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Original Article

The Green Synthesis and Characterization of Zinc Oxide Nanoparticles from the Leaf Extracts of *Satureja hortensis*

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Abstract

Background and Aim: Zinc oxide nanoparticles are widely used in various industrial and medical fields. In order to prevent and reduce the environmental effects of commercial nanoparticles synthesized via conventional chemical methods, this study was designed to prepare green synthesized zinc oxide nanoparticles using *Satureja hortensis*.

Materials and Methods: In this study, the aqueous extracts of the Satureja plant were prepared as reducing agents. ZnO nanoparticles were synthesized using zinc nitrate and sodium hydroxide. The powder samples were analyzed using an infrared spectroscopy (UV-Vis), scanning electron microscopy (SEM), Energy-dispersive X-ray analysis (EDX), Fourier Transform Infra-red Spectroscopy (FTIR), and X-ray diffraction (XRD).

Results: The findings of this research, including the change in solvent color, the absorption spectrum at 330 nm, electron microscopy images, the determination of zinc and oxygen as the main elements in the composition, and the peaks in the X-ray diffraction spectrum, confirmed the synthesis of zinc oxide nanoparticles.

Conclusion: The green synthesis of nanoparticles using plants is easier to use. It is non-toxic, and more environmentally friendly. Moreover, it should be replaced with physical and chemical methods of synthesis of nanoparticles.

Keywords: Savory summer, *Satureja hortensis*, Green synthesis, ZnO nanoparticle

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Introduction

Metal oxide nanoparticles are among the most significant nanoscale classes in the industry (1). Among the metal nanoparticles, ZnO nanoparticles are highly useful used in medicine, pharmacy and various industries (2). After silver oxide, titanium dioxide and silicon dioxide nanoparticles, ZnO nanoparticle is the most wildly used nanoparticle for the production of nanoscale materials. Overall, the 36 nano-products that were made are quantitatively estimated to be more than 550 tons per year (3). The nanoparticles are usually synthesized using chemical methods, and their excessive use has led to an increase in nano-sewage, most of which enter into aquatic environments that is estimated to be more than 370,000 tons per year (4). The introduction of nanoparticles into water has resulted in genetic effects and increased mortality of fish and other aquatic organisms (5). Chemical methods of making nanoparticles are also costly in the environment (6). Hence, for various reasons, especially for environmental benefits, the biological synthesis methods or green synthesis of nanoparticles have been considered by the researchers (7). In these novel methods (i.e. green synthesis), plants, microorganisms or fungi are used as the base to produce nanoparticles, and they have been identified as an efficient way to exploit the biological substances found in nature (8, 9). Plants could be widely used due to their abundance, easy availability, and environmental compatibility without inducing adverse environmental impacts (10). The procedures that are used to produce nanoparticles from different parts of plants are technically easier than chemical methods. Hence, biological materials play a pivotal role in the synthesis of nanoparticles, and they are referred to as novel solutions that could be replaced with chemical methods (11). Plants such as aloe vera (12), *Glycosmis pentaphylla* leaf (13), *Limonia acidi*ssima (14) are used to synthesize ZnO nanoparticles. Even though the significance of plants for the synthesis of nanoparticles has been highlighted, unfortunately, only a limited number of studies have been carried out about this subject in Iran. Since Iran is characterized by a remarkable diversity of plants, more studies are required to be conducted in this regard. One of the most important herbs in Iran, which is native to different regions of the country, is *Satureja hortensis*. *Satureja hortensis* is a plant which is widely used in various food products. This herb is a herbaceous one-year-old plant that belongs to the family of mint. This plant is resistant to various weather conditions. Its height ranges from 40-60 cm, and its dark green leaves have purple flowers. Sucral medicine has many properties, such as antioxidant, antifungal, antibacterial and antiseptic properties. Moreover, it is an LDL-cholesterol-lowering agent, and is rich in oily and protein substances (15). Regarding this issue, this study sought to synthesize the green nanoparticles of zinc oxide from satureja plants.

Materials and Methods

Materials

All the chemicals used in this study, such as Zinc nitrate, sodium hydroxide, etc., were purchased from Sina Teb (a representative of Merck Company).

Collecting the Satureja Plant

The leaves of *Satureja hortensis* were purchased from the market of medicinal herbs in Khorramabad, Lorestan. The identification was carried out by the herbariums unit of Lorestan University. The leaves were washed with tap water and distilled water to remove the dust particles and other possible contaminants, and then were dried at room temperature (absence of light). They were finally crashed very well.

Preparation of the Leaf Extract

Extraction of the plant was conducted via mixing 10g of dried leaves of the satureja in 100 ml of double distillated water in a 250 ml beaker. The container was placed on an electric stirrer at 150 ° and 300 rpm for 1 h. The solution was then filtered using Whatman No 40 filter paper after cooling to room temperature.

Green Synthesis of ZnO Nanoparticles

The extract of *Satureja hortensis* leaves was used for the reduction of zinc ions ($Zn2+$) to zinc oxide nanoparticles. 100 ml of the Satureja extract was stirred at 400 rpm and 150 ° C. After ten min, 7g of zinc nitrate and 10 ml of sodium hydroxide (2.5g of hydroxide sodium in 50 ml of distilled water) were added. Consequently, the color changed after a few minutes. After cooling down, the solution was centrifuged at 5000 rpm for 5 min. The precipitate was then dried at 85°C for 90 min. To complete the process, the precipitated solids were crushed in a porcelain mortar and passed through a sieve with a pore size of %15 mm. ZnO nanoparticles synthesis steps have been indicated in Figure 1.

Characterization:

UV-Vis Spectrometry

To carry out the spectroscopy analysis, 200 µg of ZnO nanoparticle powder was added to 1 ml distilled water, and then the resulting substance was scanned in the range of 220-500 nm by a spectrophotometer (model T80-uv-vis, England) and the DR5000 (model HACH, Germany).

Scanning Electron Microscope - (SEM)

Scanning electron microscope (FE-SEM, TESCAN Company, Czech Republic) analysis was used to analyze the morphological features of synthesized zinc oxide nanoparticles from *satureja hortensis*.

Energy-Dispersive X-ray Analysis - (EDX)

X-ray energy equipment (mira3-tescan device, Czech Republic's) analysis was performed to determine the main composition of the elements present in the synthesized nanoparticles. FTIR Analysis was used to identify the potential presence of functional groups in the biosynthesis of zinc oxide nanoparticle powder from Satureja plant. The analysis of the specimen was performed by thermo company Nicolet 4700 model, USA.

X-ray Diffraction - (XRD)

XRD analysis (STOE Company, Germany) was conducted to observe the phase change and crystallite size of the samples. The analysis of the specimen was performed using the staid model of the X-ray diffraction.

Results and Discussion

The present study resulted in the production of brown powder of ZnO nanoparticles. The initial confirmation of the synthesis was made by changing the color of the colloidal solution from red to green mustard. After observing the color change, the spectrophotometric method was used to confirm the synthesis of ZnO nanoparticles. The absorption peak at 330 nm indicated the presence of zinc oxide nanoparticles and a confirmation of its synthesis. Hence, the change in solvent color and absorbance at 330 nm confirmed the synthesis of zinc oxide nanoparticles from *S.hortensis* plant (Figure 2). Moreover, different images (at the range of magnifications of 500 nm to 20 µm) of the synthesized nanoparticles clearly indicated the presence of nanoparticles in different shapes of spherical and crystalline forms in a dense and clinging manner and confirmed the nanoparticle's synthesis (Figure 3). As a semi-quantitative method for determining the elements, EDX analysis confirmed the presence of zinc and oxygen signals in zinc oxide nanoparticles produced from Satureja plant (Table 1). In this table, oxygen content with 51.43% and zinc with 35.5% were identified as the main elements in the sample. In addition to the main elements in the zinc oxide nanoparticle, the carbon element with a percentage weight of 13% was also revealed in the composition.

Several peaks in the EDX spectrum indicated that these peaks have identified zinc, oxygen, and carbon elements, with the highest concentration of zinc, followed by oxygen and then carbon (Fig. 4). The

Figure 1. The Steps of ZnO Nanoparticles Synthesis Using *s.hortensis*.

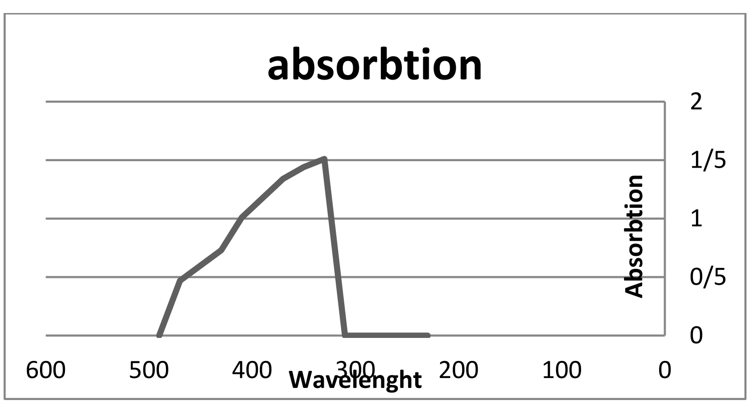

Figure 2. UV Spectra of ZnO NP Synthesized by *S.hortensis*.

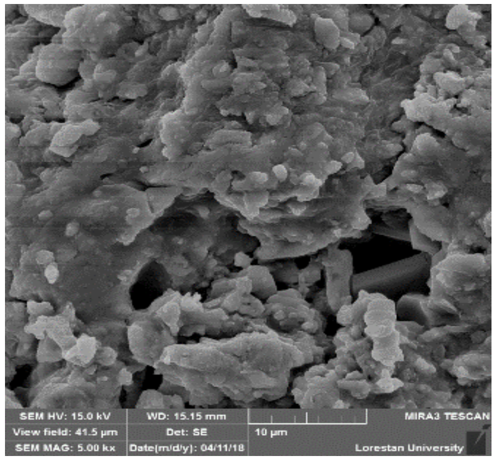

Figure 3. The SEM Image of Zinc Oxide Nanoparticles Synthesized by *S.hortensis*.

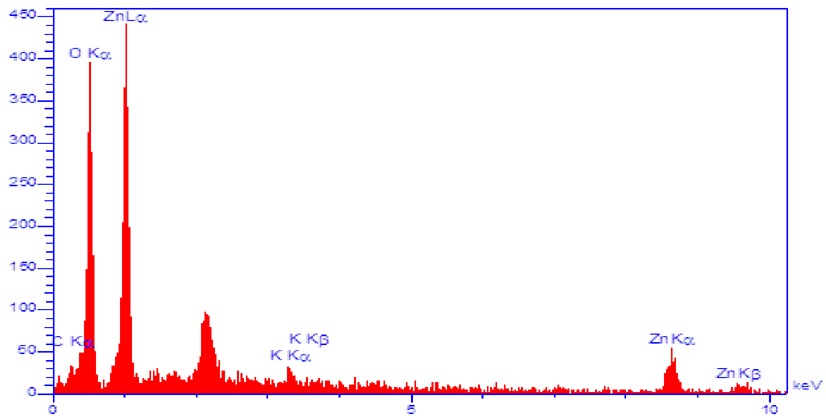

Figure 4. The EDX Spectrum of ZnO Nanoparticles by *Satureja hortensis*.

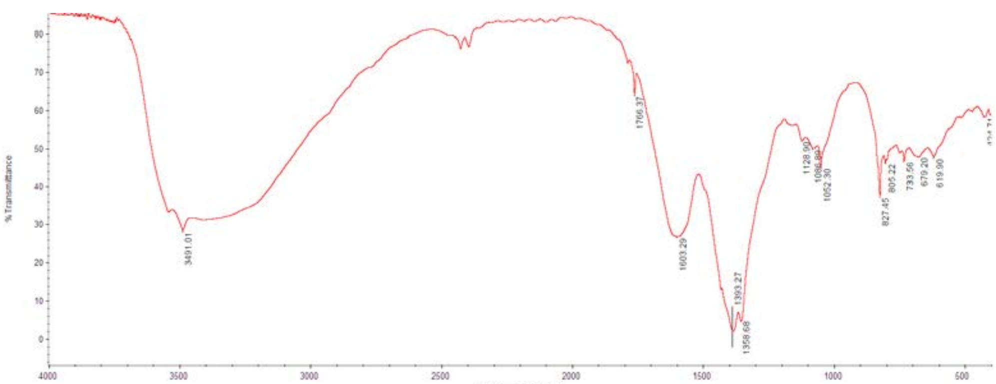

Figure 5. The FT-IR Spectra of Synthesized ZnO Nanoparticles Using *S.hortensis*.

main peak of the Zno nanoparticle is in the range of 1 keV. Other weak peaks are observed in higher ranges such as 3 and 8.5 keV. Figure 5 shows the FTIR spectra of the sample powdered in the range of 500-4000 cm^{-1}. According to Fig 5, the broad peaks could be observed at 3491.01 cm^{-1}, 1766.37 cm^{-1}, 1393.27 cm^{-1}, 1128 cm^{-1} and 827.45 cm^{-1}. The X-ray diffraction spectra of the Satureja plant has also been indicated in Fig. 6. Based on this pattern, it was determined that the most zinc-synthesized phase belongs to the zinc element that represents the formation of zinc oxide nanoparticles from the sorghum extract. Multiple couriers have been shown in the diagram that indicates the value of two in the range of 20-80 degrees, which is related to the hexagonal structure of zinc oxide. A number of reflections were observed with 29.52 degrees, 30.24 degrees, 34.32 degrees, 36.02 degrees, 48.93 degrees and 58.51

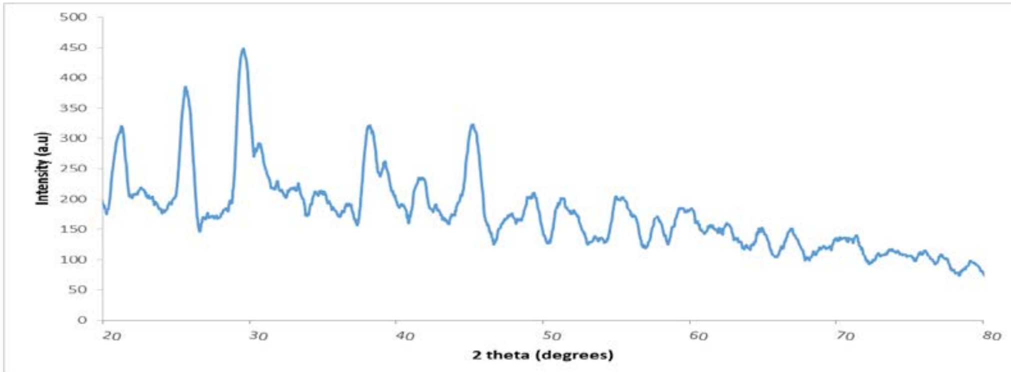

Figure 6. The XRD Pattern of Zno Nanoparticles by *Satureja hortensis*.

Table 1: The Major Elements in the Composition According to EDX Analysis.

Element	% Weight	% Atomic
Zn	35.5	11.25
O	51.43	66.2
C	13.07	22.4

degrees, respectively, relating to crystalline plates (100), (002), (101), (102) and (110). Particle diameter via considering the Debye-Scherrer formula was calculated from 24.78 to 64.19 nanometers with an average of 35.88 nm.

In the present study, the biological reduction method was used to prepare zinc oxide nanoparticles using the aqueous extract of Satureja plant. The aqueous extract of the Satureja plant contains compounds that can reduce the zinc nitrate salt to the zinc nanoparticles (15). These compounds include thymol (29.1%), carvacrol (26.6%), gamma terpinene (24.7%) and parasmene (7.5%) (16). The compounds in the Satureja plant could play a pivotal role in the synthesis of zinc oxide nanoparticles because of their active functional groups (16). The change in the color of the solution from red to green mustard is the first sign of the nanoparticle synthesis that occurs due to the interaction of the plant extract and the zinc solution (17). Reduction of zinc ions into zinc oxide nanoparticles throughout contact to plant extracts was observed as a result of the color change. Change in the color of the solution also indicated the existence of zinc oxide nanoparticles, and confirmed the formation of the zinc oxide nanoparticle (18). The color modification is due to the Surface Plasmon Resonance (SPR) phenomenon. The metal nanoparticles have free electrons that give the SPR absorption band due to the joint vibration of electrons of metal nanoparticles in resonance with light wave (19). In fact, the color change is the first indication of the synthesis of ZnO nanoparticles (20). This study is unique, probably because of laboratory conditions at the time of nanoparticle generation and plant type (21). Jayandran *et al.*, reported the duration to be roughly 1 h (19), whereas Raj *et al.*, reported this period to be about 2h (22). Similar to this study, the use of zinc nitrate salt for the synthesis of nanoparticles has also been addressed in many other studies (23, 24). This research eventually led to the preparation of a brown powder of zinc oxide nanoparticles from the sorghum, which is compatible with the observations of Devasenan. In his research, he reported that the color of the zinc oxide nano powder is greenish-brown (25). Joel's observations revealed that nanoparticles of zinc oxide produced by chemical methods are white, but nanoparticles produced by biological methods are brown in color (17). In several studies, the variation in the color of the solution has been mentioned as the initial indication of the nanosized metal oxide synthesis (12). The results of available studies indicate that zinc oxide nanoparticles often have absorption peaks between 305-345 nm (26-30). According to the results of this study, the max absorption peak was at 330 nm, which is in complete agreement with the published reports. From the morphological analysis point, the shape of the spherical of crystalline nanoparticles were the dominant form of synthesized ZnO nanoparticles in the present research. Similar studies have also reported the shape of zinc oxide nanoparticles in various forms, particularly the spherical shape (28, 29, 31). The EDX analysis was carried out in addition to the determination of zinc and oxygen as the main components found in the carbon element sample with a weight percentage of about 13%. The presence of carbon in the spectrum indicates the activity of plant phytochemical groups to limit and reduce the synthesis of zinc oxide nanoparticles (32). In the ultra-peak spectrum, the nanoparticles of zinc oxide are determined at energy 1kev, which is consistent with the results of Parthiban and Saravanakkumar (33). Chaudhuri also described the peaks at energy levels up to 1-2kev (34). The peak at 3491.01 cm^{-1} indicated -OH stretching vibrations of phenolic compounds and hydrogen-bonded alcohols functional groups in the sample, while the peak observed at 1766.37 cm^{-1} could be assigned to C=O stretching due to carboxylic acid and ester, aldehyde and ketones (27). The peak at 1393.27 $^{cm-1}$ is the characteristic absorption of C-H bending (35). the peak at 1128.9 cm^{-1} could be attributed to the characteristic absorption of C-O stretching, and finally weak band at 827.45 cm−1 is the result of C=C bending (35).

The XRD spectrum indicated the hexagonal structure of zinc oxide nanoparticles of an average particle diameter of 35.88. In various studies, the hexagonal structure of the zinc oxide nanoparticle has been confirmed, and the diameter of particles has been calculated with a small difference at the same range, depending on the testing conditions, plant type and region and the genetic properties (36, 37). The presence of additional couriers in the graph of the elements in the sample has a crystalline structure (6).

Conclusion

In this study, Satureja plant was used to synthesize ZnO nanoparticles, because it contains effective chemical compounds for antioxidant and regenerative activities. Nanoparticle specification was determined using the available tools. The results indicated that this method is a low-cost, simple, low-risk and environmentally friendly method that could produce nanoparticles of appropriate size, and replace chemical methods.

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Conflict of Interest

The authors declare that they have no conflict of interest.

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