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New Frontiers in the Bio-inspired Green Synthesis of NiO NPs and Their Applications: An Overview

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ABSTRACT

Nanoparticles are an important tool for new updations and advancements in diverse sectors. The inorganic metal and metal oxide nanoparticles have enormous research interest because of their great relevance in medicine, wastewater treatment, catalysis, biotechnology, and in the formation of energy storage devices. The NiO NPs can be synthesized using different physical and chemical methods and exploring all their possible applications. Green synthesis is the easy, safe, and effective nanoparticle synthesis route. Green metal and metal oxide nanoparticle syntheses provide the most affordable, convenient, and biocompatible approach for fabricating NiO NPs. This way is a good alternative to the conventional methods of synthesis. Green synthesis, being more constructive, is widely used in research and gives promising outcomes. This review highlighted the unique feature of the NiO nanoparticles. This paper brings forth the usage of green synthesis for synthesizing NiO nanoparticles. It also provides readers with a collective review of the recent development in the green synthesis of NiO NPs and their potential application in different fields.

INTRODUCTION

Nanoparticles are one of the important explorations that are remarkably contributing to bringing advancements in the field of science and technology. This area is drawing recognition because of several advantages of nanoparticles over their macro counterparts. Due to their benefits and wide applications, it has become a renowned area of research in recent times.

The term 'Nanoparticles' consists of two words, 'nano'+ 'particle,' which means extremely small fragments usually in the 1-100 nm range. Depending on the shape of nanoparticles, they can be categorized into 0D, 1D, 2D, or 3D (Ayyub et al. 2001). This field drew attention when researchers found that size can influence the physiochemical properties of a substance. A substance in nanoform behaves differently than its bulk part owing to its changed optical, thermal, and electrical properties and larger surface-to-volume ratio, making them more chemically reactive (Khan et al. 2019). These properties of the nanoparticles are responsible for their use in different fields like electronics, textiles, cosmetics, medicines, and many more.

Some of the important contributions of nanoparticles in different industries are:

- In cosmetic industries, TiO_2 nanoparticles are used in sunscreens to block the dangerous UVB and UVA light.
- In PET (Polyethylene Terephthalate) bottles, SiO_2 nanosheets act as an oxygen barrier, thus preventing the beverage in the bottle from deteriorating.
- In the textile industry, Nanosilver with antimicrobial properties, TiO_2 with water-repellent, and SiO_2 with dirt-repellent attributes are used in textiles.
- The nanoscale additives in polymer composite materials are used in tennis rackets and motorcycle helmets, making them lightweight and durable.
- Nano TiO_2 is also used in coatings to form self-cleaning surfaces. A sealed film of water dissolves the dirt making the surface clean.

SYNTHESIS OF NANOPARTICLES

The importance of nanoparticles was well known, so different methods to synthesize these were searched. The top-down and bottom-up approaches were traditionally used to synthesize nanoparticles. The top-down approach includes reducing the size of the bulk material into nanosized

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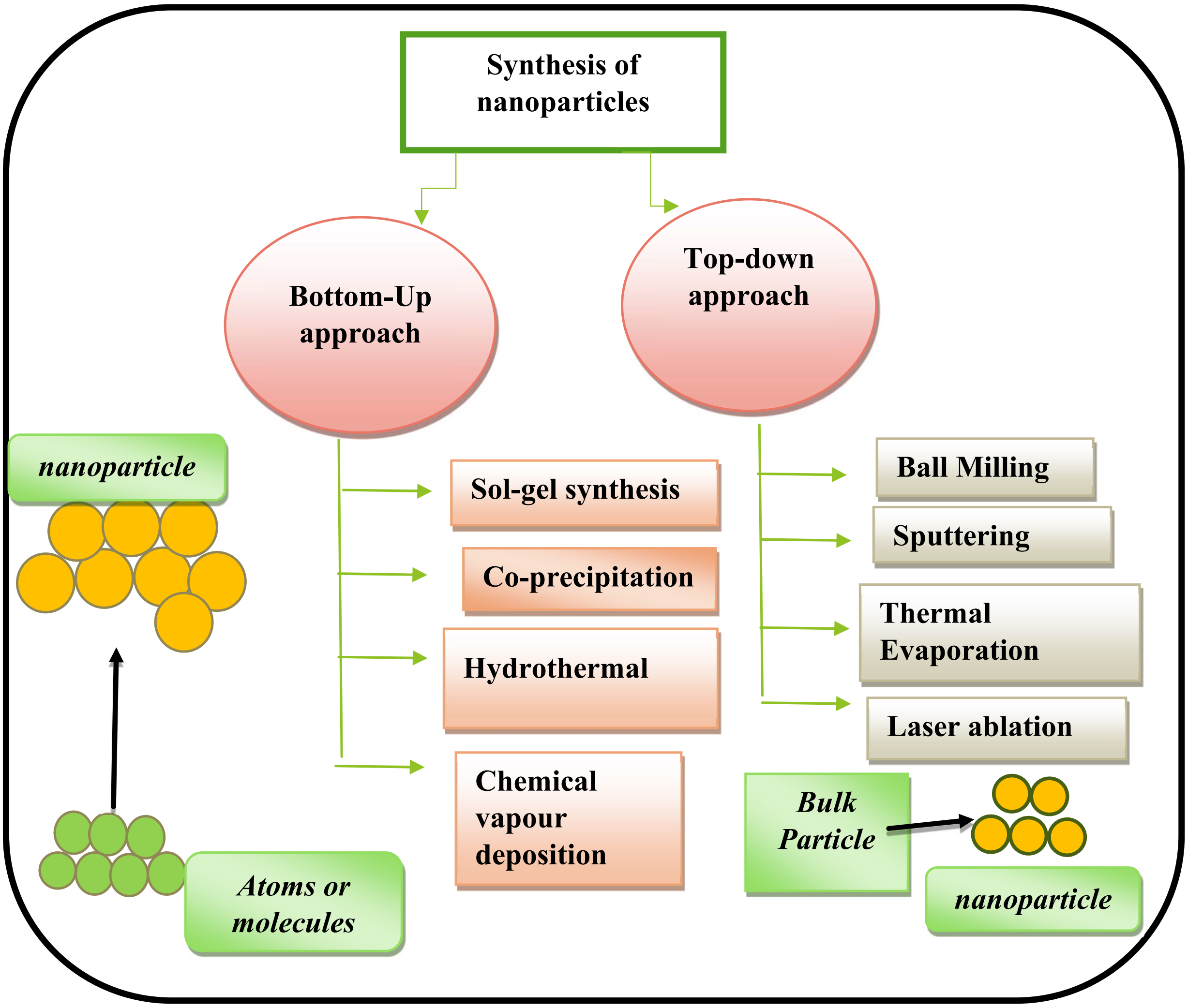

Fig. 1: Flowchart showing conventional methods for nanoparticle synthesis.

particles. Some methods involved in this approach are grinding, cutting, ball milling, Sputtering, and laser ablation (Tiwari et al. 2012). Though these methods are simpler, they have major drawbacks and are unsuitable for nanoparticle synthesis. They require large installations, so they are quite expensive. Besides this, these methods are unsuitable for the soft samples because the mechanical devices used for grinding are stiff and hard (Fig. 1).

The bottom-up approach includes small units like atoms or molecules that combine to form a nanoscopic dimension particle. This approach is based on the principle of molecular recognition. The problem with this method is that various undesired atoms and molecules may be present in the deposited material, and the cost of the chemicals is high. Moreover, these chemicals are toxic, and various poisonous by-products are obtained.

The physical and chemical methods for nanoparticle synthesis, along with their advantages and disadvantages, have been discussed by Namra Abid and the team (Abid et al. 2022). Though these methods possess some advantages, we utilize a different route for nanoparticle synthesis due to their major drawbacks.

GREEN SYNTHESIS

The difficulties faced with the classical approach emphasized the need for an alternative pathway for nanoparticle synthesis. This drove our attention towards herbs due to the availability of effective phytochemicals in various plant extracts, which can potentially reduce metal salts into metal nanoparticles. In today's era of rapid industrialization, urbanization, and population explosion, methods are absolutely necessary to minimize toxic chemicals: green synthesis is an effective tool to serve this purpose. Green synthesis paves the way for using plants and their extracts to synthesize nanoparticles. Thus, green synthesis uses environment-friendly procedures and focuses on minimizing the usage of hazardous chemicals. It comprises parts of plants such as leaves and fruit peels for nanoparticle synthesis because these contain phytochemicals that act as good reducing, capping, and stabilizing agents for metal nanoparticle synthesis. The advantages of green synthesis are that it is an easier, cost-effective, biologically safe, environment-friendly, and reliable method (Fig. 2).

This bioinspired route gives productive outcomes: therefore, it is being adopted by many for nanoparticle synthesis. Among various nanoparticles, metal oxide nanoparticles are especially drawing recognition due to their vast applications in different fields. Green synthesis is adopted for their synthesis because of their usage as antifungal agents, antibacterial agents, semiconductors, catalysts, nano-medicines (Hussain et al. 2023), and more (Fig. 3).

This biological way provides another important benefit of contributing to keeping the environment clean (Singh et al. 2018) describe that the bio-inspired green synthesis of metal and their oxide nanoparticles are a remedial approach to keeping the environment clean. Many nanoparticles have been synthesized through this eco-friendly route like Ag (using *Tectona grandis*) (Rautela et al. 2019), ZnO (using *Coriandrum sativum*, Varada V (Ukidave & Ingale 2022), NiO (using *Aegle marmelos*) (Angel Ezhilarasi et al. 2018) and more.

The plant-mediated green synthesis plant acts as chemical factories that act as reducing agents. Different phytochemicals are present in the plant extract, which probably act as reducing stabilizing agents and prevent the accumulation of developed nanoparticles. The plant-mediated nanoparticles also show low toxicity as the extract is an adsorbent to absorb the toxicity associated with the plant. Further, the other one of the most important advantages of the plant-mediated green synthesis of the nanoparticles is the rate at which the synthesis occurs (Angel Ezhilarasi et al. 2020). The

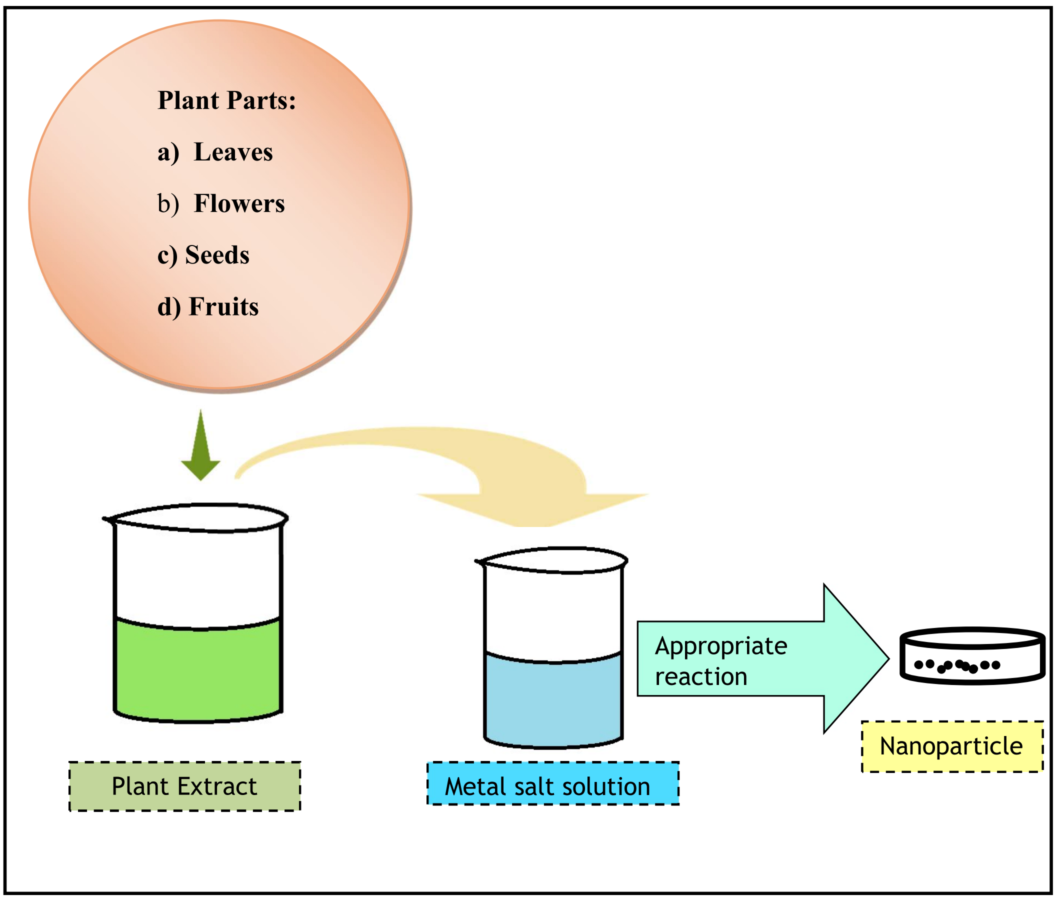

Fig. 2: Diagrammatic overview of the procedure involved in green synthesis.

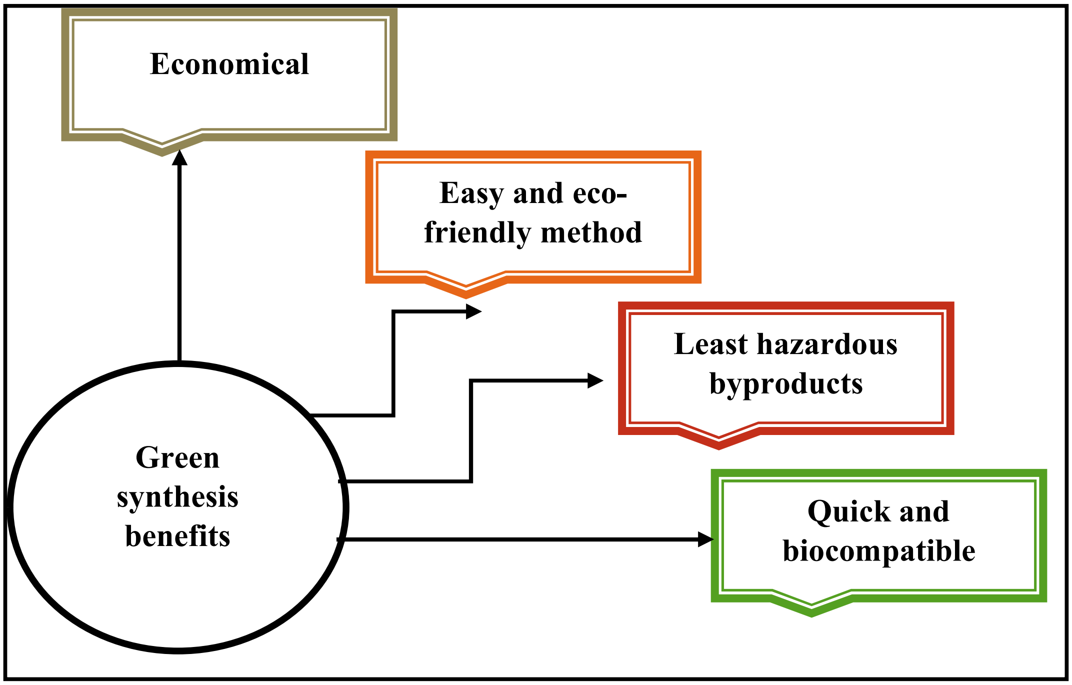

Fig. 3: Some advantages of green synthesis.

plant-mediated synthesis of the nanoparticles occurs much faster than the physical and chemical methods. In the plant-mediated green synthesis of nanoparticles, the leaf, fruit, bark root, and steam play prominent roles in the green synthesis of nanoparticles.

METAL-OXIDE NANOPARTICLES

Scientists are paying more heed to metal oxide nanoparticles because of their easy mode of formation and multidisciplinary implementations. Metal oxide nanoparticles can tackle some major issues owing to their unique properties. They possess antibacterial activity and are useful in wastewater treatment (Naseem & Durrani 2021). They serve as therapeutic agents and are used in paints, cosmetics, and ion batteries. The different useful applications of metal oxide nanoparticles in nanotechnology have been discussed by Murthy & Chavali (2019).

The transition elements have properties like high charge density, catalytic behavior, and the ability to form coordination compounds, so their metal oxides are important. Iron oxide nanoparticles possess unique features such as nontoxicity and biocompatibility and are successfully used in drug delivery systems, electrochemical sensing, hyperthermia, photothermal therapy, nanozymes, and MRI (Vallabani et al. 2018). Zinc oxide nanoparticles are used in drug-delivery systems, cosmetics, biomedical, agriculture, biosensors, and gas sensors (Bedi & Kaur 2015). Tin oxide nanoparticles are electrochemical/chemo-resistive sensors (Sharma et al. 2021). Ahmad et al. (2021) states various methods for synthesizing tin oxide nanoparticle and their properties and highlights their various biological and physio-chemical applications. Copper oxide nanoparticles have catalytic, anti-cancer, and anti-bacterial applications (Akintelu et al. 2020). TiO_2 nanoparticles have promising uses in drug-delivery systems. Their property to act as photo-sensitizing agents against malignant tumors and their effectiveness in photo-dynamic inactivation of antibiotic-resistant bacteria have been discussed by Ziental et al. (2020). The recent achievement in the field of metal oxide nanoparticles includes the development of nanostructured oxides with two or more metallic components. Different uses of these metal oxide nanoparticles have made these fascinating for researchers, and explorations in this field are continuing for the betterment of existing technologies. NiO nanoparticles are one of many useful metal oxide nanoparticles synthesized via green synthesis. These possess distinctive features like low porosity, high photo absorption, and a larger surface-to-volume ratio, making them convenient to use in different fields. NiO nanoparticles possess superior ferromagnetic properties, chemical stability, and high coercive forces (Ahmad et al. 2022a), contributing to their role in various day-day technologies.

APPLICATIONS OF NIO NANOPARTICLES

Photocatalytic Applications

(i) Wastewater Treatment by Degradation of Toxic Dyes

In the world of rapid development and industrialization, one of the major issues we face is the effective treatment of thousands of liters of wastewater daily for sustainability. Wastewater contains many impurities like sulfates, nitrates, and phosphates and may even contain synthetic dyes and other toxic pollutants. Synthetic dyes are non-biodegradable, toxic, and potentially even carcinogenic, so their removal is paramount. Adsorption is a productive and cost-efficient way to remove these impurities, thereby reducing contaminants in wastewater. Due to their high surface area, photocatalytic activity, and other chemical properties, NiO nanoparticles are used as adsorbents in purifying water. Hamidian and team carried out the bio-inspired synthesis of NiO nanoparticles using Biebersteiniamultifida extract to examine their photocatalytic activity against acid orange 7 (AO7) dye and found these particles to exhibit excellent adsorption as well as photocatalytic behavior (90.2%) (Hamidian et al. 2021). NiO nanoparticles synthesized using Allium cepa peels aqueous extract were used against the Congo red direct dye by Rafique et al. (2021) reported that decolorization of the dye was up to 90% at optimized conditions (Rafique et al. 2021). Nateghi et al. (2021) investigated the decolorization of synthetic wastewater containing mono azo orange II dye at the laboratory scale. They reported that 0.6 $g.L^{-1}$ was the optimum amount of the adsorbent required for the complete decolorization of synthetic wastewater under the condition of 50 $mg.L^{-1}$ initial dye, PH 3, and agitator speed 100 rpm for 30 minutes (Nateghiet al. 2021). Adinaveen et al. (2019) studied the photocatalytic properties of NiO nanoparticles and found these to exhibit high degradation activity against Rhodamine b (92.3%) under UV light illumination (Fig. 4).

Motahari and their team members synthesized NiO nanoparticles through the hydrothermal process in the presence of H2acacen ligand and found them productively work to remove Rhodamine B (Motahari et al. 2015). NiO nanopowder is also useful for removing heavy metals in wastewater treatment. Its application as a productive adsorbent for the heavy metals Zn and Pb in an aqueous solution is discussed in Abdl El Fatah & Ossman (2014).

(ii) Photodegradation

NiO nanoparticles also exhibit photocatalytic activity in the degradation of polyethylene films. Olajire & Mohammed (2020) carried out the biological synthesis of NiO nanoparticles using *Ananas comosus* extract. They suggested that using synthesized nanoparticles in the polymer matrix of LDPE can enhance its photodegradation (Olajire & Mohammed 2020). Wang et al. (2005) reported that the catalytic activity of NiO nanoparticles lowers the decomposition temperature for the thermal decomposition of ammonium perchlorate by 93°C compared to that of the bulk NiO particle. It also increases the heat of decomposition for the same. NiO nanoparticles are important in degrading pharmaceutical products like paracetamol (Ahmad et al. 2022b). The catalytic role of the NiO nanoparticles is also important as they have applications in synthesizing substituted Imidazole (Mahadevaiah et al. 2018).

Medical Applications

(i) Anti-Cancer Activity

NiO nanoparticles are useful in the medical field. (AlSalhi et al. 20202) Worked on the cytotoxicity of NiO nanoparticles towards HeLa cancer cells and reported these to exhibit good response at a concentration of 180 $\mu g.mL^{-1}$. NiO nanoparticles also have an inhibitory role against Hep-G2, MCF-7, and HT-29 cancer cell lines responsible for liver, breast, and colon cancer, respectively (Kouhbanani et al. 2021). Angel Ezhilarasi et al. (2016) synthesized NiO nanoparticles and explored their cytotoxicity effect against HT-29 cancer cells. They found that green synthesized nanoparticles showed better cytotoxicity. In Zhang et al. (2021), NiO nanoparticles were examined against the carcinoma cell lines of FLO-1, ESO26, 0E33, and KYSE-207, and it was reported that the nanoparticles showed significant cytotoxicity against all these cell lines. Sabouri et al. (2021) conducted a bio-synthesis of NiO nanoparticles using Arabic gum and reported their cytotoxic effects on normal CNs cell lines and cancer U87MG cell lines Sabouri et al. (2021). Betageri et al. (2021) analyzed the cytotoxic effects of green synthesized NiO nanoparticles against A549 cancer cell lines and demonstrated that the synthesized nanoparticles exhibited significant anticancer activity.

Anti-Bacterial Activity

NiO nanoparticles also possess antibacterial properties. Therefore, these serve as remarkable antibacterial agents. These nanoparticles resist the growth of gram-positive and gram-negative bacteria. Ilbeigi and the team worked on the antibiotic activity of NiO nanoparticles against some bacterial strains. They reported the effectiveness of these nanoparticles even for those bacteria which had developed resistance to antibiotics like rifampicin, cefazolin, penicillin, ampicillin, erythromycin, and streptomycin (Ilbeigi et al. 2019). Khashan et al. (2016), in their study, indicated that NiO nanoparticles could potentially cause growth inhibitions of bacterial strains because, on their application, the bacterial cell wall showed increased permeability, so amoxicillin accumulations occur in them. Khan et al. (2021) conducted a comparative study of green synthesized NiO nanoparticles and chemically synthesized nanoparticles for antibacterial, anticancer, and antioxidant activities. They reported the effectiveness of biologically synthesized nanoparticles against the bacteria *E. coli, B. bronchiseptica, B. subtilis* and *S. aureus*.

Anti-diabetic, Anti-pseudomonal, and Anti-oxidant activity

NiO nanoparticles also have significant antidiabetic effects because they exert a hypoglycemic effect, as discussed in Betageri et al. (2021). These also exhibit antimicrobial and antioxidant properties (Haq et al. 2021). Khan et al. (2021) reported that phytomolecules-coated NiO nanoparticles showed excellent antioxidant activity. Irum et al. (2021) carried out the chemical synthesis of NiO nanoparticles. They worked on the antipseudomonal activity of Al-doped NiO nanoparticles and reported the resistance of these nanoparticles against pathogens, especially *Pseudomonas aeruginosa* (Irum et al. 2021). Zhang et al. (2021) worked on the anti-oxidant properties of NiO nanoparticles and reported in their result the effectiveness of these nanoparticles as an anti-oxidant through DPPH assay. Srihasam et al. (2020) evaluated NiO nanoparticles' anti-oxidant and anti-microbial activity. They reported that the nanoparticles were found efficient against fungi *A. niger* and *A. fumigatus,* and they also exhibited strong anti-oxidant properties shown through DPPH reduction assay. Uddin et al. (2021), synthesized NiO nanoparticles from *Berberis balochistanica* stem and reported the nanoparticles to exhibit total antioxidant capacity (64.77%) and for 2,2- diphenyl-1-picrylhydrazyl (71.48%).

Agricultural Applications

Uddin et al. (2021) also reported that the nanoparticles could have stimulatory efficacy in enhancing the rates of seed germination and seedling growth rates. Singh et al. (2022) carried out the biosynthesis of NiO nanoparticles using Spirogyra sp. cell-free extract. They reported that the nanoparticles showed stimulatory and inhibitory effects on Mung bean seed germination and seedlings at varying concentrations.

Electronic Devices

NiO nanoparticles also exhibit a vital part in enhancing the structural properties of other nanoparticles: TiO_2 has limitations because of the rapid recombination rates of electron/hole pair and the wide band gap, so NiO nanoparticles were used productively in enhancing the structural properties of TiO_2 (Mannaa et al. 2021). NiO

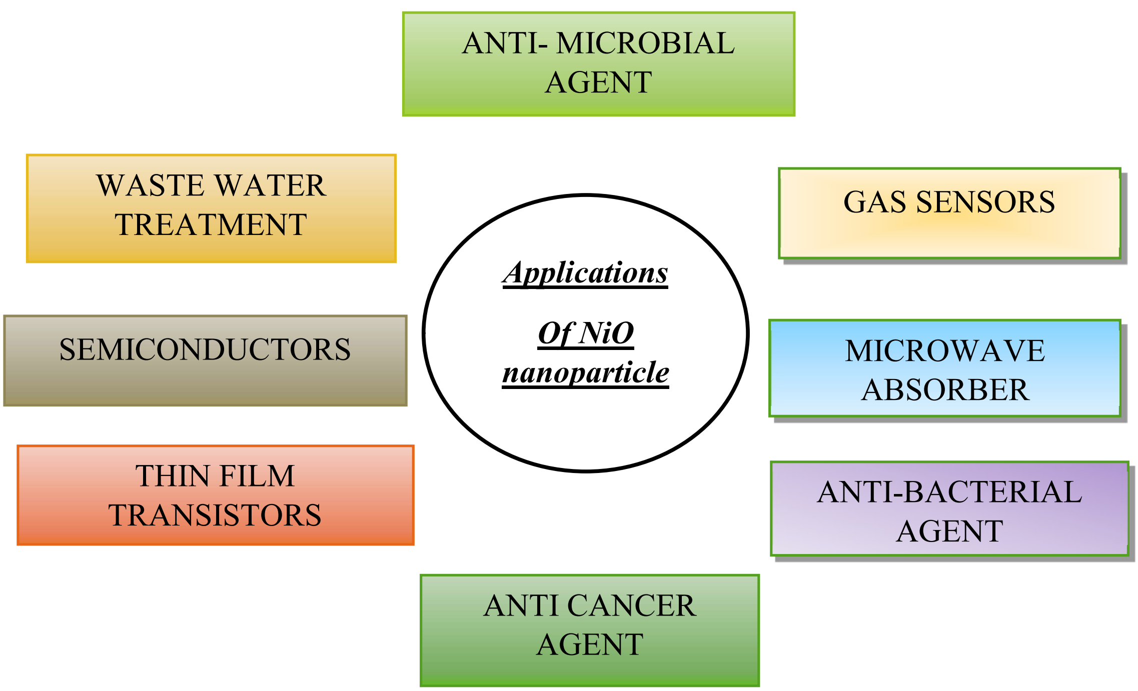

Fig. 4: Different applications of NiO nanoparticles.

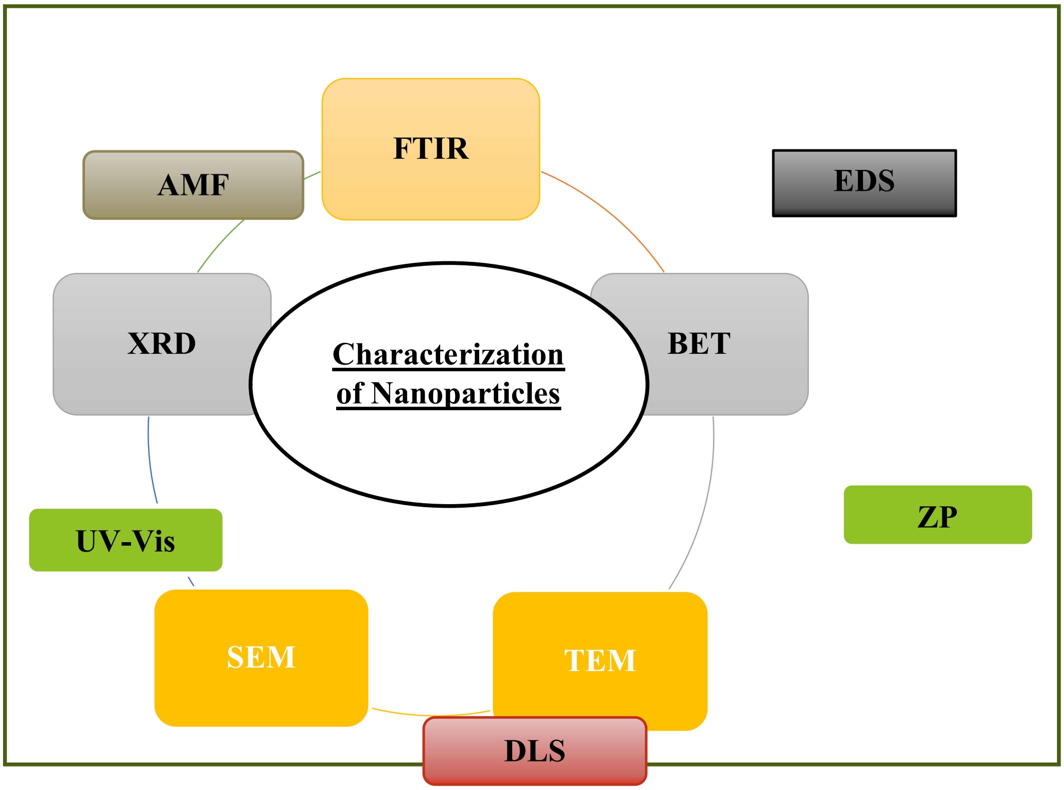

Fig. 5: Different characterization techniques of NiO NPs.

nanoparticles are also used in carbon-based perovskite solar cells to increase the device performance by reducing charge recombination (Cai et al. 2019).

Niu et al. (2019) synthesized NiO-decorated tetragonal rutile SnO_2 nanosheet-based sensors and found these to have improved sensitivity and excellent selectivity for ethanol gas (Niu et al. 2019). NiO nanoparticles also exhibit electrochromic properties (Obaida et al. 2022). Arif et al. 2018: prepared a chlorine gas sensor based on the NiO nanoparticles (Hamidian et al. 2021).

CHARACTERIZATION OF NANOPARTICLES

Nanoparticles can be synthesized differently, leading to their different sizes and shapes, ultimately affecting their properties. It is, therefore, necessary to know the size, shape, crystal structure, and other properties of the synthesized nanoparticle for its efficient usage. Nanoparticle characterization hence plays a major role. Characterization of nanoparticles is done to know the size, distribution, crystal structure, surface charge, and other properties of the nanoparticles. Various techniques are used for the characterization, such as UV-Visible spectroscopy, Fourier Transform Infrared Spectroscopy (FT-IR), Zeta Potential (ZP), transmission electron microscopy (TEM), Scanning Electron Spectroscopy (SEM), Atomic Force Microscopy (AMF), Dynamic Light Scattering (DLS), Brunauer-Emmett-Teller (BET), X-ray Diffraction (XRD), Nuclear Magnetic Resonance (NMR), Thermogravimetric Analysis (TGA), Diffuse Optical Spectroscopy (DCS) and many more (Fig.5). Different techniques derive different information for a particular sample to be analyzed. For example, XRD gives information about the crystal structure, BET tells us about the surface area and the pore size of nanoparticles, FTIR gives an idea about the surface composition of ligand bindings and the analysis of the functional groups present on the surface of the green synthesized nanoparticles. For this analysis, the developed nanoparticles were generally scanned in the 400-4000 cm^{-1}. With the help of FTIR analysis, the binding capping and stabilizing agents were identified. AFM gives knowledge about nanoparticle size and shapes in 3D mode (Table 1). The scanning electron microscope uses a beam of electrons to capture the image of the fabricated nanoparticles. With the help of SEM images, the particle size, surface morphology, and distribution of the particles were estimated.

Table 1: Summary of the synthesis, characterization and applications of NiO NPs.

S. No	Name of the plant	Size	Morphology	Characterization Techniques	Applications	Reference
1	*Biebersteinia multifida*	54-58 nm	Cubic structure	SEM, EDX, PXRD, UV-Vis, Raman analysis	Photocatalytic degradation of acid orange 7 dye under visible light	Hamidian et al. (2021)
2	*Aegle marmelos*	8-10 nm	Single crystalline with fcc phase	XRD, HR-TEM, HR-SEM, FT-IR	Antibacterial activity Cytotoxicity towards A549 cell culture Photocatalytic degradation of 4- chlorophenol(4-CP)	Angel Ezhilarasi et al. (2018)
3	*Andrographis paniculata*	24 nm	Cubic structure	XRD, FTIR, SEM, HRTEM, UV-Vis	Photocatalytic and anti-cancer activity	Karthik et al. (2019)
4	*Berberis balochistanica*	31.44 nm	Rhombohedral agglomerated shape	FTIR, XRD, UV-Vis, SEM	Antioxidant activity, stimulatory effect to fasten the rates of seed germination and seedling growth	Uddin et al. (2021)
5	*Ananas comosus*	0.63-5.75 nm	fcc crystalline structure	UV-Vis, XRD, HRTEM, FTIR, EDX	Photocatalytic activity	Olajire et al. (2020)
6	*Salvia hispanica* L.	30 nm	Spherical	TGA, FTIR, UV-Vis, XRD, EDAX	Cytotoxicity and photocatalytic activity	Sabouri et al. (2021)
7	*Stevia*	20-50 nm	Spherical, and a few are agglomerated	XRD,FTIR, FE-SEM, TEM, UV-Vis	in -vitro oxidant and antimicrobial activity against multi-drug resistant microbes	Srihasam et al. (2020).
8	*Ageratum conyzoides* L.	11.5 nm	Cubic structure	XRD, FTIR, TEM	Catalytic activity	Wardani et al. (2019)
9	*Rhamnus virgata*	34 nm	-	FTIR, XRD, SEM,TEM, DLS	Antibacterial activity, anti-fungal activity, alpha-amylase inhibition, antioxidant activity, antileishmanial activity	Iqbal et al. (2019)
11	*Stevia*	2-16 nm	Spherical	-	Antibacterial activity against S. mutans	Moghadam et al. (2022)
12	*Abutilon indicum*	-	-	XRD, SEM, EDX, DLS, FTIR, UV-Vis.	Antioxidant, Antibacterial, and Anticancer activities	Khan et al.:2021
13	*Allium cepa*	-	-	SEM, FTIR, UV-Vis	Photocatalytic Activity- Degradation of Congo red direct dye	Rafique et al. (2021)
14	*Calendula officinalis*	60.39 nm	Spherical	TEM, SEM, EDS, FTIR, XRD, UV- Vis.	Antioxidant activity, Cytotoxicity, Anti-esophageal Carcinoma activity	Zhang et al. (2021)
15	*Areca catechu*	5.46 nm	Hexagonal shaped	XRD, SEM, TEM, UV- Vis.	Anti-diabetic and Cytotoxicity Effects.	Shwetha et al. (2021)

CONCLUSION

A vital and budding research field that has the potential to bring advancements in the world and provide solutions to many existing problems is nanoparticles. The nano-sized particles do not limit their usage to one field. Rather, they have multidisciplinary implementations. The major targeted areas are cancer treatment, gene therapy, and drug delivery system. Owing to their importance, synthesizing them also draws our attention.

The synthesized NiO nanoparticles have their role in various areas such as water treatment, pharmaceutical and medical sector, electronic devices, photodegradation and catalysis, and many other fields. The principle is that various phytochemicals in the plant extracts can remarkably reduce the metal salts into corresponding nanoparticles, thus synthesizing desired nanoparticles. This route helps keep the environment clean and less toxic by minimizing the usage of hazardous chemicals. The overview of the diverse benefits of NiO nanoparticles reflects that we should focus more on them and other nano-dimensional particles and explore more of this area to bring forth new innovative solutions for the welfare of mankind.

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