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Green Nanomaterials for Drug Delivery

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Synonyms

Biodegradable polymers, Drug delivery, Green nanomaterials, Green nanotechnology, Nanotechnology

Definition

Rapid advances in drug discovery methods have led to an exponential increase in new drugs. Due to the diversity of physical and chemical properties of various drugs, we need more innovative drug delivery systems. In order for a drug to remain therapeutically effective, it is necessary to protect the site of action and preserve its biological and chemical properties. So far, a lot of research has been done in this field. Green nanotechnology refers to developing "clean technologies" to produce nanomaterials and safe products for the environment or human health. It produces nanoproducts that provide solutions to environmental problems. Using nanotechnology in pharmaceutical science research, achieving targeted drug delivery has also been considered by researchers. Studies have shown that nanometal particles, polymers, liposomes, and biological materials are used as green nanomaterial as examples of drug delivery systems. Nanosized polymers, low molecular weight molecules, and biodegradable polymers are some of the best candidates for developing more efficient green chemistry methods to synthesize nanodrug delivery vehicles. Nanoparticles are used to solve major drug delivery problems, including protein delivery, cancer delivery, drug delivery to the eye, control of intracellular infections, and reduction of multidrug resistance. These particles can enter the human body faster than larger particles due to their tuning. Environmentally friendly development of nontoxic, renewable nanocarriers for drug delivery to target disease sites dramatically enhances the therapeutic effect of the original drugs. It eliminates potential risks to human health

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and the environment. The use of green nanomaterials is crucial for the future development of pharmaceutical technologies, especially in drug delivery.

Introduction

With particle dimensions in the range of 1–100 nm, nanotechnology is a popular subject of current research that deals with particle design, synthesis, and manipulation, applied in science ranging from material science to biotechnology (Guo et al. 2005). The synthesis of nanoscale materials and the research or application of their unique physicochemical and optoelectronic properties have opened new basic and applied frontiers in this up-and-coming technology. Nanotechnology is fast gaining traction in various fields, including cosmetics, health care, environmental health, food and feed, mechanics, optics, chemical industries, electronics, biomedical sciences, and especially drug-gene delivery. Figure 1 illustrates the green material classification and their varieties that have found wide applications in drug delivery based on green nanomaterials (Schulte et al. 2013; Han et al. 2013).

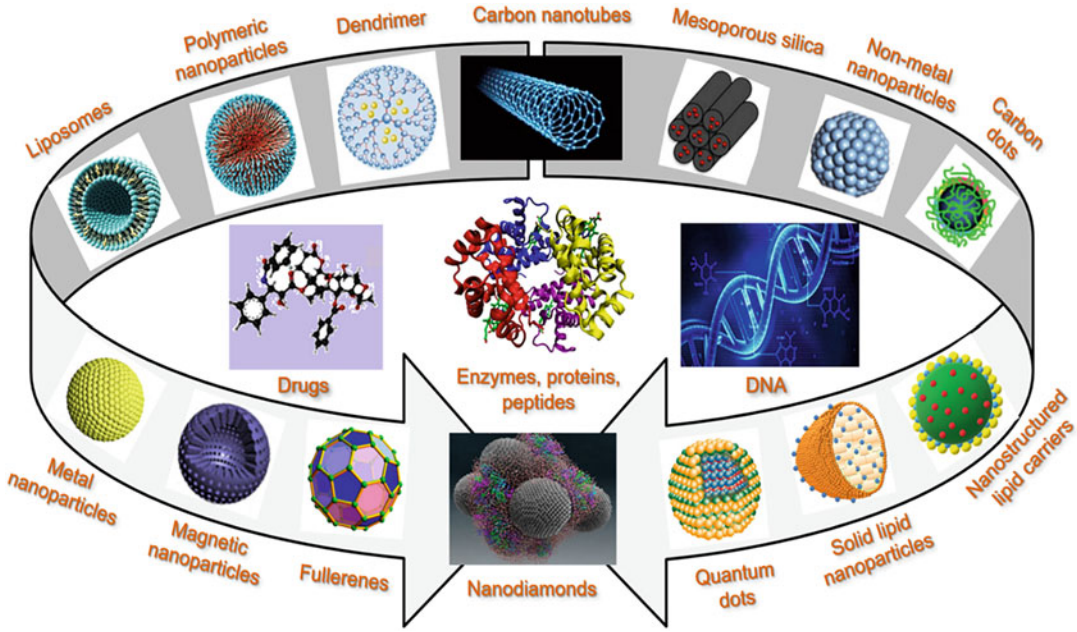
Over the past few years, nongreen NPs (nanoparticles) have been developed for use in various biomedical apps which their prominent methods of synthesis have been focused on chemical reaction methods. Such chemical methods have some disadvantages that frequently involve applying hazardous chemicals that raise the speculation of high toxicity. In the past conventional chemical methods, stabilizing agents commonly are used for metal ions undesired agglomeration in NP formation process, while their bioincompatible entity makes them hazardous materials to the environment (McNaughton et al. 2005).

As a result of the use of environmentally friendly and less dangerous nanotechnology, the term green nanotechnology has been coined. When it comes to green nanotechnology, protocol design is critical. Nanomaterials with many functions, such as those used in medication delivery, have been the subject of several studies and developments. Green chemistry demonstrates that nanotechnologies improve process environmental sustainability while lowering costs and reducing hazardous materials. Due to its environmental friendliness, green nanotechnology is an emerging topic with a wide range of applications. Thus, it is an excellent example of new research efforts by scientists to apply sound information to reduce or eliminate environmental pollution and improve human well-being. Sustainability, health, and safety are all impacted by nanotechnology and green approaches when they are used together to produce nanoproducts (Koo et al. 2005; Commission, C.f.t.E 2004; Watlington 2005; Sinha et al. 2007).

Green Nanotechnology-Driven Drug Delivery

Active pharmaceutical molecules, including medications, vaccines, antibodies, enzymes, peptides, proteins, and other related substances, are being investigated and produced to increase the efficacy and administration of DDS (Anselmo and Mitragotri 2014). DDS (emulsions, suspensions, and solutions) must develop an advanced type due to the limitations of conventional DDS, such as first-pass effect, instability, intolerance, fluctuations in plasma levels of the drug, absence of a long-term effect, limited effectiveness, lack of selectivity, and poor bioavailability (Hans and Lowman 2002; Kumar et al. 2017). To avoid fast deterioration of medications or protect them against clearance, a slew of controlled DDS has been devised. The first generation of DDS was created between 1950 and 1980, with a focus on transdermal and oral sustained-release methods. The second generation (2G) of DDS arose between 1980 and 2010, focused mostly on green nanotechnology (Park 2014). The third generation of DDS (to be launched in 2010) will have to overcome both physiochemical (poor water solubility, controlled drug release kinetics) and biological (target site delivery) challenges with the help of nontoxic excipients (Kanwar et al. 2019).

Improved bioavailability, controllable and sustained drug release, high drug-loading capacity, prolonged circulation time, enhanced intracellular penetration, and targeted delivery to specific



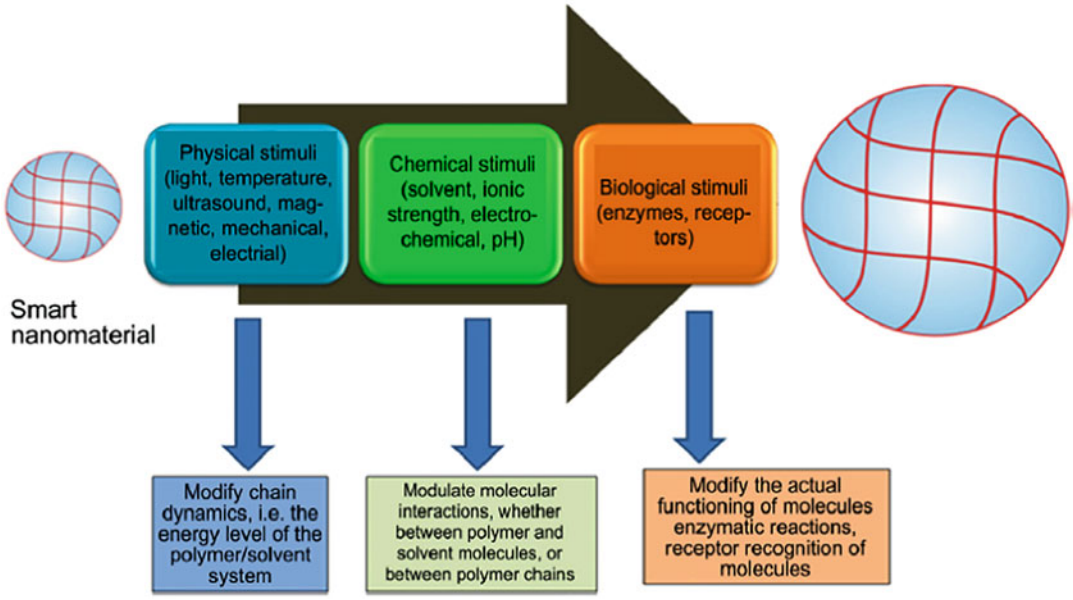
Green Nanomaterials for Drug Delivery, Fig. 1 Nanotechnology of green materials in drug delivery application areas. (Schulte et al. 2013)

sites or organs are just some of the advantages of using NPs as potent DDS (for the prevention and treatment of different diseases). NPs can be administered in a variety of ways, including parenteral and intravenous (Khosa et al. 2018).

Green Polymers

Various processes are used to create polymeric nanoparticles, depending on their intended use and the sort of medicine they would enclose (Hans and Lowman 2002; Soppimath et al. 2001). These nanoparticles are commonly employed to nanoencapsulate a variety of bioactive compounds for nanomedicine. Biodegradable polymeric nanoparticles such as polyaniline, polylactide, polyglycolide, polyorthoesters, poly (lactic-co-glycolide), polycaprolactone, poly (vinyl alcohol), polyglutamic acid, poly(vinyl alcohol), and poly(methyl methacrylate) are highly preferred to other polymers, because they have shown to be extremely effective in the delivery of drugs. Controlled release, subcellular size, and biocompatibility are all characteristics of nanoparticles that should be used in this application (Zambaux et al. 1999). Despite the fact that these nanomedicines need to be stable in the blood, non-thrombogenic, non-immunogenic, biodegradable, bypassing the reticular-endothelial system, and applicable to a variety of molecules like drugs, proteins, peptides, or nucleic acids, they can be used to treat a wide range of diseases and conditions, including cancer (van Vlerken et al. 2007). The general synthesis and encapsulation of biodegradable nanomedicines are shown in Fig. 2 (Tiyaboonchai 2013). For the past two decades, numerous work has been done to develop effective nanomedicines from biocompatible and biodegradable nanopolymers (Mundargi et al. 2008).

Various green polymeric micelles for anticancer drugs have been developed and well-known to obtain highly selective delivery and accumulation around the tumor microenvironment. A series of pH-sensitive drug conjugates were fabricated by Du et al. to deliver DOX anticancer drugs. Using extracellular pH gradients that are around tumor microenvironments, the drug-loaded polymer carriers respond to tumor presence by simultaneously releasing the drug to the accumulations that are as a tumor. Polymeric carrier enhances not only the



Green Nanomaterials for Drug Delivery, Fig. 2 Illustration of stimuli-responsive green polymer function and responses. (Thambiraj et al. 2018)

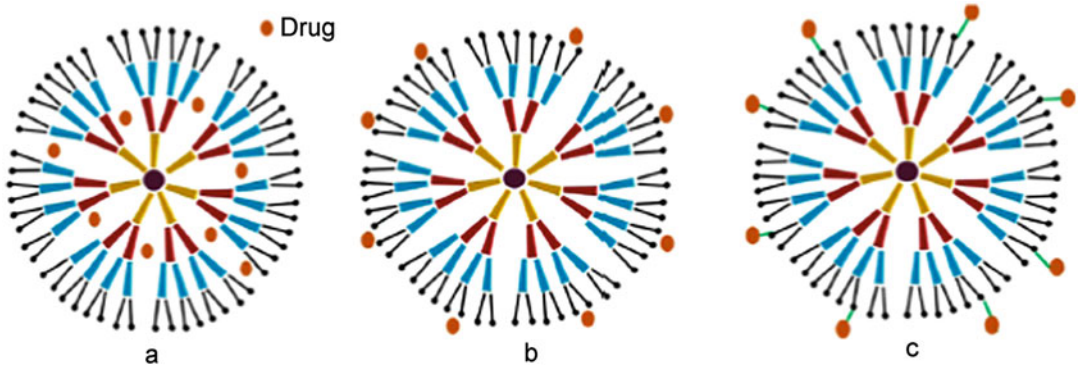
permeability but retention effect. Roque et al. reported the delivery of nystatin as an antifungal drug added to PNP bioadhesive. They greatly increased the adhesion capacity for oral delivery applications to provide long-term release in vitro and in vivo. Modified types of D,L-lactide/glycolide NPs also were used for 5-fluorouracil and indomethacin drugs. The entrapment efficiency was attained >75% for submicron PNP-loaded indomethacin. For harnessing the multidrug resistance that occurs in the chemotherapy process, block copolymer made by PEG-PLL-PAsp can code the simultaneous delivery process of BCL-2siRNA and DOX for hepatic carcinoma therapy (Sun et al. 2018).

Liposomes

Liposomes are lipid vesicles that were developed in the 1960s in order to comprehend new polymer nanocontainers better. Liposomes were first conceived as a model system for studying biological membranes, but by 1970, they had evolved into a drug delivery mechanism (Graff et al. 2004). Liposomes are lipid vesicles that were developed in the 1960s to understand novel polymer nanocontainers better. Liposomes were first designed as a model system for studying biological membranes in the 1960s, but by 1970, they had evolved into a drug delivery method (Graff et al. 2004). Liposomes offer a wide range of applications in the pharmaceutical sector, including as a targeted delivery system for anticancer, antifungal, and vaccine medicines. They're also used in the cosmetics industry to make shampoos and other skincare products. They are very important tools in diagnostics as they can degrade in the cells after delivery (Graff et al. 2004; Salata 2004). Figure 4 shows that liposomes were claimed to be the first artificial nanoparticles used for medication administration, but one major downside was their penchant for fusing in aqueous environments and releasing contents before reaching the target location. Nanoparticle replacement or stabilization methods employing newer nanoparticle substitutes are now being sought (Nath and Banerjee 2013).

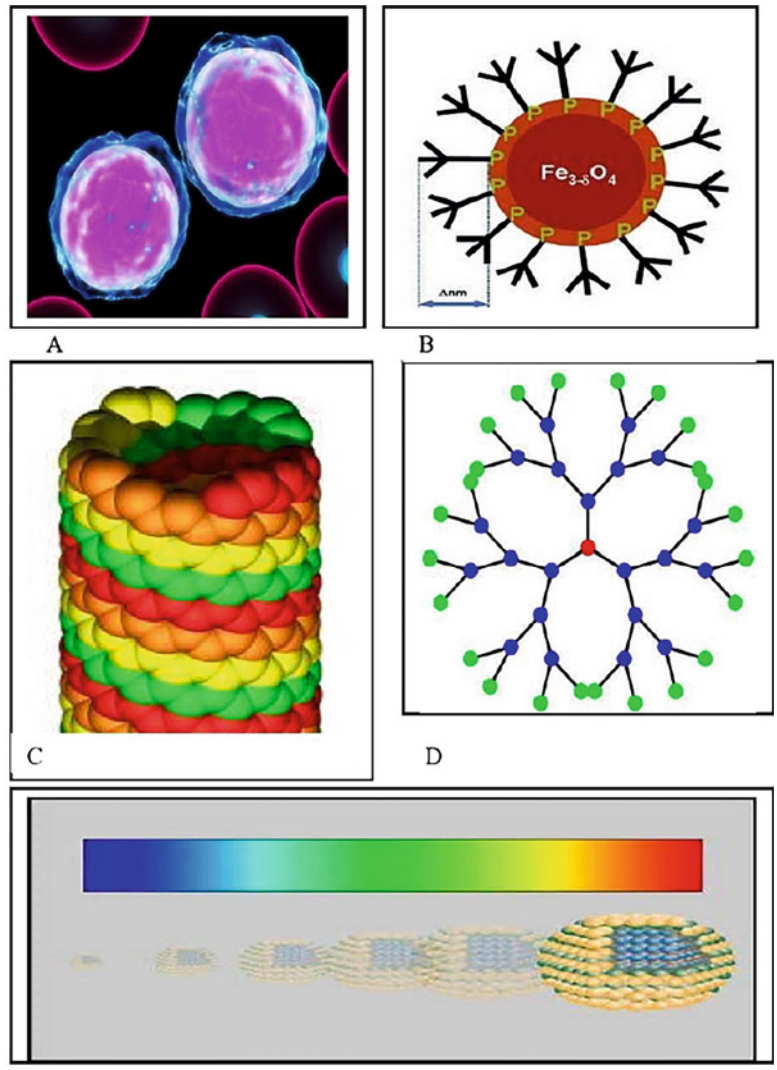
Superparamagnetic Nanoparticles

These iron oxide-based SPIONs (superparamagnetic nanoparticles) can be coated with either inorganic materials like silica and gold or organic components such as phospholipids, fatty



Green Nanomaterials for Drug Delivery, Fig. 3 Models of drug-dendrimer interactions: (**a**) entrapment, (**b**) adsorption, and (**c**) conjugation. (Yoon et al. 2005)

Green Nanomaterials for Drug Delivery, Fig. 4 Different classes of nanoparticles. (**a**) Nanoparticle in liposomes; (**b**) superparamagnetic nanoparticle; (**c**) nanotube; (**d**) dendrimer; (**e**) quantum dots with changing optical properties. (Nath and Banerjee 2013)



acids, peptides, and surfactants that have a phospholipid or fatty acid core (Fig. 3b) (Gupta and Curtis 2004; Lu et al. 2007). SPIONs have a unique property that sets them apart from other nanoparticles: the ability to adhere to a magnetic field without maintaining any residual magnetism after the field has been removed. MRI contrast-enhancing agents, selective bio-separations, MRI contrast-enhancing agents in drug delivery systems, magnetic hyperthermia (local heat source in tumor therapy), and magnetically assisted cell transfection are all examples of applications that benefit from this ability (Neuberger et al. 2005; Aptekar et al. 2009).

Solid Lipid Nanoparticles (SLNs)

SLNs, as the forerunners of the nanocarrier family, have demonstrated drug delivery potential. They have submicron colloidal forms, making them suitable as drug carriers from a structural standpoint. For secondary and tertiary drug delivery, SLNs have the benefit of encapsulating hydrophobic pharmaceuticals, indicating a novel prototype with the potential to be used. They described the use of chitosan- and PEG polymer-functionalized SLNs for targeting the distribution of cancer-healing PTX medication. For M109-HiFR cells, the PTX inhibitory dose was dramatically lowered by this compound in vitro. Another study found that loading olmesartan medoxomil on SLNs with a hot homogenization approach increased oral bioavailability by nearly 2.3-fold compared to the marketed formulation of the medication.

Dendrimer

When in solution, dendrimers take on the shape of a circle due to their uniform size, strong branching, and radial symmetry (Graff et al. 2004). Repetitive covalent bond-forming processes build dendrimers layer by layer from core to periphery. Because of the geometric expansion at each branching point, the density of the dendrimers grew with each layer generated in each phase (Tomalia 2005). When it comes to medication delivery, the SLNs have an edge since they can target secondary and tertiary stages of absorption with hydrophobic compounds. PEG polymer-functionalized SLNs were used by Matthias to transport PTX to tumors in lung tissue; as described by Matthias and coworkers for M109-HiFR cells, the PTX inhibitory dose was dramatically lowered by this compound in vitro. Oral bioavailability of the medication olmesartan medoxomil was shown to be about 2.33 times greater than that of the commercial version when it was loaded onto SLNs through heat homogenization in another research (Smith and Diederich 2000).

Dendrimer application in the drug delivery incorporates some polymers involving PEG, poly(L-glutamic acid) (PG), polyamidoamine (PAMAM), polypropylene imine, polyethylenimine, poly(ethylene glycol), chitin, etc. The terminal groups of the dendrimers play a key role in drug-loading capacity. The one-pot divergent synthetic routes frequently provide water-soluble, biocompatible, and more biodegradable green types of dendrimers that are synthesized under supercritical carbon dioxide ($scCO_2$). The models of interaction between drugs and dendrimers are illustrated in Fig. 3 (Yoon et al. 2005).

Liquid Crystals

Dendrimers are made up of layers of covalent bonds that are formed layer by layer from the center to the perimeter. Because of the geometric expansion at each branching point, the dendrimer density increased with each layer generated in each phase (Nath and Banerjee 2013).

Metallic NPs

Metallic and their oxide-based NPs, which involve Ag, Au, etc. and ZnO, Fe_2O_3, etc., are applied in pharmaceutical formulation. For example, AuNPs are widely utilized in anticancer applications, or AgNPs recently are found utilized in wound healing and dressing implants due to antimicrobial properties of such materials. Also, similar features were reported for PtNPs and PdNPs. SeNPs and CuNPs are widely used in antibacterial agents and in cosmetic formulations. Also, ZnONPs showed great antimicrobial disinfectant features (Gericke and Pinches 2006). For example, Thirumurugan and coworkers proposed a

reported biological synthesis route for PtNPs and AgNPs by applying neem extracts and curry leaves. The encapsulation process of DOX was implemented, and under in vitro conditions, the anticancer properties were considered against MCF-7 cells (Table 1).

Use of Bacteria to Synthesize Nanoparticles

Bacteria play an important role in metal biogeochemical cycling and mineral formation in both surface and subsurface settings. One of the most effective ways to make metal nanoparticles is to use microorganisms to create them (see Fig. 5). Microbes' capacity to remove and/or accumulate metals is exploited in commercial biotechnological processes, including bioleaching and bioremediation, despite the fact that synthesizing nanoparticles has only recently become a focus of attention. A variety of inorganic chemicals can be produced by bacteria, both inside and outside of the cell. Gold, silver, and cadmium sulfide nanoparticles may be produced in a biofactory using microorganisms (Lowenstam 1981; Gericke and Pinches 2006).

Among microorganisms, prokaryotic bacteria have received the most attention for their ability to produce metal nanoparticles. Metal nanoparticles have been reported to be produced by bacteria such as *Escherichia coli*, *Pseudomonas stutzeri*, *Pseudomonas aeruginosa*, *Plectonema boryanum*, *Salmonella typlus*, *Staphylococcus currens*, *Vibrio cholerae*, and others (Klaus et al. 1999).

Synthesis of Nanoparticles Using Actinomycetes

Like fungus and prokaryotes, such as bacteria, actinomycetes share many characteristics. They were formerly classed as ray fungus, despite the fact that they are now classified as prokaryotes. Actinomycetes have received a lot of attention because of their outstanding ability to create secondary metabolites like antibiotics. When exposed to gold ions under alkaline circumstances, a unique alkalothermophilic actinomycete, *Thermomonospora* sp., produced gold nanoparticles extracellularly (Syed and Ahmad 2012).

Use of Fungi to Synthesize Nanoparticles

Fungi play an important role in the production of MNPs. A variety of fungi have been used in the biosynthesis of nanoparticles, and the molecular features controlling nanoparticle production have also been described for a number of them. Fungi can also produce nanoparticles with well-defined dimensions in addition to monodispersity. In comparison to bacteria, fungi might be employed to produce enormous quantities of nanoparticles. This is because fungi release more proteins, which directly translates to better nanoparticle production productivity (Balaji et al. 2009).

Nanoparticles may be made using yeast, a fungus that belongs to the *Ascomycetes* class of fungi. Inside the cell, the *Verticillium luteoalbum* fungus was employed to synthesize gold nanoparticles. Physical elements such as pH, temperature, metal concentration (gold), and exposure time can influence nanoparticle size and production rate. A biological process would greatly benefit from being able to precisely regulate the shape of the particles. Yeast, a member of the *Ascomycetes* class, has been found to have considerable potential for the synthesis of nanoparticles. Newly found CdS nanocrystals can be made by *Schizosaccharomyces pombe* cells. As the yeast grows exponentially, Cd has an impact on the organism's metabolic processes (Das et al. 2009).

Use of Plants to Synthesize Nanoparticles

A wide range of chemicals found in plants can assist in the reduction of nanoparticles since they are widely available and safe to handle (Gardea-Torresdey et al. 2002).

Nanoparticles can be formed by a variety of plants, now under investigation. Gold nanoparticles with a size range of 2–20 nm were synthesized using live alfalfa plants. *Brassica juncea*, *Medicago sativa* (alfalfa), and *Helianthus annuus* plants have also been shown to contain nanoparticles of silver, nickel, cobalt, zinc, and copper (sunflower). Hyperaccumulators are plants that accumulate larger

Green Nanomaterials for Drug Delivery, Table 1 Details on types of synthesis, advantages, and disadvantages of physical and chemical methods

Type of physical and chemical methods	Types of synthesis	Advantages	Disadvantages
High-energy ball milling[a]	–	Affordable. It can be combined with chemical treatments to obtain desired product with minimal effort	Noise pollution, requires high amount of energy
Laser ablation[a]	–	Nanoparticles in stable form without the usage of any capping or stabilizing agent can be synthesized	A high amount of energy is required – Low productivity
Electrospraying[a]	–	A most efficient way to produce solid-lipid nanoparticles for drug Delivery	Material loss to the surrounding resulting in lower yields The high amount of energy required
Inert gas condensation[a]	–	The size of the particles synthesized can be controlled effectively	Time-consuming hence not suited for industrial-level production
Chemical vapor synthesis[b]	Vapor-phase synthesis	Multicomponent nanoparticles can be formed by using multiple precursors	Hazardous byproducts are formed. Some of the reactions entail high temperatures
Sol-gel process[b]	Liquid-phase Synthesis	Reactions can be carried out at low temperatures	Organic solvents used are harmful
Hydrothermal process[b]	Liquid-phase Synthesis	Materials that cannot be synthesized using solid-phase synthesis can be synthesized using this method	Involves high temperature and pressure. Fatal accidents may occur if not handled properly
Mechanochemical[b]	Solid-state chemical synthesis	Materials with mean particle size as small as 4 nm, showing low agglomeration and uniformity of crystal structure and morphology, can be synthesized	High energy is consumed
Thermomechanica[b]	Solid-state chemical synthesis	The reactions can be easily Controlled	Requires high temperature

Source: Kiriyanthan et al. (2021)
[a]nanoparticles synthesized by physical methods; [b]nanoparticles synthesized by chemical methods

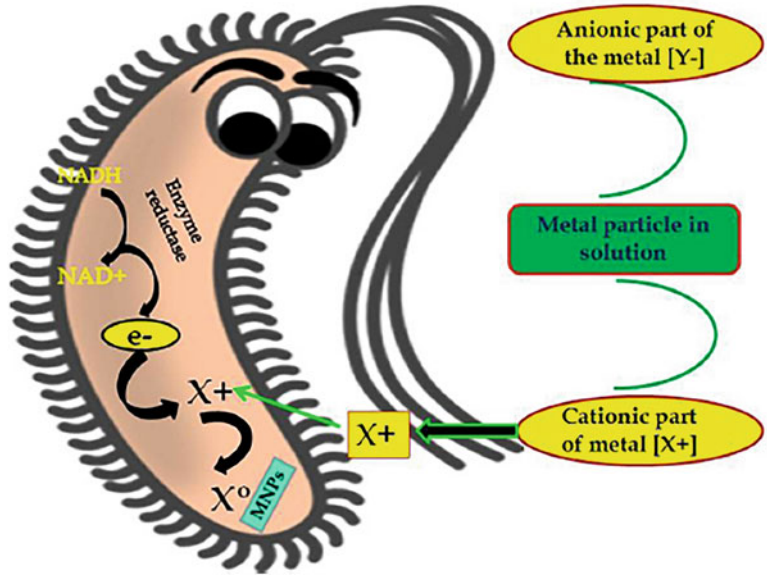
quantities of metals than ordinary plants. In the study, *B. juncea* was shown to be the most capable of collecting metal and subsequently absorbing it as nanoparticles (Song et al. 2009).

Applications of Metal Nanoparticles in Medical Biology

Plants are shown schematically as a biological source of green nanoproduction, its characterization, and biomedical applications. Metal nanoparticles have gotten a lot of attention in the ever-expanding field of nanomaterial research because of its wide range of applications in catalysis, electronics, sensing, photonics, environmental remediation, imaging, and drug delivery (Guo et al. 2005).

Conclusion

Green nanotechnology is an effective method for producing nanoparticles that can be easily separated without the use of harmful reagents. Nanotechnology advancements are spreading their potential into the realm of environmental remediation and protection. The demand for nanoscale materials, gadgets, and tools is growing every day across all industries, including the pharmaceutical business. However, some aspects of nanoparticle bioavailability, bioaccumulation, transport, toxicity, and the environmental and health effects of nanoparticle manufacture need to be better understood. Nanotechnology incorporates green



Green Nanomaterials for Drug Delivery,
Fig. 5 Bacteria may be able to synthesize metal nanoparticles using this hypothetical method

manufacturing practices into the design process, removing contaminated waste products from the equation. More research into the effective production of nanoparticles is needed to use these nanoscale particles for purification and catalysis for environmental remediation. As a result, green nanotechnology's future goals appear optimistic and good. Green nanotechnology will play a key role in balancing sustainable approaches and hazardous agents in the future.

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