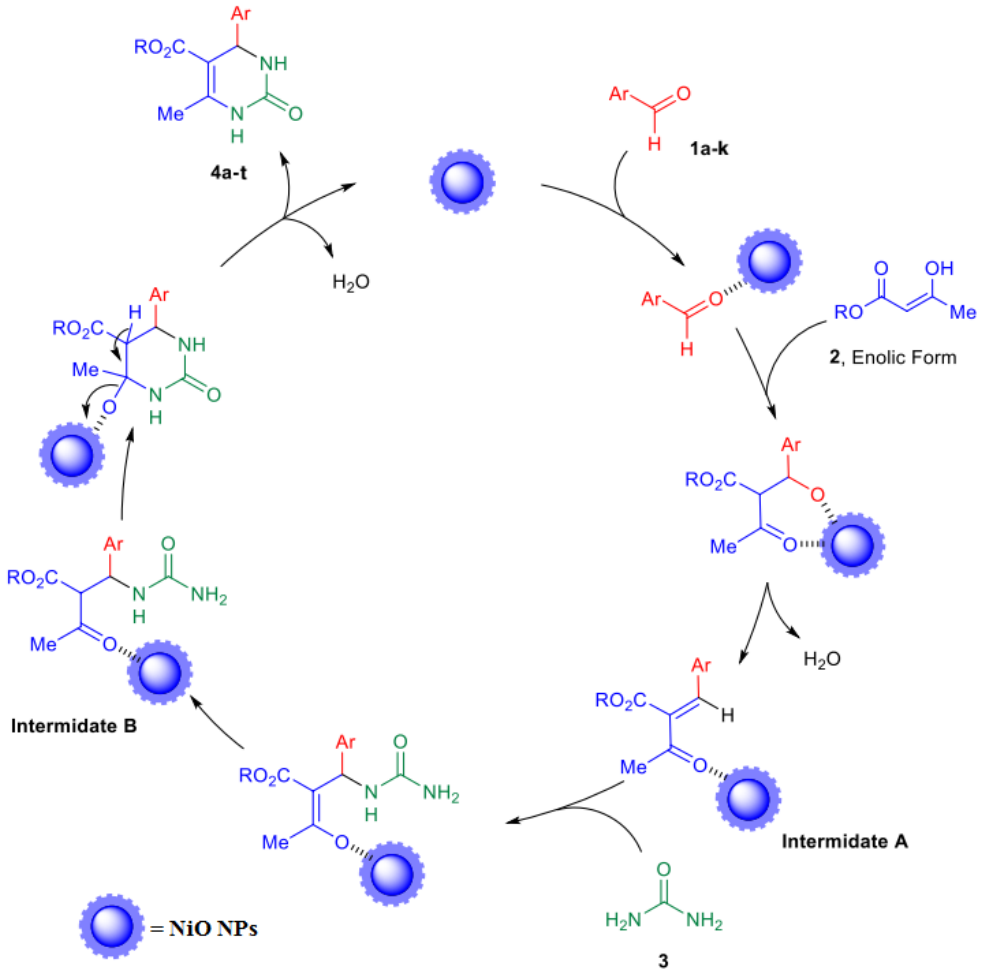
Scheme 5. Synthesis of 2-(4-morpholinyl)-4,6-diarylpyrimidines by means of NiO nanoparticles.



Scheme 6. Synthesis of 2-(1H-tetrazol-5-yl) acrylonitrile derivatives using NiO NPs.



Scheme 7. Synthesis of DHPMs using NiO NPs.



Scheme 8. Proposed Mechanism for the synthesis of DHPMs using NiO NPs [211].

all reaction stages, especially under solvent-free conditions, utilizing its Lewis acid and basic sites to activate reactants and facilitate product formation.

3. Conclusion

Plant extracts have become a dependable approach for synthesizing nanoparticles because of their safety, environmental friendliness, and cost-effectiveness. The utilization of this environmentally friendly method to synthesise nanoparticles has a wide range of uses in fields such as nanomedicine, pharmaceuticals, cosmetics, environmental science, and the food industry. The high toxicity of traditional methods for NP preparation presents a major challenge, which poses a danger to sectors related to human health. As a result, the scientific community has been continuously looking for novel, secure, and environmentally-friendly methods. In recent decades, there has been significant progress in the production of nanoparticles by biosynthesis, which is considered an environmentally friendly option.

These eco-friendly methods provide exceptional advantages, including as cost-efficiency, environmental compatibility, ease of testing, and lack of toxicity, which make them extremely beneficial for NP synthesis. An additional noteworthy benefit of green synthesis is the use of stability and reducing chemicals, facilitating convenient regulation of nanoparticle shape and size. NiO NPs are extensively used due to their advantageous features in catalysis, magnetism, antibacterial activity, optics, and photocatalysis. Notably, NiO nanoparticles produced by the use of environmentally benign plant extracts are expected to possess safety, sustainability, and non-harmful properties, rendering them appropriate for diverse applications including catalysis, photocatalysis, and antibacterial purposes.

Ethical Approval
This manuscript does not report on or involve the use of any animal or human data or tissue. So the ethical approval is not applicable.

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Authors Contributions
All authors have contributed equally to prepare the paper.

Availability of Data and Materials
There are no associated datasets used in this manuscript.

Conflict of Interests
The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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