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Review article

A review on green synthesis of zinc oxide nanoparticles – An eco-friendly approach

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ABSTRACT

Nanotechnology deals with the production and usage of material with nanoscale dimension. Nanoscale dimension provides nanoparticles a large surface area to volume ratio and thus very specific properties. Zinc oxide nanoparticles (ZnO NPs) had been in recent studies due to its large bandwidth and high exciton binding energy and it has potential applications like antibacterial, antifungal, anti-diabetic, anti-inflammatory, wound healing, antioxidant and optic properties. Due to the large rate of toxic chemicals and extreme environment employed in the physical and chemical production of these NPs, green methods employing the use of plants, fungus, bacteria, and algae have been adopted. This review is a comprehensive study of the synthesis and characterization methods used for the green synthesis of ZnO NPs using different biological sources.

1. Introduction

Nanomaterials are particles having nanoscale dimension, and nanoparticles are very small sized particles with enhanced catalytic reactivity, thermal conductivity, non-linear optical performance and chemical steadiness owing to its large surface area to volume ratio [1]. NPs have started being considered as nano antibiotics because of their antimicrobial activities [2]. Nanoparticles have been integrated into various industrial, health, food, feed, space, chemical, and cosmetics industry of consumers which calls for a green and environment-friendly approach to their synthesis [3].

1.1. Nanoparticle synthesis methods

Two approaches have been suggested for nanoparticle synthesis: Bottom up and top down approach. The top–down approach involves milling or attrition of large macroscopic particle. It involves synthesizing large-scale patterns initially and then reducing it to nanoscale level through plastic deformation. This technique cannot be employed for large scale production of nanoparticles because it is a costly and slow process [4]. Interferometric Lithographic (IL) is the most common technique which employs the role of top–down approach for nanomaterial synthesis [5]. This technique involves the synthesis of nanoparticles from already miniaturized atomic components through self-assembly. This includes formation through physical and chemical means. It is a comparatively cheap approach [6]. It is based on kinetic and thermodynamic equilibrium approach. The kinetic approach involves MBE (molecular beam epitaxy).

1.2. Different methods used in nanoparticle synthesis

In the physical method, physical forces are involved in the attraction of nanoscale particles and formation of large, stable, well-defined nanostructures. Its example includes nanoparticle synthesis through colloidal dispersion method. It also includes basic techniques like vapor condensation, amorphous crystallization, physical fragmentation and many others [7–10]. Nanoparticle synthesis is mediated by physical, chemical and green methods [11–13]. The physical method involves the use of costly equipment, high temperature and pressure [14], large space area for setting up of machines. The chemical method involves the use of toxic chemicals which can prove to be hazardous for the environment and the person handling it. The literature states that some of the toxic

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chemicals that we use in physical and chemical methods may reside in the NPs formed which may prove hazardous in the field of their application in the medical field [15]. Thus, we needed an environment-friendly and cost-effective method for nanoparticle synthesis. Physical process involves the use of high vacuum in processes like pulsed laser deposition, MBE (molecular beam epitaxy), thermal evaporation etc. [16] and chemical methods include chemical micro emulsion, wet chemical, spray pyrolysis, electrodeposition [16], chemical and direct precipitation and microwave assisted combustion [17]. Additional capping and stabilizing agent are needed in physical and chemical methods [18–21].

1.3. Green approach

Biosynthesis of nanoparticles is an approach of synthesizing nanoparticles using microorganisms and plants having biomedical applications. This approach is an environment-friendly, cost-effective, biocompatible, safe, green approach [22]. Green synthesis includes synthesis through plants, bacteria, fungi, algae etc. They allow large scale production of ZnO NPs free of additional impurities [23]. NPs synthesized from biomimetic approach show more catalytic activity and limit the use of expensive and toxic chemicals.

These natural strains and plant extract secrete some phytochemicals that act as both reducing agent and capping or stabilization agent; for example, synthesis of ZnO nanoflowers of uniform size from cell soluble proteins of *B. licheniformis* showed enhanced photocatalytic activity and photo stability clearly depicted by 83% degradation of methylene blue (MB) pollutant dye in presence of ZnO nanoflowers considering the fact that self-degradation of MB was null (observed through the control value) and through three repeated cycles of experiment at different time interval, degradation was found at 74% which clearly showed photo stability of ZnO nanoflowers produced [24]. Oblate spherical and hexagonal shaped ZnO NPs of size ranging from 1.2 to 6.8 nm have been synthesized using fungal strain Aspergillus fumigatus TFR-8 and these NPs showed stability for 90 days confirmed by measuring hydrodynamic diameter of NPs using particle size analyzer which showed agglomeration formation of NPs only after 90 days suggesting high stability of NPs formed using the fungal strain [25]. ZnO NPs of size 36 nm synthesized from seaweed *Sargassum myriocystum* (microalgae) obtained from the gulf of Mannar showed no visible changes even after 6 months clearly demonstrating the stability of NPs formed. From FTIR result studies, it has been confirmed that fucoidan soluble pigments secreted from microalgae were responsible for the reduction and stabilization of the NPs.

Plant parts like roots, leaves, stems, seeds, fruits have also been utilized for the NPs synthesis as their extract is rich in phytochemicals which act as both reducing and stabilization agent [26–32]. ZnO NPs synthesized from *Trifolium pratense* flower extract showed similar peaks in UV-Vis spectrophotometer after 24, 48, 72, 96 and 120 hours of NPs formation showing the stability of NPs formed [33]. Similarly, fruit extract of Rosa canina acted as both reducing and stabilizing agent for synthesized ZnO NPs, confirmed by FTIR studies. Bio-capping is done by carboxylic and phenolic acid present in fruit extract. Spherical shaped ZnO NPs were formed by Aloe Vera leaf extract where free carboxylic and the amino group of plant extract acted as both reducing and capping agent.

1.4. Zinc oxide nanoparticles

ZnO is an n-type semiconducting metal oxide. Zinc oxide NP has drawn interest in past two–three years due to its wide range of applicability in the field of electronics, optics, and biomedical systems [34–40]. Several types of inorganic metal oxides have been synthesized and remained in recent studies like TiO_2, CuO, and ZnO. Of all these metal oxides, ZnO NPs is of maximum interest because they are inexpensive to produce, safe and can be prepared easily [41]. US FDA has enlisted ZnO as GRAS (generally recognized as safe) metal oxide [42]. ZnO NPs exhibit tremendous semiconducting properties because of its large band gap (3.37 eV) and high exciton binding energy (60meV) like high catalytic activity, optic, UV filtering properties, anti-inflammatory, wound healing [43–49]. Due to its UV filtering properties, it has been extensively used in cosmetics like sunscreen lotions [50]. It has a wide range of biomedical applications like drug delivery, anti- cancer, anti-diabetic, antibacterial, antifungal and agricultural properties [51–55]. Although ZnO is used for targeted drug delivery, it still has the limitation of cytotoxicity which is yet to be resolved [56]. ZnO NPs have a very strong antibacterial effect at a very low concentration of gram negative and gram positive bacteria as confirmed by the studies, they have shown strong antibacterial effect than the ZnO NPs synthesized chemically [57–59]. They have also been employed for rubber manufacturing, paint, for removing sulfur and arsenic from water, protein adsorption properties, and dental applications. ZnO NPs have piezoelectric and pyroelectric properties [60,61]. They are used for disposal of aquatic weed which is resistant to all type of eradication techniques like physical, chemical and mechanical means [62]. ZnO NPs have been reported in different morphologies like nanoflake, nanoflower, nanobelt, nanorod and nanowire [63–65].

2. Literature study

Due to the increasing popularity of green methods, different works had been done to synthesize ZnO NPs using different sources like bacteria, fungus, algae, plants and others (Fig. 1). A list of tables had been put to summarize the valuable work done in this field.

2.1. Green synthesis of ZnO NPs using plant extract

Plant parts like leaf, stem, root, fruit, and seed have been used for ZnO NPs synthesis because of the exclusive phytochemicals that they produce. Using natural extracts of plant parts is a very eco-friendly, cheap process and it does not involve usage of any intermediate base groups. It takes very less time, does not involve usage of costly equipment and precursor and gives a highly pure and quantity enriched product free of impurities [66]. Plants are most preferred source of NPs synthesis because they lead to large-scale production and production of stable, varied in shape and size NPs [67]. Bio-reduction involves reducing metal ions or metal oxides to 0 valence metal NPs with the help of phytochemicals like polysaccharides, polyphenolic compounds, vitamins, amino acids, alkaloids, terpenoids secreted from the plant [66,67].

Most commonly applied method for simple preparation of ZnO NPs from leaves or flowers is where the plant part is washed thoroughly in running tap water and sterilized using double distilled water (some use Tween 20 to sterilize it). Then, the plant part is kept for drying at room temperature followed by weighing and then crushing it using a mortar and pestle. Milli-Q H_2O is added to the plant part according to the desired concentration and the mixture is boiled under continuous stirring using a magnetic stirrer [66–70]. The solution is filtered using Whatman filter paper and the obtained clear solution was used as a plant extract (sample). Some volume of the extract is mixed with 0.5 Mm of hydrated Zinc nitrate or zinc oxide or zinc sulfate and the mixture is boiled at desired temperature and time to achieve efficient mixing [69,70]. Some perform optimization at