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Green Synthesis of Oxide Nanoparticles in Water and Ethanol at Low Energies for Photodetector Applications

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Introduction

In recent years, oxide nanoparticles have attracted great interest because of their multidimensional applications, especially in the development of optoelectronic devices such as photodetectors which demand high standards such as low noise, fast response times and high sensitivity at operational wavelengths [1]. The applications of these materials are widely spread in varied disciplines such as heterogeneous catalysis, biotechnology, medicine, photonics, solar energy conversion, and microelectronics [2].Metal oxides nanoparticles (ZnO, SnO2, Fe2O3) are especially useful to such applications, because they have a high surface to volume ratio, can be tuned to optical properties, and are exceptionally chemically stable [3], [4], [5].One such material, which has come up as a promising photodetector technology, is Zinc oxide nanoparticles due to their broad direct bandgap and high electron mobility [6], [7]. ZnO has high excitation binding energy which in combination with its high thermal stability has made it suitable in applications of UV light sensors and detectors [8]. The traditional physical and chemical synthesisroutes to these nanoparticles tend to be labor-intensive, consume a lot of energy, and use toxic reagents, and hence there is need to look at other sustainable alternatives that are cost-effective and environmentally friendly[9], [10]. Green synthesis methods are also sustainable development targets as they work with biocompatible materials, reducing toxicity, thus being a less risky alternative to conventional reducing agents, such as sodium citrate and sodium borohydride, that have dangerous physical characteristics [11], [12]. These environmental-friendly procedures make use of natural resources, such as extracts of plants and microorganisms to act as reducing and stabilizing agents throughout the synthesis process which has considerable benefits, which are low cost, nontoxicity, scalability, and the ability to give nanostructures the desired size, shape and morphology [11], [13]. In particular, the plant-mediated synthesis that leveraging bioactive compounds, such as polyphenols, flavonoids, and terpenoids, is a potential reducing and capping agent to counteract the nucleation and growth dynamics without hazardous chemicals [11], [14], [15]. It is a bio-mediated technique that is able to remove the environmental risks of the traditional chemical techniques as well as precisely control the texture and morphology of the particles at ambient temperatures [16], [17]. Moreover, aqueous solvents and ethanol solvents used in these green protocols can lead to the generation of nanoparticles having a high degree of biocompatibility and lower toxicity profiles that are important in the fabrication of photodetectors where the level of impurity can have a profound effect on the performance and reliability of the device [18], [19]. Thus, the optimization of the solvent composition and the energy input is an important field of research to adjust the optoelectronic characteristics of oxide nanoparticles to achieve excellent photodetection parameters. The elimination of undesirable environmental impacts generally linked with high-energy chemical processes is a core benefit of these low-energy green synthesis methods where in most cases use mild reaction conditions for facilitating the nanostructures' formation through natural biochemical pathways [15], [19].

Literature Review

The shift toward environmentally benign fabrication techniques was catalyzed through the sense of urgency to reduce the footprints of the traditional nanomanufacturing processes, which often involve hazardous precursors as well as energy-consuming procedures for producing metal oxide nanoparticles [20]. This poses serious concerns on their environmental and bio-compatibility and hence the use of green and bio-based synthesis methods that use non-toxic solvents and benign reaction conditions need to be adopted [21], [22]. Green synthesis processes employ natural, renewable, and non-toxic starting materials and in many cases use aqueous solvents which minimize the amount of dangerous chemicals, solvents, and waste as compared to more traditional methods of synthesis [23], [24]. Plants, fungi, bacteria and algae are all biological agents and this means that they make good solvents and stabilizing agents building a sustainable synthetic pathway that does not only cut down on environmental pollution but also convert heavy metals from industrial waste into safer compounds [25].

These sustainable methods have been associated with such benefits as low cost and the possibility of mass production without excessive use of toxic organic solvents and reducing agents that harm the environment and health of the population with these green-principled approaches [19], [21]. Such approaches must prevent the production of undesirable or harmful by-products by accumulating credible and sustainable and environmentally friendly synthesis processes [26]. Plants, algae and fungi provide biological extracts which contain secondary metabolites in the form of alkaloids and flavonoids; these extracts are often used as reducing agents and capping agents which reduce wastes generated and toxic substances are not used [27], [28], [29]. Green chemistry principles can be applied to create the biodegradable, sustainable and environmentally friendly nanomaterials because such materials are based on the renewable resources to reduce the use of the hazardous chemicals [25], [30]. In addition, biomolecules and phytochemicals that are extracted out of plants serve as active compounds as reducing and stabilizing agents, and the general procedure provides less energy requirements and less cost of production in contrast to the traditional synthesis [25], [31]. Moreover, the financial viability of these green procedures can be seen in the fact that they can use feedstocks that are renewable, and they do not need costly purification processes that may be part of most traditional pathways to eliminate toxic residues [25], [32].

Methodology

In this paper, a green synthesis methodology that used aqueous and ethanol extract of chosen plant species was used to enable the zinc oxide nanoparticles to be synthesized using low energy that enables its application in photodetectors. These extracts contain phytochemical constituents known as polyphenols and flavonoid which serve as dual-purpose reducing and capping agents to aid the generation of stable nanoparticles at ambient temperatures [25], [33]. The synthesized nanoparticles were also characterized through UV-Vis spectroscopy, XRD, and the FE-SEM to determine optical characteristics, crystalline structure, and surface structure [25], [34]. These properties were analyzed and confirmed the successful forming pure zinc oxide phases and gave information about the effect of solvent polarity on particle size distribution and optical band gap energy [11], [35]. Optical measurements showed that there was a clear ultraviolet absorption edge which was an indication that there are quantum confinement effects which are typical of nanoscale ZnO. The analysis of FE-SEM showed that the synthesized nanoparticles under aqueous media had a spherical morphology with an average particle size of about 20-30 nm, and those under ethanol medium had slightly elongated shapes with an average particle diameter of about 25-40 nm indicating the effect of the solvent polarity to the growth kinetics and final morphology. FE-SEM analysis revealed that the nanoparticles produced under aqueous conditions were found to have a spherical morphology with an average particle size of about 20-30 nm and those produced under ethanol conditions had slightly elongated morphology with an average particle size of about 25-40 nm, showing the effect of solvent polarity on the growth kinetics and the final morphology [33], [36]. Also, the elemental composition analysis through energy-dispersive X-ray spectroscopy was performed to determine the high purity of the samples synthesized since only Zinc and oxygen signals showed without any significant impurity peaks. The presence of a hexagonal wurtzite structure was then supported with X-ray diffraction analysis that revealed clearly defined peaks on the characteristic planes which are indicative of a high level of crystallinity [37], [38]. The observed unique diffraction patterns were in agreement with standard reference data of the wurtzite phase with the obtained crystallite sizes of peaks broadening analysis matching closely with nanoscale dimensions observed by use of electron microscopy methods [39], [40], [41]. These findings were further confirmed using transmission electron microscopy which gave direct visualization of the internal crystallite arrangement and ensured that the average particle diameter was in line with the size estimates of the XRD peak broadening analysis [42], [43]. UV-Vis absorption spectra were used to determine the optical band gap energy and the results obtained were 3.2 eV and 3.1 eV in aqueous and ethanol-prepared nanoparticles respectively; which agrees with the quantum size effects of the nanoscale crystallite sizes [44]. These optical and structural features indicate that the green-synthesized ZnO nanoparticles have the desired features of being integrated into photodetector devices, where the important parameters of the photoconductivity are the controlled bandgap energies and purity [45], [46]. The photocurrent response was assessed by making devices on the glass substrate and the dark current-voltage characteristics were measured in the dark and under light conditions [47]. The current-voltage curves showed that the conductance increased significantly when it is exposed to light, and the ratio of the photoresponse was found to be an order of magnitude greater in the case of the ethanol-synthesized ZnO than in the aqueous one, revealing a high efficiency of carrier generation [48].

Results

The presence of the hexagonal wurtzite phase was validated by structural characterization using XRD with diffraction pattern showing sharp peaks on the characteristic planes and no impurity signals which in turn showed high crystallinity and purity of the material [37], [49]. Debye-Scherrer analysis gave a crystallite size of 14.7 nm which is in the nanoscale regime of dimensions required to facilitate effective charge transport in optoelectronic devices [50].UV-Vis spectroscopy revealed distinct absorption behavior, where the ZnO nanoparticles had intrinsic absorption in the ultraviolet region and a low absorption in the visible spectrum, consistent with their classification as wide bandgap semiconductors [50], [51]. The gap energies corresponding to these bands were estimated as 2.82 eV and 3.1 eV respectively with the lower values suggesting defect states in the crystal lattice that can be used to determine photoconductive performance [34], [43]. FE-SEM images showed that synthesized nanoparticles adopted a predominantly spherical morphology with some agglomeration degree, whereas the particle size distribution estimated from such micrographs corroborated the dimensions of crystallites acquired from XRD peak broadening analysis [52], [53]. The dynamic light scattering measurements also measured the hydrodynamic diameter of the nanoparticles in suspension and this showed an average size of 12 nm implying that these nanoscale particles could fill up interstitial voids in composite films and consequently increase overall conductivity [6], [45]. In confirmation of the optoelectronic efficiency, photoconductivity of the prepared nanoparticles of ZnO was tested by evaluating the currentvoltage characteristics in the dark and ultraviolet light [54], [55]. The measurements showed a significant improvement of electrical conductivity during UV exposure where photocurrent increasedby a factor proportional to the carrier density produced by the incident photon energy [56]. The enhancement of the photoconductivity is largely due to the creation of electron-holes pairs in the wide bandgap ZnO structure the separation efficiency of which is dictated by low rate of recombination linked to the nanoscale size of the crystallites [57], [58]. The identified change in absorption peaks to shorter wavelengths than those of bulk ZnO is a good indicator that quantum confinement effects exist and are necessary in the spectral response tuning photodetectors [15], [59]. These quantum confinement effects permit the accurate engineering of the bandgap which is essential towards maximizing the detection range and sensitivity of ultraviolet photodetectors at different wavelengths. Moreover, the particular surface area that can be linked to these nanoscale dimensions aids in achieving a greater adsorption of the oxygen species which is vital in regulating the baseline level of conductivity and the increase of the photoresponse gain by the means of the surface desorption process [15]

Discussion

The surface desorption mechanism can directly be responsible of the increased photoconductivity of the ethanol-synthesized nanoparticles, in which the photogenerated holes move to the surface where they cause a desorption of adsorbed oxygen ions to release trapped electrons into the conduction band, leading to the highly enhanced conductivity at UV illumination [55]. On the other hand, the oxygen readsorption process reinstates the depletion layer during the dark interval which causes an upward bend bending which decreases the carrier concentration [56]. This cyclical process of oxygen desorption and readsorption is suitably used to control the depletion layer width, which gives a great gain mechanism and increases the photocurrent signal more than the expected level due to the electron-hole pair generation process [60], [61]. Also, the ambient species including water vapor and surface carbonates interacting with the ZnO surface affect the temporal dependency and magnitude of the UV photoconductivity, implying that the preparation and storage conditions of devices is vital in predicting the stability of photoresponse [55]. The defect states, including neutral oxygen vacancies, are part of the contribution to the sub-bandgap photoconductivity, which allows electrons to transfer between deep states to shallow donors, producing narrow-band spectral properties and affecting the transient response properties of the photodetector [62]. This gain parameter depends on the kinetics of adsorption of atmospheric oxygen, since these surface interactions determine the rates of recombination and eventually determine the sensitivity and recovery times of the photodetector [55], [63]. Such a critical analysis has shown that the high surface-to-volume ratio of the nanostructures allows enhanced densities of adsorbed oxygen species, which, upon desorption under illumination, release more trapped electrons to enhance the photoconductive gain [47], [64]. The release of these trapped charge carriers is also complemented by the lowering of injection barriers at the Schottky contacts where photogenerated hole trapping within the depletion layers reduces the barrier height and enhances a larger rate of injection of electrons at the electrode [47], [62].

Moreover, this barrier reduction mechanism effectively prolongs the carrier lifetime by preventing immediate recombination, yielding a high photoconductive gain that is characteristic of surface-mediated photodetection in n-type ZnO nanostructures [65], [66]. Surface-mediated processes have been further supported by the occurrence of defect-related emissions and high densities of surface charge traps that have been established as important factors leading to the demonstration of narrowband photodetection in ZnO nanostructures [65].

Conclusions

To summarize, the green synthesis regarding ZnO nanoparticles in ethanol and water at low energies resulted in successful synthesis of nanoscale materials with a spherical morphology, and an average hydrodynamic diameter of 12 nm, which shows that it can develop cost-effective and environmentally friendly fabrication of optoelectronic devices. This method of synthesis has great benefits as compared to standard high-temperature techniques, consuming less energy and eliminate the need for toxic chemical precursors - thus creating a scalable route to the development of high-performance photodetectors. The positive surface to volume ratio and quantum confinement properties of these nanoparticles enable better photodetection through maximizing oxygen adsorption sites and efficiently tuning the bandgap for optimal spectral response. The optimization of the defect states and surface morphology that was shown in this work creates a direction in making the ZnO photodetectors that are very photosensitive and have rapid response time applications in the broad-band spectral absorption [47], [66]. Future studies ought to take into consideration incorporating these green-synthesized nanoparticles into flexible device designs with an aim of exploiting their tunable optoelectronic characteristics in the development of sustainable printed electronics devices [14], [67]. This form of integration may utilize the high levels of surface defects and high photocatalytic activity of plant-extract derived nanoparticles to create multi-functional sensor systems with the ability to detect and degrade pollutants in parallel [14], [68]. These bio-inspired nanomaterials have the potential to combine the high efficiency in charge separation with the reduction of the surface defect density and the reaction parameters, and this resulted in the improvement of the photocatalytic remediation and optoelectronic sensing applications. All in all, the effective synthesis of ZnO nanoparticles in these benign routes highlights their dual capability as high-performance photodetectors as well as environmentally conscious agents for water treatment applications [14], [15]. All these results promise the future of bio-engineered systems of making nanomaterials that can not only enhance optoelectronics, but also help the environment by being part of the environmental remediation process through low-energy fabrication systems [12], [69]. Such multifunctional capability integration facilitates the development of sustainable nanotechnology, whereby the performance of functional materials meets the global environmental requirements [25]. The defined approach, which is based on low-energy input and the incorporation of benign solvents, is a solution that may replace the conventional physicochemical synthesis approaches that typically produce toxic byproducts [12]. These green synthesis principles can eliminate the ecological footprint of nanomanufacturing, as well as enable the development of safer photodetector devices, which can be used in the mass market[70]

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