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Synthesis and Characterization of Zinc Oxide Nanoparticles (Zno-NPs) From Leaves of Some Plants

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Abstract

The current study focuses on producing zinc oxide nanoparticles (ZnO-NPs) from aqueous leave extracts because it is more environmentally friendly and cost-effective than traditional physical and chemical methods. Synthesis of aqueous extracts of *Moringa Oleifera*, *Ocimum tenuiflorum (Tulsi)*, and Neem *(Azadirachta indicia)* leaves. Zinc acetate dihydrate (Zn $(CH_3COO)_2.2H_2O$) was used as a precursor to biosynthesize ZnO-NPs at different temperatures (50, 75, and 100 ^oC) for 1 hour of the synthesis of Zinc oxide nanoparticles (ZnO-NPs) were characterized using X-ray diffraction (XRD), scanning electron microscopy (SEM), Fourier transforms infrared (UV). The presence of biomolecules such as polyphenols, carboxylic acid, polysaccharides, and amino acids was revealed by FTIR analysis of green synthesized nanoparticles. The results of the SEM study of green synthesized ZnO nanoparticles revealed that the sample exhibits a compact arrangement of homogeneous nanoparticles with a spherical shape and successfully unfolded it was crystalline structure nature.

Keywords: Green synthesis; ZnO nanoparticles; characterization; X-ray; SEM; FTIR; UV.

Introduction

Nanotechnology is one of the strategies that appears to have the most potential, as nanomaterials present a larger specific surface area and particular physical, chemical, and biological properties (developed due to small particle size) that are suitable for applications in the environmental field **(Deak *et al.*, 2019)**. When compared to traditional methods, which involve toxic substances and convoluted procedures, green nanoparticle synthesis is an essential component of nanotechnology that provides exceptional advantages for the environment as well as significant financial savings **(Sierra *et al.*, 2018)**.

Zinc oxide, also known as ZnO, is a crystalline powder that can range in color from white to yellowish-white and is almost completely soluble in water. Wurtzite crystals were the most typical of their crystalline forms (hexagonal). The Zinc Oxide Nanoparticles were a type of inorganic compound that had the formula ZnO for their molecular structure. ZnO powder finds widespread application as an additive in a wide variety of materials and products, including ceramics, glass, cement, and rubber, amongst others **(Sabir *et al.*, 2014)**. Nanoparticles made of zinc oxide are an important type of metal oxide that finds widespread use in the field of materials science due to the unique physical, chemical, and biological properties that they possess. These properties include biocompatibility, environmental friendliness, low cost, and non-toxicity **(Alwan *et al.*, 2015)**.

Moringa oleifera leaves, *Ocimum tenuiflorum* leaves, and Neem (*Azadirachta indica*) leaf extract are being used in this study to develop improved and more reliable methods for the bio-fabrication of zinc oxide (ZnO) nanoparticles using green methods. These methods involve the utilization of *Moringa oleifera*, *Ocimum tenuiflorum*, and *Neem* leaves Several different characterization techniques, such as X-ray diffraction spectroscopy (XRD), high resolution scanning electron microscopy (SEM), and Fourier transform infrared analysis, were utilized in order to investigate the electrochemical activity of ZnO nanoparticles (FTIR). Also, the characterization of the ZnO-NPs and their use in the treatment of contaminated water, as well as the determination of the optimal conditions for their production using leaf extract. **Mlitan *et al.*, (2014)** prepared the leaves of *Ocimum tenuiflorum* that were collected from three different locations in the Misurata region of Libya (Zaroge, Tamina, and Daphnia) in order to conduct an analysis of the proximate and minor minerals constituents in the samples. According to the results of the proximate analysis of the samples (Zaroge, Tamina, and Daphnia), the percentages of crude protein content, moisture, lipids, ash, and carbohydrate of the leaves on a dry weight basis were as follows: (9.10, 10.60, 41.30, 28.92, 10.08%), (9.80, 10.40, 12.85, 14.50, 52.45%), and (9.22, 10.60, 35.55).

The ZnO nanoparticles were assembled by **Ghorbani *et al.*, (2015)** by combining the Zn $(NO_3)2.6H_2O$ and the KOH in an aqueous solution. The precipitated compound was calcined, and its properties were analyzed using transmission electron microscopy (TEM), UV-Vis spectroscopy, and dynamic light scattering (DLS). Around 372 nm was the wavelength at which the characteristic surface plasmon resonance peak was observed for the ZnO nanoparticles. Based on the results of the dynamic light scattering technique (DLS), it was determined that the particles had a size ranging from 20 to 40 nanometers. On the other hand, **Noorjahan *et al.*, (2015)** used an aqueous extract of Neem (*Azadirachta indica*) leaf to synthesize zinc oxide nanoparticles. When it comes to the production of nanoparticles, the extract of the leaf in aqueous form serves not only as a solvent but also in multiple other capacities, including those of promoter, stabilizer, and template. The FTIR analysis of green nanoparticles revealed the presence of biomolecules such as polyphenols, carboxylic acid, polysaccharides, amino acids, and proteins. Additionally, the results of the SEM analysis of the green synthesized ZnO nanoparticles showed the formation of spindle-shaped nanoparticles as well as nano-flakes of zinc oxide.

Okiki *et al.*, (2015) studied assessing the nutritional and phytochemical constituents of the leaves of *Moringa oleifera*. The proximate analysis of dried leaves of *Moringa oleifera* showed that it was highly rich in protein (28.00±0.33%) and metabolizable energy (2625.25±79.30 Kcal/Kg), with appreciable levels of fat (3.88%), ash (9.88%), crude fiber (12.57%) and carbohydrate (37.87%).

Pinto and Nazareth (2016) used a variety of methods, including chemical, physical, and biological, to prepare the ZnO particles. The green synthesis was accomplished through the utilization of an aqueous solution containing *Ocimum tenuiflorum (Tulsi), Phyllanthu Emblicaca* (*Gooseberry*), Neem (*Azadirachta indica*) extract, and zinc nitrate. The nanoparticles were characterized by using FTIR, XRD, and SEM. The size of the particles that were synthesized ranged from 10 to 30 nanometers (nm). The XRD analysis of each of the prepared samples revealed the presence of zinc oxide in nanoscale form, as indicated by a distinctive peak in the analysis. The structure of the zinc oxide nanoparticles appeared to be elongated and floral when viewed through a scanning electron microscope (SEM). ZnO nanoparticles were found to be present, as demonstrated by the IR spectra. Antimicrobial activity was also observed in the samples that were synthesized.

The zinc oxide nanoparticles were synthesized by **Thirunavukkarasu *et al.*, (2016)** utilizing an aqueous extract of *Moringa oleifera* obtained from the leaf, blossom, and bark. The green synthesis sol-gel method was used for the preparation. X-ray diffraction (XRD), scanning electron microscopy (SEM), ultraviolet-visible (UV-Vis), and Fourier transform infrared spectroscopy (FTIR). Was all utilized in order to investigate Zinc Oxide Nanoparticles. The results of an analysis using UV-visible spectroscopy showed that the band gap energy was highest in the near-visible region at a wavelength of 286.5 nm. This value was 4.3 eV. The use of *Moringa oleifera* bark to produce ZnO nanoparticles resulted in the strong appearance of peaks in XRD, which demonstrated the crystallinity of the particles. The synthesis of green nanoparticles was investigated by **Khaing *et al.*, (2018)**, who used an aqueous solution of *Neem (Azadirachta indica)* leaf extracts along with zinc nitrate solution in their research. In addition, we analyzed them by employing techniques such as spectroscopy (UV-Visible), FTIR, EDXRF, XRD, and scanning electron microscopy (SEM). They discovered that the FT-IR spectrum of the aqueous neem extract showed the presence of O-H stretch of 3377.41cm^{-1} from the extract of Neem leaves, and they found that the typical absorption peak of the Zn-O bond was identified at 832.30 cm^{-1}. The high crystallinity was clearly indicated by the strong peaks in the XRD pattern, and the size of the ZnO nanoparticles at 100 degrees Celsius was found to be 37.79 nm and 24.60 nm respectively. The green ZnO that was synthesized was found to have formed nanoflakes and nanoparticles in the shape of spindles, according to experiments using a scanning electron microscope (SEM).

Researchers from **Pal *et al.*, (2018)** investigated the process of precipitating ZnO nanoparticles from a natural precursor known as Moringa Oleifera (Drumstick) leaves. Using UV-VIS spectroscopy, we were able to establish both the formation of ZnO nanoparticles as well as their characterization. They performed an X-ray diffraction examination and found that the produced nanoparticles have a hexagonal wurtzite structure with an average grain size of 52 nm. In addition, ZnO nanoparticles that have been artificially produced have been used as photocatalytic agents in order to break down the organic dye. Titanium appears yellow in visible light and can be made to do so by exposing it for one hour to visible light. The yellow titanium dye was nearly completely destroyed by ZnO nanoparticles. The chemical contents of the aqueous extract of Neem (*Azadirachta Indica*) leaves were investigated by **Garba and Mungadi (2019).** The findings indicated that there was a presence of tannin in the amount of 1.4%, oxalate in the amount of 1.41mg/100g, phytate in the amount of 6.12mg/100g, saponin in the amount of 22.55%, cyanogenic glycoside in the amount of 2.05mg/100g, an alkaloid in the amount of 12.22%, trypsin inhibitor in the amount of 6.25%, flavonoid in the amount of 32.06%.

Neem (*Azadirachta indica*) and Tulasi (*Ocimum tenuiflorum*) leave extracts to serve as the bulk molecules that **Ajayan and Hebsur (2020)** use to produce zinc oxide nanoparticles. With the help of a particle size analyzer and scanning electron microscopy, the newly created particles were examined to determine their dimensions and form. When measured with a particle size analyzer, the zinc oxide nanoparticles that were generated using neem and Tulasi had an average size (diameter) of 101.6 and 122.4 nm, respectively. ZnO nanoparticles produced from Neem recorded a flakes-like or flower-like appearance when observed by scanning electron microscopy (SEM), and their sizes ranged from 100 to 300 nm. The ZnO-NPs that were produced from the Tulasi leaves extract recorded ovoid forms and sizes that ranged from 70-400 nm. The synthesis of zinc oxide (ZnO) nanoparticles was performed by **Sharma *et al.*, (2020)** using the green synthesis method. They used various parts of the Neem tree, such as the leaves, bark, twigs, and fruits, for the synthesis of ZnO nanoparticles, primarily by two methods: one that involved soaking and the other involved boiling. After the nanoparticles that were generated were characterized, the XRD technique was utilized, and the results revealed that the size of the particles ranges from 16 nm to 31 nm. In addition, the produced nanoparticles have a crystalline character, which allows them to be controlled and exact, and they have well-defined sizes and forms of a variety of different configurations.

ZnO nanoparticles were synthesized and described by **Yasotha *et al.*, (2020)** using an environmentally friendly synthesis approach that involved the extract of *Ocimum Tnuiflorum* leaves. They discovered that the acquired results show that the crystalline size, shape, and composition meet the common values and will be useful for applications involving antibacterial treatment. Before characterization, the ZnO-NPs created by **Adam *et al.*, (2021)** were subjected to heat treatment at temperatures of 300, 350, 400, 450, and 500 degrees Celsius. These temperatures were used during the preparation and synthesis of the ZnO-NPs. Methods include Fourier transform infrared spectroscopy, X-ray diffraction, and (UV-Vis). They discovered that the FTIR analysis reveals an absorption band at (422-497 cm^{-1}), which was associated with the stretching of Zn-O, and the intensity of the O-H bond (3417 cm^{-1}) decreases with increasing calcination temperature. Both of these findings are related to the stretching of ZnO. As the temperature rises, the XRD spectrum reveals a discernible improvement in the electron characteristics and bond formation of the material.

Materials and Methods

Moringa Oleifera, *Ocimum tenuiflorum*, and Neem (*Azadirachta indicia*) leaves were collected from the Faculty of Agriculture, Benha University, Qalyubia Governorate. Leaves were washed thoroughly with water and were allowed to dry in the air at room temperature.

Chemicals: - Zinc acetate dihydrate ($Zn(CH_3COO)_2.2H_2O$)
ammonium carbonate $(NH_3)_2CO_3$, methanol were obtained from El-Gomhouria Com and El-Nasr Com.

Preparation of green leaves extracts: Green leaves of *Moringa Oleifera, Ocimum Tenuiflorum,* and Neem (*Azadirachta Indicia*) extracts were prepared by the method described by **pal *et al.*, (2018).**

Green synthesis of ZnO nanoparticles using green leaves: Ten mL of extract from green leaves was combined with 50 mL of a solution containing twenty percent NaOH 20%. After that, 5 mL solutions of that mixture were added to 50 mL of distilled water in a beaker with a capacity of 250 mL, and the mixture was stirred for an hour. After that, solutions of zinc acetate (2.1 grams dissolved in 100 mL of water) and ammonium carbonate (0.96 grams dissolved in 100 mL) were simultaneously and drop wise put into the beaker while being stirred. After the completion of the reaction, the suspension was allowed to remain stirred at room temperature for a period of one hour. In the end, the precipitate was filtered and washed repeatedly with an ammonia solution before being repeatedly treated with ethanol. The precipitates were then dried in a vacuum for a period of 12 hours before being calcinated in a hot air oven at a temperature of 350 degrees Celsius for a period of 5 hours. The zinc oxide nanoparticles were collected and kept for future use in a vacuum storage facility **(pal *et al.*, 2018)**.

Chemical composition of green plants under investigation:

Moisture, ash, crude protein, and crude fat content were determined according to the method described by **A.O.A.C (2019).** The total carbohydrates were determined by difference.

Characterization of zinc oxide nanoparticles: -

X-ray diffraction spectroscopy (XRD): provides the calculation for determining whether or not ZnO nanoparticles are, on average, crystalline. The intensity of the rays that have been diffracted is displayed as a function of the diffraction angles on the diffractogram. The spectra provide an illustration of the intricate characteristics of the crystal planes. At the Egyptian atomic energy authority in Nasr City, the testing of the samples was performed in an XRD with the model number XRD-6000. The Debye-Scherrer equation was utilized in order to get an accurate reading of the nanoparticles' sizes **(Jayachandran *et al.*, 2021).**

$$D = \frac{K\lambda}{\beta\cos\theta}\ A$$

Where: **D =** the average crystalline size in A°.
K = the Scherrer constant (0.9).
λ = X-ray wavelength (1.5406 A°) s.
β = peak width of half maximum.
θ = the Bragg diffraction angle.

Scanning electron microscopy (SEM) The morphology of the produced nanoparticles, including their size and shape, were analyzed with scanning electron microscopy (SEM) in accordance with the approach developed by **Ajayan and Hebsur (2020).** In the laboratory of the Mansoura Faculty of Agricultural University, the samples were analyzed using a scanning electron microscope with the model number JEOL JSM 6510 lv.

Fourier transform infrared spectroscopy (FTIR) was performed to study the surface functional group present in ZnO-NPs according to the method of **(Shamhari *et al.*, 2018).** The analyzed samples were measured within a range of 450-4000 cm^{-1}. The samples were carried out in the Fourier transform infrared spectroscopy in the Benha Faculty of Science model Nicolet™ iS10.

UV Visible Spectra Analysis was done for preliminary confirmation of green synthesized ZnO nanoparticles. The samples were recorded as a photograph of the UV-Vis spectra model V-670 of ZnO in Benha Faculty of Science, according to the method of **(Ajayan and Hebsur 2020).**

Estimation of optical band gap: For the purpose of determining the optical characteristics of amorphous semiconductors, several models are utilized. The Tauc model is the one that is used the most frequently. This model enables us to obtain the band gap energy Eg from E(ε) 1/2 as a function of the incident energy E. To calculate the Tauc optical gap associated with the thin films, an extrapolation of the linear trend seen in the spectral dependence of (hv) 1/2 is performed across a specific range of photon energies h. This allows for the gap to be calculated. At the point where this linear extrapolation meets the abscissa axis is where the Tauc optical gap will be found. In many amorphous semiconductors, the absorption coefficient (α) near the band edge displays an exponential dependence upon the photon energy. This dependence typically obeys the empirical relation developed by **Bensaada Laidani *et al.*, (2008)**.

$$(\alpha h\nu) = B(h\nu - Eg)^n$$

Where: (α) is the absorption coefficient
$(h\nu)$ is the photon energy in electron volts unit
(B) is a constant.
$(\alpha = 4\pi k/\lambda)$; k is the absorption index.
(λ) is the wavelength in nm.
(Eg) is the optical energy gape
(n) is an index that can have different values.

Results and Discussion

The present work reveals that the synthesis ZnO-NPs via a simple and cost-effective precipitation approach employing green leaves extracts such as *Moringa Oleifera, Ocimum Tenuiflorum,* and *Neem (Azadirachta Indicia),* XRD, FTIR, SEM, and UV studies confirmed the production of ZnO nanoparticles. The emergence of a white precipitate plainly demonstrates the synthesis of ZnO-NPs in the reaction mixture.

Chemical composition of green leaves under investigation: The data on the proximate of the biomass samples are tabulated and presented in **Table (1)**. The proximate analysis obtained resulted in the proximate analysis like moisture content being higher in *Ocimum Tenuiflorum* leaves (10.60±0.01g/100g). While the ash was found higher in *Ocimum Tenuiflorum* (14.50±0.01g/100g). but, *Moringa Oleifera* has a content of higher crude protein (13.5±0.01g/100g). On the other hand, the higher crude fat and carbohydrate contents for Neem *(Azadirachta Indicia)* were found to be (7.96±0.01g/100g), (62.36±0.18g/100g). These results are different from those reported by **Okiki *et al.*, (2015), Mlitan *et al.*, (2014)** and **Garba and Mungadi (2019).**

Table 1. Chemical composition of green leaves (g/100g) on the dry weight basis:

Green leaves	Components (%)				
	Moisture	Ash	Crude protein	Crude Fat	*Total carbohydrate
Moringa Oleifera	$9.37±0.01^{c}$	$8.60±0.01^{c}$	$13.50±0.01^{a}$	$7.30±0.01^{b}$	$61.23±0.01^{b}$
Ocimum Tenuiflorum	$10.60±0.00^{a}$	$14.50±0.01^{a}$	$7.42±0.01^{c}$	$6.52±0.01^{c}$	$60.96±0.02^{c}$
Neem (Azadirachta Indicia)	$9.50±0.01^{b}$	$11.62±0.01^{b}$	$8.56±0.01^{b}$	$7.96±0.01^{a}$	$62.36±0.18^{a}$
LSD at 0.05	0.02	0.02	0.03	0.04	0.37

a, b & c: No significant difference (P>0.05) between any two means, within the same column and with the same superscript letter.
*The difference gave total carbohydrate

Yield percent of prepared ZnO Nanoparticles from green leaves at different temperatures: The current study demonstrates that the preparation of an aqueous extract from the green leaves of *Moringa Oleifera, Ocimum Tenuiflorum,* and Neem (*Azadirachta Indicia*) can be used to produce zinc oxide nanoparticles (ZnO-NPs) at three different temperature ranges (50, 75, and 100 degrees Celsius) for one hour. The outcomes that were achieved are detailed in **Table (2).** According to the findings from their research, the temperature ranges had an effect on the yield percent of ZnO-NPs that were extracted from the leaves of *Moringa Oleifera* and *Ocimum tenuiflorum*. According to these findings, the amount of ZnO-NPs that could be isolated from Moringa Oleifera amounted to 0.727% of the total yield. The temperature was determined to be (50 degrees Celsius), and the concentration of green leaves was found to be 10 grams per 100 mL of the leaves. The yield percent of ZnO-NPs extracted from *Ocimum Tenuiflorum* was 0.775% when the conditions were optimal. However, at a temperature of (100^{o}C) and a concentration of 10g/100 mL, it was discovered that the yield percent of ZnO-NPs recovered from Neem (*Azadirachta indicia*) was 1.05%. The similarities between these results and those reported by **(Khaing *et al.*, 2018)**.

Table 2. Yield percent of prepared ZnO Nanoparticles from leaves of Moringa Oleifera, Ocimum tenuiflorum, and Azadirachta Indicia at different temperatures**.**

Plant leaves	Concentration (g/100mL)	Yield percent (%) at different temperatures		
		50^{o}C	75^{o}C	100^{o}C
Moringa oleifera	**5**	**0.59**	**0.68**	**0.63**
	10	**0.72**	**0.71**	**0.68**
Ocimum tenuiflorum	**5**	**0.71**	**0.76**	**0.65**
	10	**0.78**	**0.65**	**0.54**
Neem (Azadirachta indicia)	**5**	**0.65**	**0.96**	**0.97**
	10	**0.77**	**0.91**	**1.05**

Morphology and size distribution:

In order to evaluate the crystal structure of the nanoparticles, the powder X-ray diffraction patterns (XRD) of ZnO-NPs that were biosynthesized utilizing *Moringa Oleifera, Ocimum Tenuiflorum,* and Neem (*Azadirachta indicia*) were analyzed. The results that were obtained from the analysis of the various leaves are presented in **Figure (1).** The findings from the Moringa Oleifera leaves extract are presented in **Table (3).** The characteristic peaks of ZnO-NPs can be seen in these patterns the angles of 31.75 degrees, 32.99 degrees, and 34.51 degrees. Using the Scherrer formula and the data that was received from the X-ray diffraction patterns, it was possible to calculate the size of the crystal. The *Moringa Oleifera* ZnO-NPs were found to have an average size of 8.56 nm as a result of this experiment. The samples were also estimated with the help of origin software from the contribution to the intensity of the amorphous and crystallized fields by utilizing a single line fitting. The findings presented here are consistent with those reported by **(Adam *et al.*, 2021).**

Table 3. The Structure and geometric parameters of ZnO-NPs from Moringa Oleifera leaves extract

Peak Index	Peak Type	Area Intg (a.u.)	FWHM (degree)	Max Height (a.u.)	2th (degree)	Area IntgP (a.u.)	D (nm)	D Average (nm)
1	Gaussian	144.2567	0.93573	144.8283	31.75727	14.94835	8.49003	
2	Gaussian	59.1556	0.80776	68.73316	32.99436	6.12990	9.83508	**8.56905**
3	Gaussian	142.7893	1.07618	124.6464	34.51107	14.79629	7.382035	

The results that were achieved from the X-ray diffraction of the nanoparticles that were generated from the extracts of *Ocimum Tenuiflorum* leaves are listed in **Table (4).** The ZnO-NPs *Ocimum Tenuiflorum* leaves came out to have an average size of 12.10 nm as a result of the experiment. The results of the investigation were consistent with those that were reported by **(Yedurkar *et al.,* 2016) and (Sierra *et al.,* 2018). (Sharma *et al.,* 2020)** found that the particle size of the Ociumum Tenuiflorum zinc oxide nanoparticle was determined to be 28.13 nm. The results presented here are in contrast to those that were found and reported by **(Sharma *et al.,* 2020).**

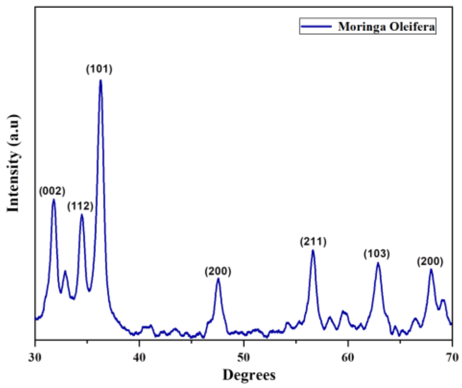
Table 4. The Structure and geometric parameters of ZnO-NPs from Ocimum Tenuiflorum leaves extract

Peak Index	Peak Type	Area Intg (a.u.)	FWHM (degree)	Max Height (a.u.)	2th (degree)	Area IntgP (a.u.)	D (nm)	D Average (nm)
1	Gaussian	193.8282	0.658384	276.5701	31.79155	29.14793	12.06649	
2	Gaussian	153.835	0.666174	216.9377	34.45515	23.13375	11.92541	**12.10629**
3	Gaussian	153.835	0.644474	462.548	36.26756	47.71833	12.32697	

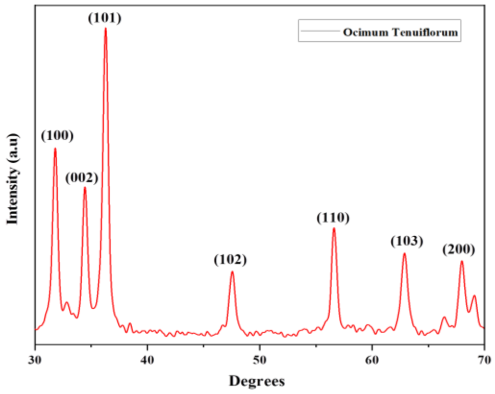
On the contrary hand, X-ray diffraction was performed on the nanoparticles that were extracted from Neem *Azadirachta indica* leaves, and the results are given in **Table (5)**. The ZnO-Nps Neem (*Azadirachta indicia*) leaves came out to have an average size of 23.68 nm as a result of the experiment. The results of the inquiry were consistent with those stated by **(Khaing *et al.,* 2018)**. These findings are quite similar to those that were reported by **(Noorjahan *et al.,* 2015)** they discovered that the particle size of Azadirachta Indica zinc oxide nanoparticles was determined to be 24.7 nm. These findings are very promising.

Table 5. The Structure and Geometric Parameters of ZnO NPs from Neem (Azadirachta indicia) leaves extract

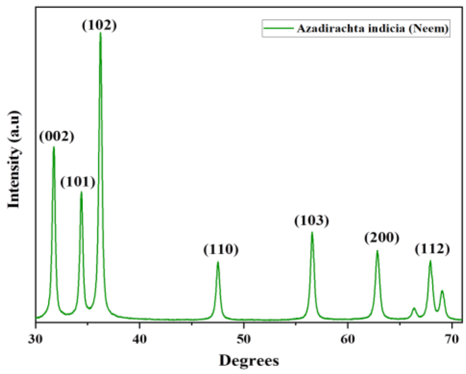
Peak Index	Peak Type	Area Intg (a.u.)	FWHM (degree)	Max Height (a.u.)	2th (degree)	Area IntgP (a.u.)	D (nm)	D Average (nm)
1	Gaussian	0.4471	0.3345	128432.10	31.72497	17.93681	23.75002	
2	Gaussian	0.4289	0.3359	92719.29	34.40506	13.69759	23.65108	**23.68173**
3	Gaussian	0.4665	0.336	210443.58	36.2159	30.65584	23.64408	



(a) ***Moringa Oleifera*** **leaves extract**



(b) ***Ocimum Tenuiflorum*** **leaves extract**



(c) Neem (*Azadirachta indicia*) leaves extract

Fig. (1) XRD patterns of nanoparticles of ZnO-NPs from different leaves extracts

Scanning electron microscopy (SEM) analysis: Following the confirmation of the XRD data, the sample was prepared for the SEM analysis. SEM photographs of ZnO nanoparticles at various resolutions are shown in **Figs (2), (3), and (4),** which illustrate the SEM results of ZnO nanoparticles extracted from the green leaves of *Moringa Oleifera, Ocimum tenuiflorum*, and Neem (*Azadirachta Indicia*). The produced products are spherical and crystalline, according to detailed structural characterizations. There are no holes and the particles have a flakes-like shape. The SEM pictures of zinc oxide nanoparticles show a dense collection of homogenous nanoparticles with a spherical form. It was discovered that the particles are not aggregated, but rather distributed, and that the average size of nano zinc oxide derived from *Moringa Oleifera, Ocimum tenuiflorum*, and (*Azadirachta Indicia*) was (64, 74, and 51 nm), respectively. The achieved results differ from those provided by **(Pal *et al.*, 2018)**, although the data acquired from Moringa Oleifera are the same. A particle size analyzer and scanning electron microscopy were used to examine the size and morphology of zinc oxide nanoparticles synthesized from Azadirachta indica and *Ocimum tenuiflorum* leaf extracts. In scanning electron microscopy (SEM), the average size of neem and OcIimum *tenuiflorum* was 101.6 and 122.4 nm, respectively, and size ranged from 100-300 nm for ZnO-NPs generated from *Ocimum tenuiflorum* leaves extract recorded ovoid shape and size ranging from 70-400 nm **(Ajayan and Hebsur 2020).**

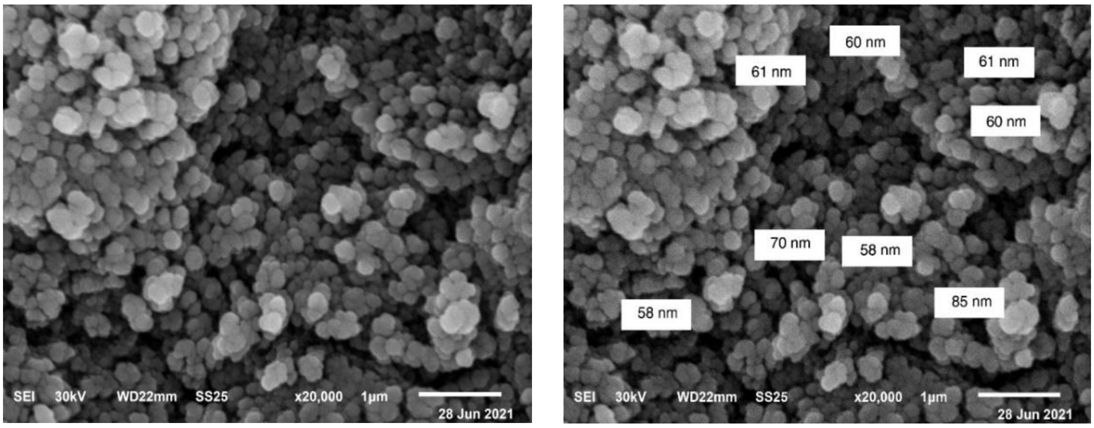


Fig. (2) SEM micrograph of ZnO-NPs nanoparticles from *Moringa Oleifera* leaves extract

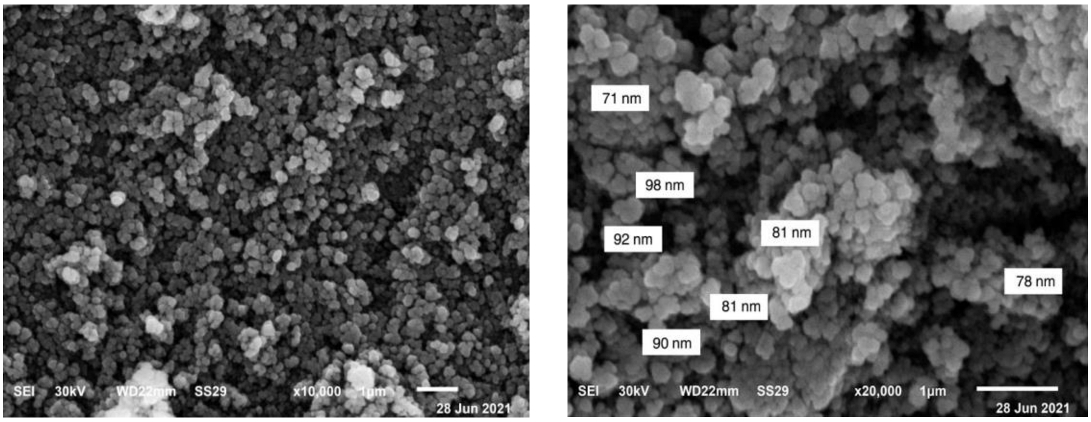


Fig. (3) SEM micrograph of ZnO-NPs nanoparticles from *Ocimum Tenuiflorum* leaves extract

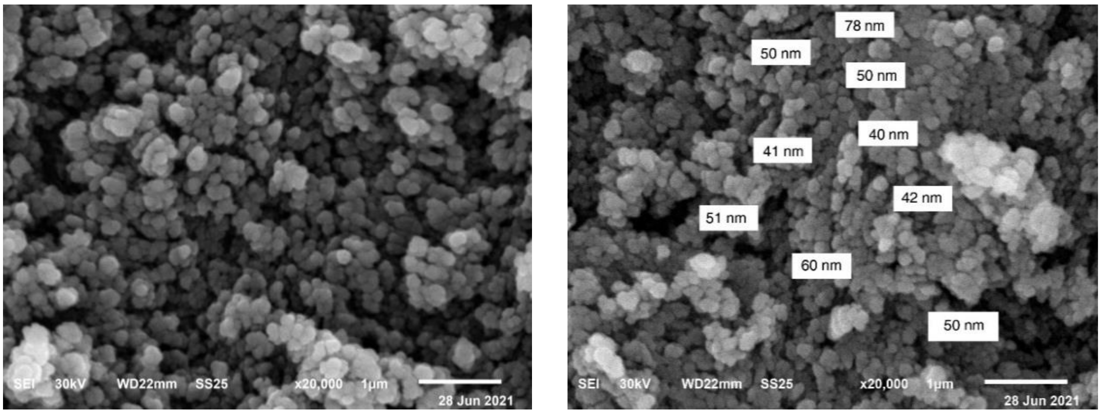
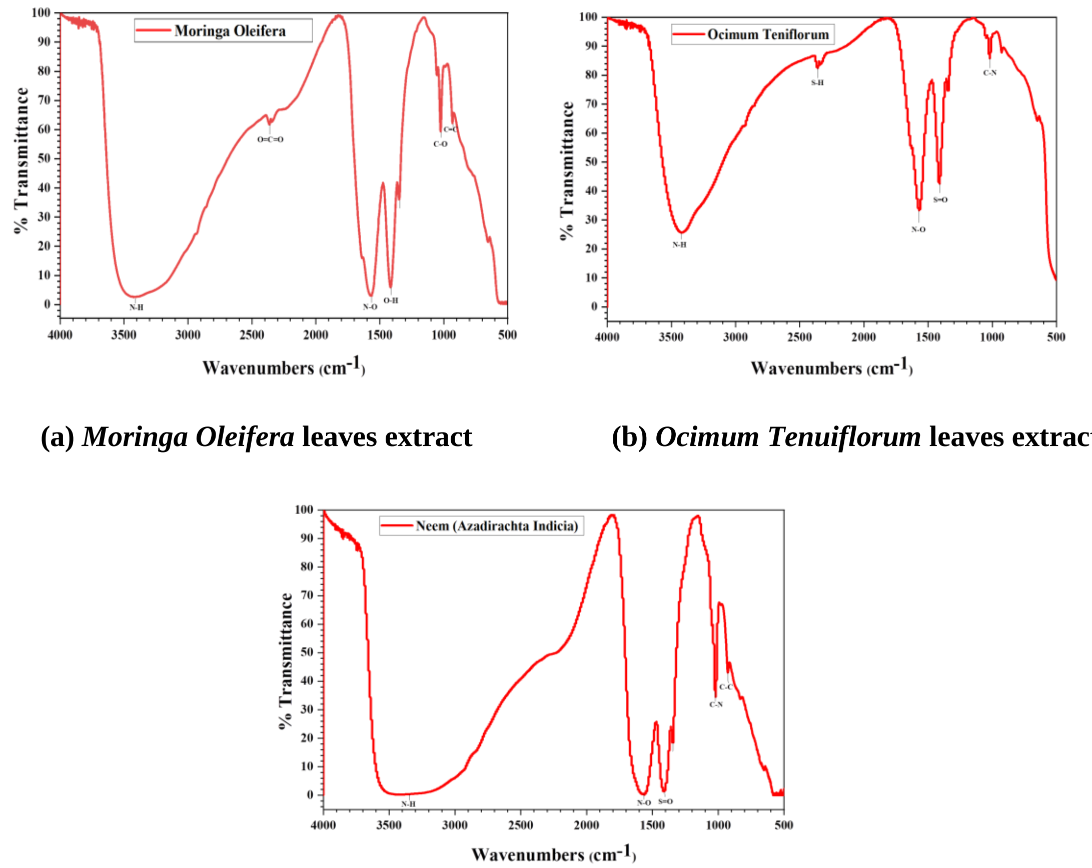


Fig. (4) SEM micrograph of ZnO-NPs nanoparticles from Neem (*Azadirachta indicia*) leaves extract

Fourier transform infrared spectroscopy Analysis (FTIR): functional groups can be provided on the surface The FTIR spectra of functionalized *Moringa Oleifera* leaves extract are given in **Fig (5),** with the peaks at 3405, 2365, 1562, 1409, 1020, and 928 cm^{-1}, respectively, assigned to N-H stretching, O=C=O stretching, N-O stretching, O-H bending, C-O stretching, and C=C bending. The data are identical to those reported by **(Pal *et al.*, 2018).** While the FTIR spectra of functionalized *Ocimum Tenuiflorum* leaves extract are presented in **Fig (7),** the peaks detected at 3418, 2366, 1568, 1410, and 1019 cm^{-1}, respectively, are attributable to N-H stretching, S-H stretching, N-O stretching, S=O stretching, and C-N stretching bending. The findings are consistent with those published by **Pal *et al.*, (2018) and Yasotha *et al.*, (2020).**

FTIR spectra of functionalized Neem (*Azadirachta indicia*) leaves extract are given in **Fig (5),** with peaks found at 3374, 1567, 1410, 1016, and 924 cm^{-1}, attributed to N-H stretching, N-O stretching, S=O stretching, C-N stretching, and C=C bending, respectively. The data are the same as those reported by **(Pal *et al.*, 2018).** These findings are similar to those published by **Khaing *et al.*, (2018),** who discovered in the FT-IR spectrum of aqueous neem extracts the existence of O-H stretching the frequency at 3377.41cm^{-1} from Neem leaves extract and the typical absorption peak of Zn-O bond at 832.30 cm^{-1}. The strong peaks in the XRD pattern clearly demonstrated the nanoparticles' great crystallinity. At 100°C, the crystallite size of the ZnO nanoparticles was determined to be 37.79 nm and 24.60 nm, respectively.



(a) ***Moringa Oleifera*** **leaves extract**

(b) ***Ocimum Tenuiflorum*** **leaves extract**

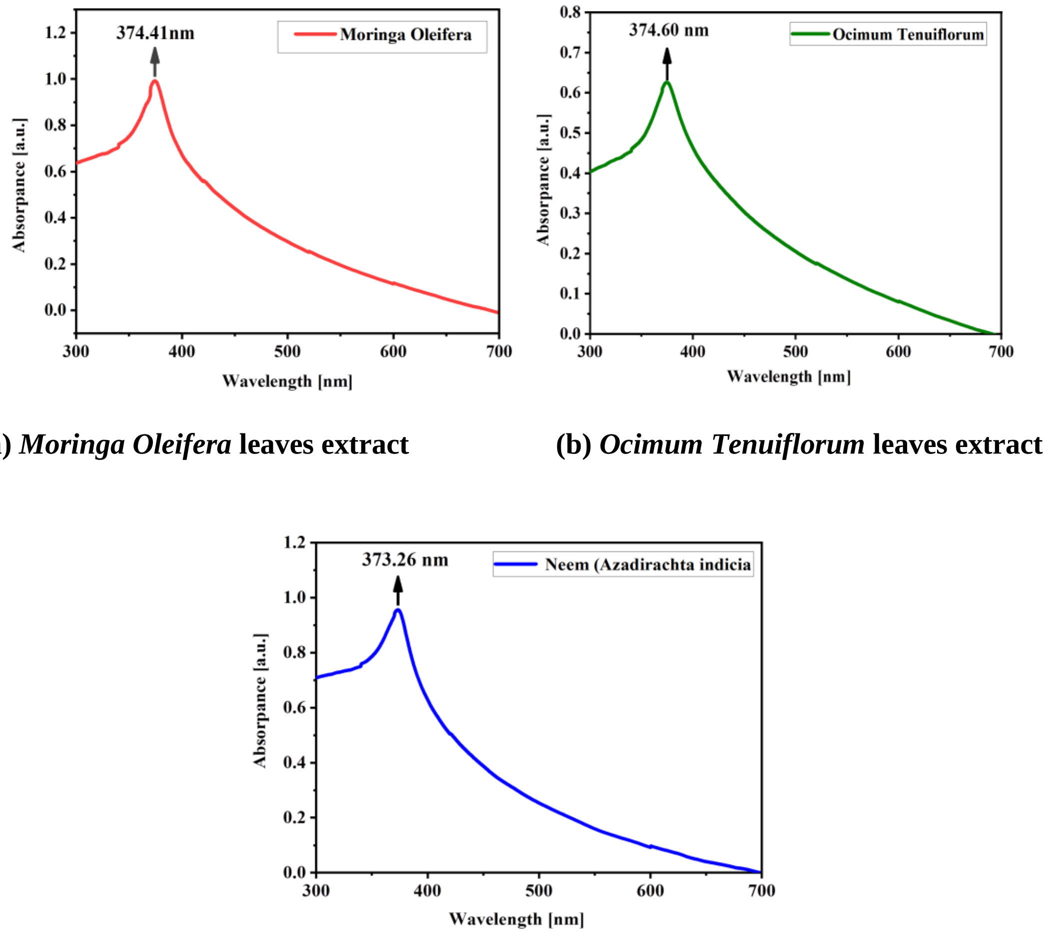
(c) Neem (*Azadirachta indicia*) leaves extract

Fig. (5) FTIR of ZnO NPs nanoparticles from different leaves extracts

UV-Visible absorption spectrum Analysis: UV-VIS absorption spectroscopy was used to investigate the optical characteristics of ZnO-NPs produced utilizing various extracts derived from green leaves. Natural extracts were obtained from *Moringa Oleifera*, *Ocimum Tenuiflorum*, and Neem (*Azadirachta indicia*), and the results are shown in **Table (6)** and **Fig (6).** Furthermore, a similar absorption edge was detected in natural extracts *Moringa Oleifera*, *Ocimum Tenuiflorum*, and Neem (*Azadirachta indicia*) at 374.41 nm, 374.60 nm, and 373.26 nm, respectively. The electrical transition from filled valence bands to empty conduction bands relates to ZnO absorption in the U.V range **(Musleh *et al.*, 2019).** These absorption edge values corresponded nicely with the data obtained by **(Shaat *et al.*, 2019).**

Table 6. The UV-Visible absorption spectrum analysis in green leaves extracts under investigation

Green leaves extract	Moringa Oleifera	Ocimum Tenuiflorum	Neem (Azadirachta indicia)
UV-Visible absorption (nm)	374.41	374.60	373.26



(a) ***Moringa Oleifera*** **leaves extract** **(b)** ***Ocimum Tenuiflorum*** **leaves extract**

(c) Neem (*Azadirachta indicia*) leaves extract

Fig. (6) UV-VIS absorption spectra for leaves extracted from pure ZnO-NPs

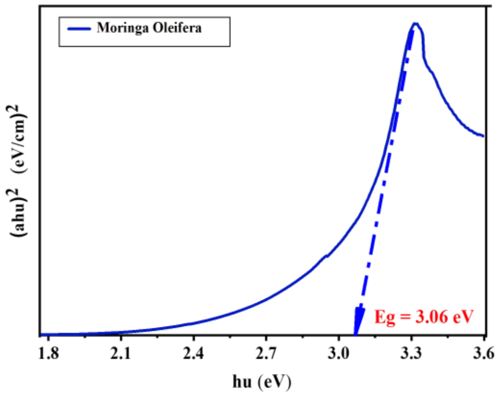
Estimation of optical band gap: The optical energy gap (Eg) in the wavelength between 300 nm to 700 nm of pure ZnO-NPs has been estimated from the UV-Visible absorption spectrum using the Tauc relation was an illustration in the following equation:

$$(\alpha h\nu) = B(h\nu - Eg)^n$$

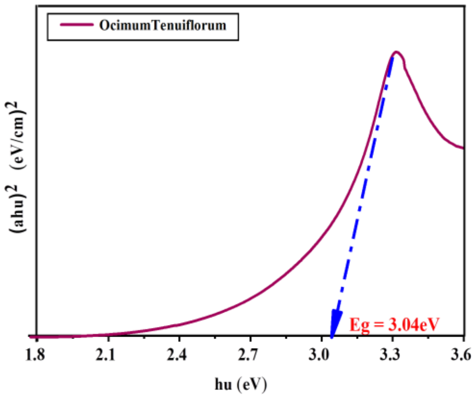
The opptical energy gap in ZnO is allowed to be a direct energy gap, therefore n=1/2. Extrapolating the linear portion of $(h\nu)^2$ in the y-axis against h, which intersects with the photon energy axis, yields an estimate of the optical energy gap. The acquired results are shown in **Table (7)** and **Fig. (7)** for the Eg of the produced ZnO-NPs samples. The calculated values of Eg for the samples *Moringa Oleifera*, *Ocimum Tenuiflorum*, and Neem (*Azadirachta indicia*) were 3.06 eV, 3.04 eV, and 3.14 eV, respectively. These values were consistent with the calculated values **(Shaat *et al.*, 2017).**

Table 7. The estimation of the optical band gap in green leaf extracts under investigation

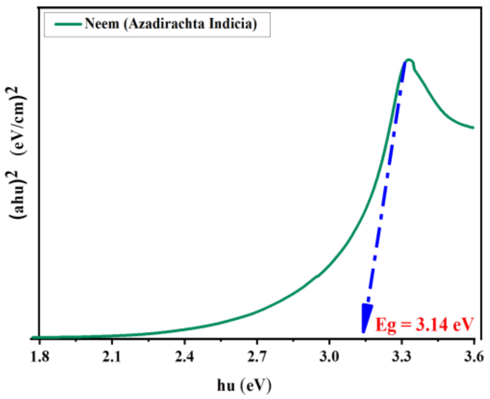
Green leaves extract	Moringa Oleifera	Ocimum Tenuiflorum	Neem (Azadirachta indicia)
Optical energy gab (ev)	3.06	3.04	3.14



(a) ***Moringa Oleifera*** **leaves extract**



(b) ***Ocimum Tenuiflorum*** **leaves extract**



(c) Neem (*Azadirachta indicia*) leaves extract

Fig. (7) Optical energy gap (Eg) for different leaves extracts

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تخليق وتوصيف جسيمات أكسيد الزنك النانونية من أوراق بعض النباتات

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يمثل التخليق الأخضر للجسيمات النانونية جزءاً مهم في تقنية النانو حيث تعد من التقنيات الحديثة التي تدخل في مجالات عديدة ويرجع ذلك الى خواصها الكيميائية والفيزيائية وارتفاع نسبة سطحها الى حجمها نظراً لتناهي صغرها، وبذلك تزيد من سطح تماسها مع الأجسام الأخرى لذلك فإن الهدف من هذه الدراسة تخليق جسيمات أكسيد الزنك النانونية وهو مركب غير عضوي له الصيغة الجزيئية ZnO وهو عبارة عن مسحوق بلوري أبيض مائل للصفرة قابل للذوبان في الماء تقريبًا، يوجد عده طرق مختلفة لتخليق أكسيد الزنك النانونية، ولكن تعد طريقة التخليق الخضراء من أبسط الطرق، وأقلها تكلفة، سريعة والآمنة بيئياً. تناولت هذه الدراسة أحدث الدراسات حول تخليق جسيمات أكسيد الزنك النانونية باستخدامات المستخلصات النباتية حيث تضمن العمل البحثي إستخدام مستخلصات أوراق المورينجا وأواق الريحان وأوراق النيم بهدف تعزيز الإستدامة البيئية وتشجيع إستبدال المنتجات الحالية بمنتجات النانو الجديدة الصديقة للبيئة خلال دورة حياتها وغير سامة، والتي لا تسبب أضرارًا للبيئة أو لصحة الإنسان. ولتحضير جسيمات النانو تم تجميع الأوراق الخضراء تحت الدراسة ومن ثم غسلها جيدًا وتجفيفها ثم عملية الطحن وحفظها لحين الإستخدام ثم عمل المستخلصات لتخليق أكسيد الزنك النانونية.

وتم دراسة التركيب الكيميائي لأوراق النباتات وتوصيف جزيئات أكسيد الزنك النانونية للتأكد من وصولها لحجم النانو حسب الدراسات السابقة وذلك بإستخدام الأشعة السينية، المجهر الإلكتروني الماسح، مطياف الأشعة تحت الحمراء، الأشعة فوق البنفسجية وفجوة الطاقة.

أوضح التركيب الكيميائي أن نسبة الرطوبة لأوراق المورينجا، وأوراق الريحان، وأوراق النيم هي على التوالي (9.37±0.01)، (10.60±0.00) و(9.50±0.01) جم/100جم مادة جافة بينما نسبة الرماد هي (8.60±0.01)، (14.50±0.01) و (11.62±0.01) جم/100جم مادة جافة لأوراق الأشجار على التوالي، وكذلك نسبة البروتين هي (13.50±0.01)، (7.42±0.01) و (8.56±0.01) جم/100جم مادة جافة لأوراق الأشجار تحت الدراسة على التوالي، ونسبة الدهون هي (7.30±0.01)، (6.52±0.01) و (6.96±0.01) جم/100جم جافة على التوالي ، بينما سجلت نسبة الكربوهيدرات اعلى نسب فكانت (61.32±0.01)، (60.96±0.02) و (62.36±0.18) جم/100جم مادة جافة لأوراق الأشجار المورينجا، وأوراق الريحان و أوراق النيم.

بينما كانت نتائج توصيف جسيمات النانو للأشعة السينية فأظهر البناء البلوري لأوراق المورينجا وكان متوسط حجم النانو 8.56 نانوميتر، ولأوراق الريحان 12.10 نانوميتر، بينما كان متوسط حجم النانو لأوراق النيم 23.68 نانوميتر. وكان متوسط حجم جسيمات النانو المستخدمة في الدراسة بالمجهر الإلكتروني الماسح هي (51، 84، 64 نانوميتر) لنبات المورينجا والريحان والنيم على التوالي. كما أظهر مطياف الأشعة تحت الحمراء المجموعات الوظيفية الفعالة لأوراق أشجار النباتات تحت الدراسة، والأشعة فوق البنفسجية يوضح طيف الامتصاص لأكسيد الزنك المحضرة من مستخلصات الأوراق لنبات المورينجا والريحان والنيم، وتم الحصول على أقصى امتصاص عند الطول الموجي (374.50، 374.70، 373.09 نانومتر)، على التوالي ، وفجوة الطاقة لمعرفة أعلى نطاق تكافؤ كانت (3.06, 3.04، 3.14 الكترون فولت) لنبات المورينجا والريحان والنيم على التوالي .

وبذلك يمكن خلاصه القول إن النتائج المتحصل عليها من دراسة تخليق وتوصيف جسيمات النانو بإستخدام أوراق النباتات تحت الدراسة المختلفة من أوراق المورينجا وأوراق الريحان وأوراق النيم أنها تعطي متوسط حجم النانو والمجموعة الوظيفية الفعالة وطيف الأمتصاص وكذلك فجوة الطاقة وبهذا يحقق الهدف من تطوير أفضل الطرق لتخليق جسيمات أكسيد الزنك النانونية الأقل تكلفة وانخفاض سميتها بالمقارنة بالطرق التقليدية سواء الكيميائية أو التطبيقية الأخرى.