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Zinc Oxide Nanostructures: A Review

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Abstract
The structure and unique properties of zinc oxide makes it indomitable among all the nanostructures. Nanostructures are classified based on their dimensions, origin, chemistry and counterparts types. Nanostructures show solitary and exclusive properties that are not present in their bulk counterparts. Depending on their state i.e. solid, liquid or gas; it can be characterized by various techniques. Due to its unique properties; zinc oxide nanostructures show a wide range of applications in biomedical, agriculture, cosmetics, food packaging, electronics, construction, textiles etc. Humans may suffer from potential health issues at their work places if not taken proper precautions.

Keywords: *Zinc oxide nanostructures, Characterization, Doping, Applications, Toxicity*

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I. Introduction:

To be defined as a nano-object only one of its characteristic dimensions to be in the range of 1 – 100 nm. In 2008, the International Organization for Standardization defined a nanoparticle as a discrete nano-object where all three Cartesian dimensions are less than 100 nm. Similarly, the ISO also defined the two-dimensional nano-objects i.e. nanodiscs and nanoplates and one-dimensional nano-objects i.e. nanofibres and nanotubes. It exhibit unique physicochemical properties that are not present in their bulk counterparts. There is an exception to the 100 nm rule i.e. solid-lipid nanoparticle that exhibit the unique nanoparticle related properties at diameters greater than 100 nm[1]. Nanostructures are of considerable scientific interest as they are constructively a bridge between bulk materials and atomic and molecular structures. Nanostructures can be classified according to their dimensions (zero-dimensional, one-dimensional, two-dimensional and three-dimensional particle); origin (originated from forest, fires, volcanic eruptions, lightning); chemistry (metals / metal oxides, DNA and other biological materials, carbon, polymers and clays); counterpart types and application into different classes[2]. Nanoparticles exhibit exceptional mechanical, optical, electronic, magnetic and thermal properties[3].

1.1 Metal Nanoparticles

Nano size inorganic compounds have shown remarkable biological activities at very low concentrations due to their high surface area to volume ratio. Metal, semiconductor and quantum dot nanoparticles have been extensively used as functional materials in chemistry, biology and physics. They show enormous chemical diversity with unique beneficial and adverse effects that may be related to their structure and synthetic procedure. Metallic nanoparticles have number of applications in pharmaceutical and biomedical areas as alternative antimicrobial strategy due to upsurge of many infectious diseases and emergence of antibiotic resistant strains[4]. Valence electrons present in the transition metals are in different orbitals and exhibit several common oxidation states[2]. Among the various metal nanoparticles, zinc oxide nanoparticles have gained the attention of many researchers for its divergent properties. Zinc oxide nanoparticles belong to the class of metal oxides, which is characterized by photocatalytic and photo-oxidizing capacity against chemical and biological species[5]. In nanoscale metal oxide, zinc oxide is a common hosted material that has been widely used for its perceptible properties.

1.2 Zinc Oxide Nanoparticles

In the recent years, there has been a significant emphasis on nano – sized semiconductors because of their unexpected visible properties which carries wide range of applications[6]. Zinc blende, Wurtzite and Rocksalt (or Rochelle salt) are the known crystal structures of zinc oxide. Zinc blende structure is also known as Sphalerite. Wurtzite is thermodynamically stable at ambient temperatures. The lack of centre of symmetry combined with large electromechanical coupling, results in strong piezoelectric and pyroelectric properties. The zinc blende structure can only be stabilized by growth on cubic substrate and the Rochelle salt structure may be obtained at relatively high pressures. Zinc oxide is a versatile functional material that has a varied group of growth morphologies namely, nanorings, naowires, nanobelts, nanohelixes, nanocages, and nanocombs[7]. Nanostructured zinc oxide materials have been received broad attention due to their distinguished accomplishment in optics, electronics and photonics.

Zinc is a metallic element having atomic number 30 and stable isotopes of mass number 66, 67, 68, and 70, hence averaging 65.38 atomic mass unit. The terrestrial chemistry of Zn is that of Zn (II) rather than Zn (0). The Zn (II) ion has an electron configuration of $1s^2$, $2s^2$, $2p^6$, $3d^{10}$, and it lacks unfilled *d* sub-shells in the well-known oxidation state, the requisite criterion for true transition metals. Since, zinc is required as either a structural component or reaction site in numerous proteins, the zinc-binding portions of which are highly conserved among species make it important element for terrestrial life. Zinc sites in proteins comprise of zinc polyhedra with apical S, N, or O, associated with cysteine, histidine, glutamic acid, aspartic acid, and water[8]. ZnO is an *n*-type semiconductor (means it has few extra electrons which allow it to conduct) by virtue of the electrons excited from ionized zinc interstitials. At 25 °C, the electrical resistivity is typically 0.01 Ω m. The resistivity can be decreased by quenching from high temperatures or by heat treatment in an appropriate environment to increase the concentration of interstitial zinc[9]. Due to its unique physical and chemical properties such as high chemical stability, high electrochemical coupling coefficient, broad range of radiation absorption, paramagnetic nature and high photo-stability, make it a multifunctional material[10].

ZnO is a wide band-gap (3.37eV) compound semiconductor that is expedient for various types of applications like power generators, ultraviolet lasers, solar cells, gas sensors, capacitors, field emission devices, photocatalysts, electrophotography, photoprinting, transparent UV resistance coating, electrochemical and electromechanical nanodevices, sunscreen lotion, cosmetics, anti-hemorrhoids, eczema, antibacterial agent, excoriation in the human medicine[11-16]. It is also used as an additive in various materials and products like ceramics, adhesives, ointment, lubricants, glass, rubber, plastics, pigments, sealants, paints, batteries, fire retardants[17-18] etc. To date, it has been crucially versatile material in nanotechnology applications.

II. Zinc oxide nanostructures and their characterization

Due to the ease of fabrication, a wide array of methods enables the fabrication of nanostructures with many different shapes and sizes. As their name suggests their morphology, zinc oxide nanostructures can be included in variety of morphologies, such as nanowires, nanotubes, nanoflowers, nanoribbons, nanocages, nanoplates, nanodendrites, nanorings, nanosheets, nanorods, nanospheres, nanocorals, nanocombs, nanosprings, nanoshells and more are achievable. If we consider the literature report, zinc oxide nanostructure formation is based on chemical synthesis approach. Depending on the embodiment of the applied technique, different morphologies can be obtained by controllable synthesized parameters, such as the type of reactants, alkalinity, temperature, type of solvents and its water content[19]. The different surface structures of zinc oxide could induce anisotropic growth. By modifying the composition of source materials, morphology of the grown oxide nanostructure can be drastically changed.

Under thermodynamic equilibrium conditions, the facets with higher surface energy is normally small in area and the lower energy facets are larger. In determining the nanostructures obtained, it is important to have control over growth kinetics. One of the examples to show the crucial role played by the growth kinetics in nanostructure formation is Zn-ZnO core shell structure[20]. With a variety of different techniques and materials sensitive to fabrication conditions, their properties differ significantly. Even with the same morphology, but used different fabrication approach, the nanostructure properties can differ due to different levels of background impurities[21].

The potential and application of a nanostructure is determined by its unique characteristics. It is measured by various characterization techniques depending on their states i.e. solid, liquid or gaseous phase. The size of the particle can be measured by scanning electron microscope (SEM) or transmission electron microscope (TEM) if the samples are in solid phase[22]. Photon correlation spectroscopy and centrifugation for liquid phase and scanning mobility particle size (SMPS) is used to characterize in gaseous phase. When it comes to surface area characterization, BET analysis for solids, simple titration or nuclear magnetic resonance (NMR) for liquids and SMPS or differential mobility analyser (DMA) for gaseous phase. By using X-ray photoelectron spectroscopy, the characterization of composition can be carried out[23]. Zeta potentiometer is used for measuring its surface charge for solids and liquids whereas electron microscopy is used for gaseous phase samples. The crystallography of nanoparticles are carried out by a powder X-ray, electron or neutron diffraction to determine the structural arrangements[24].

III. Doping of ZnO Nanostructures

The performance and properties of ZnO nanostructures are chiefly dependent on sizes, defects and doping of zinc oxide. During the synthesis of zinc oxide nanostructures, dopants are generally introduced by controlling their formation. It is well established that doping affects the basic physical properties like electrical, optical and magnetic properties. So, a wide range of properties like piezoelectricity, room temperature ferromagnetism, high transparency, wide bandgap semiconductivity, huge magneto-optic, chemical-sensing and a range of conductivity from metallic to insulating (including n-type and p-type conductivity) are significantly altered[25]. Small amounts of dopants are sufficient to acts as donors or acceptors inside the semiconductor crystal lattice which will remarkably amend the properties of semiconductor up to a greater extent[26]. There are two categories basis on which the dopants function: (i) doping with donor or acceptor impurities to achieve high n-type or p-type conductivity; (ii) doping with transition metals or rare earth metals to achieve the desired semiconductor properties. L. Armelao suggest that the rare earth dopants most likely affect the energy transfer and structural changes by nucleation, rather than direct radiative decay from Eu sites[27].

Literature reports reveal that doping of zinc oxide structure with metals and non-metals leads to a band gap red-shift to to produce visible-light-active photocatalysts[28]. In metal doping, metal cations get replaced by Zn^{+2} ions, which leads to the reaction of dopant cations state or creation of intra-band gap levels. Though these new band states extend response in visible light range; depending on the nature of the metal dopants and its concentrations, they can act as recombination centers to decrease the photocatalytic activity. When non-metal dopants are used, the anionic dopants are suppose to replace the oxygen atoms, the valence bond maximum elevates upward to shift the optical respose towards visible light absorption. Hence, doping acts as a significant route to change the microstructures and practical performance of zinc oxide.

IV. Applications

The versatility of physical and chemical properties and ease of formation of zinc oxide nanostructures make it a suitable and interesting candidate for researchers. It plays an important role in wide range of applications; some of them are discussed below:

4.1 Biomedical applications

The main objective of nanoparticle is to manage and influence biomacromolecular constructs and supramolecular assemblies that play crucial role in living cells, in order to improve quality of human health. Proteins, DNA/RNA, cellular lipid bilayer, cellular receptor sites and antibody variable regions critical for immunology and are engaged in events at nanoscale proportions[29]. Targeting the drug to specific site of body, bioavailability can be improved by enhancing aqeous solubility, smaller drug dose, retention of the drug at the active site and increasing half life of its clearance or increasing resistance time in the body, are the main advantages of using nanoparticle[30]. Zinc oxide nanostructures have a dominant position in transforming therapeutics. Utilising inorganic nanostructures with entrapped biomolecules have feasible applications in many frontiers of modern materials science including drug delivery system. The types of nanostructures applied in the drug delivery system includes: polymeric nanoparticles, carbon nanotubes, nanosuspensions and nanocrystals, magnetic nanoparticles, solid lipid nanoparticles, polymeric micelles etc[31]. The traditional treatments like organ transplants and artificial implants can be carried out by tissue engineering. In the era of drug therapy nanostructures have made greatest impact over new modalities for chronic disease treatment. Ferial Ghaemi[32] and his co-authors have very well summarized the role of different types of nanomaterials against, diagnosis, prevention and therapy of covid-19.

It is one of the most promising areas of the nanostructures where metal oxide devices are well established. There are numerous uses of nanostructures in medical diagnostics due to its great sensitivity, specificity, portability, reusability and speed with cost effective makes nanostructure based assays an appealing alternative to current diagnostic techniques[33]. Cardiovascular and surgical devices like nano-electromechanical systems to assist surgeons, implantable medical devices containing nano-electronic components diagnostic tools and therapeutic actuating elements are examples of sophisticated devices[34].

Zinc oxide nanoparticles have been highlighted as suitable metal based nanodrugs. They show promising potential as therapeutics with antidiabetic[35], anticancer[36], anti-inflammatory[37], antimicrobial[38] and wound healing activities[39].

4.2 Agricultural applications

There is a wide range of nanoparticulate materials and structures being developed for the agricultural purposes to be used as fertilizers, herbicides, pesticides, sensors and quality stimulants in order to improve the conventional agricultural practices[40]. Due to the factors like drifting, photolysis, hydrolysis, microbial degradation and leaching, the agrochemicals applied to the crops are lost. Nanoparticles and nanocapsules are being used to reduce collateral damage and to provide efficient distribution of fertilizers and pesticides in a controlled manner with high site specificity[41]. Nanosensors can detect soil moisture and soil nutrients. It has been reported that zinc oxide nanoparticles have the potential to boost the yield and growth of food crops[42]. L. M. Batsmanova and his co-workers developed an eco-friendly and cost-effective colloidal solution of metal nanoparticles using Zn, Co, Cu, Mn, Fe, which can be used as fertilizers[43].

4.3 Cosmetics

In cosmetic industry, nanomaterials used in various types and forms. The main advantages of using nanomaterials in cosmetics are: efficient penetration into the skin for the improved delivery of the ingredients of the products, transparency, new colour elements and long lasting effects. The purpose behind using the nanomaterials in cosmetics is to deliver the required amount of ingredient to the desired part of the body and long term of stability[44]. The nanomaterials based cosmetic products are sunscreen containing nanoparticles attenuating UV radiation while simultaneously removing the free radicals generated by UV lights, dental care products, long lasting hair dyes with carbon nanotubes providing smothering, volumizing and anti aging creams with active ingredients embedded in nanoparticles of synthetic polymers acting as skin permeability enhancers[45]. Good texture, better spreadability and enhanced sun protection factor (SPF) proved the zinc oxide nanoparticles desirable product to be used in cosmetic industry[46].

4.4 Food Packaging

As zinc is an essential trace element, zinc oxide is currently listed as generally recognized as safe by the US Food and Drug Administration and used as a food additive. Materials in nanoscale has potential application in food packaging in forms of barrier and mechanical properties, detection of pathogens, smart and active packaging with food safety and quality benefits[47]. In order to provide antimicrobial property to the packaging material, zinc oxide nanoparticles have been incorporated in polymeric matrices which leads to improvement in packaging properties. Mainly by three mechanism i.e. by release of antimicrobial ions, by damaging the integrity of bacterial cell and by the formation of ROS by the effect of light radiation; the nanomaterials have improved the quality of food packaging[48]. Aswathy Jayakumar *et. al*. developed pH sensing PVA- starch films fabricated with nutmeg oil, zinc oxide nanoparticle and jamun extract with enhanced water barrier, UV barrier, mechanical and antimicrobial properties[49].

4.5 Electronics

Zinc oxide nanostructure provides path to a new generation of devices as it is found to be an attractive material for applications in electronic, photonic and spin based devices[50]. As already discussed in the introduction part of this review, zinc oxide is a semiconductor of wide bandgap and therefore interacts with UV light. It is an excellent inorganic UV filter which is used as UV photodetector, in field emission displays as well as electrical and optical devices (light emitting diodes, optically pumped lasers, transparent thin films transistors) and solar cells[51-52]. It is used as an electron injecting layer for OLEDs, as an electron transporting layer (ETL) material as well as to improve surface and electronic properties of direct and inverted OPVs. Thomas R. Andersen *et. al*. investigated the suitability of different inorganic materials as electron transport layers in fully roll-to-roll (RTR) organic solar cells. They found ZnO electron buffer layer as the most promising ETL for roll-to-roll printed photovoltaics[53]. Due to the variety of mesophase morphologies and fast response to external stimuli such as temperature and electric or magnetic fields, make liquid crystals a unique candidate[54]. Studies show thin film with lowest areal density of ZnO nanowires showed better field emission characteristics than the carbon nanotube[55]. A sound driven piezotronic zinc oxide nanowire based nanogenerator obtained an AC output voltage, reported by Seung Nam Cha and his co – workers[56]. By utilizing the coupled piezoelectric and semiconducting dual properties of ZnO, X. Wang et. al. demonstrated a piezoelectric field effect transistor (PE-FET) which was composed of a zinc oxide nanowire or nanobelt[57].

4.6 Construction

The use of nanostructures in construction can improve the construction processes by making them quicker, inexpensive and safe. The nano-enabled products are concrete, glass, coatings, steels, bricks and timber. The advantage of the nanoparticles on the microstructure and performance on cement based materials are due to the fact that nanoparticles fill the voids between cement grains, resulting in the immobilization of free water; favor the small sized crystals; improve the structure of the aggregates' contact zone, resulting in a better bond between agreegates and cement paste; and it also provide crack arrest and interlocking effects between the slip planes, which improve the toughness, shear, tensile and flexural strength of cement based material[58-59]. The effects of zinc oxide nanoparticles on strength assessments and water permeability of concrete in different curing media was studied by Ali Nazari and Shadi Riahi. They found that the specimens containing ZnO nanoparticles have significantly higher strength when compared to specimens without it, at every age of curing[60].

4.7 Textie

Zinc oxide is one of the most promising material for the development of high performance textile products as it has antimicrobial, self cleaning, flame-retardant, thermo-insulating, UV-protective, hydrophobic, electrically conductive properties as well as textile exhibiting moisture management[61]. S. Kathirvelu *et. al*.

reported synthesis and characterization of ZnO nanoparticles and their application on cotton and polyester fabrics for the protection against UV radiation[62]. Self cleaning cotton fabrics were fabricated by coating photocatalytic zinc oxide nanoparticles on cotton surfaces by using a traditional dip-pad-dry-cure coating process, as reported by Chunhong Zhu *et. al*. The self cleaning property of the fabrics are directly dependent on the zinc oxide nanoparticle content; that means high wt% of zinc oxide nanoparticles in the coated fabric resulted in more pronounced photolytic degradation[63]. As wash fastness is correlated with the nanoparticle adhesion to the fabric; nanoparticles can impart high durability for the treated fabrics as they have large surface area and great surface energy that makes it have better affinity for fabrics and lead to an increase in durability of the desired textile functions[64].

4.8 Miscellaneous

Nanofiltration membrane technology is successfully applied to food and dairy industry, biotechnology, water and waste water treatment and pharmaceuticals. It has been applied for a long time in the edible oil processing sector, cheese and sweetner production[65]. ZnO is consumed by the rubber industry to manufacture various cross-linked rubber products[66]. The effect of zinc oxide nanoparticle morphology on activity in crosslinking of carboxylated nitrile elastomer was studied. And it was found that zinc oxide nanoparticles showed greater tensile strength as compared to microsized particles[67]. Contaminated soil is treated using nanoparticle by injecting into specific target locations for heavy metal contamination, toxic industrial wastes etc. Zinc oxide nanostructure showed their promising potential as catalysts for industrial wastewater and soil pollution treatment, studied by C. W. Zou and his co- workers[68]. Since, ZnO nanoparticles are non-toxic, economic and chemically stable in air, it has been identified as a new generation of bio-friendly cell labeling agents[69].

V. Health hazards and Toxicity

The hazardous properties of the nanoparticles and their implication within workplace safety should be investigated. Because of the increased mobility of the nanoparticles, it can travel to a great distance in air. Three main routes through which nanoparticles can enter into our body are absorption through the dermal system, inhalation into the pulmonary system and ingestion through the gastrointestinal system. It may cause potential health issues such as lung cancer, heart diseases, asthma, bronchitis, emphysema, Alzheimer's disease, Parkinson's disease, liver cancer, Crohn's diseases and many more. Through any route of exposure, nanoparticles have the tendency to accumulate in specific part of brain, where they can access the neural cells[70]. The adverse effect on the structure, function and chemistry of the central nervous system as a consequence of biological, chemical and physical influences is known as neurotoxicity. The mechanism of neurotoxicity induced by nanoparticles are astrogliosis, oxidative stress, cognitive dysfunctions, neuroinflammation, synaptic plasticity, microglial function alterations and disrupted signalling pathways. Research is required to understand the relationship between nanomaterial characteristics and their potential biological impacts; a thorough evaluation is crucial for designing safer nanocarrier systems in order to reduce their side effects[71].

VI. Discussions & conclusions

Due to tremendous research work on zinc oxide nanomaterials make it really difficult to summarize the work in a systematic way. A comprehensive review of synthesis, properties, doping, characterization, application and toxicity of zinc oxide nanostructures have been reviewed in this article. Both in structures and in properties, the unique nanostructures reveal that zinc oxide probably has the richest family of the nanostructures among all the materials. Hence, it is advantageous to choose zinc oxide nanomaterials among the variety of nanomaterials as it shows extraordinary physical and chemical properties and enables development of innovative applications in all fields of expertise. As compared to other wide band-gap semiconductors, zinc oxide is more favourable because it lends itself smoothly to the production of nanostructures from which functional devices have already been fabricated. Use of nanomaterials in biomedical applications is one of the most effective ways to control this viral pandemic outbreak. Nanomaterials based technologies has provided platforms and tools for understanding, protection, detection and treatment of SARS-CoV-2 as well as future viral diseases. Zinc oxide nano size counter parts appear to be a better alternative in many industrial processes where a combination of small size and high surface area proves to be more advantageous.

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Conflict of Interest

The authors declare that there is no conflict of interest regarding the publication of this article.

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