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Dielectric Properties Of Zinc Oxide Nanoparticles Synthesized With Terminalia Mantaly Leaf Extract.

¹B.Lavanya, ¹M.V. Ramana, ²Y.Aparna

¹Assistant Professor, ¹Professor, ¹Professor ¹Department of Physics, Anurag University, Hyderabad, INDIA ²Jawarharlal Nehru Technological University, INDIA

Abstract: ZnO nanoparticles are being used in number of industrial products such as rubber, paints, coatings, and cosmetics. Here we report the green synthesis method for the preparation of ZnO nanoparticles from Terminalia mantaly leaf extract. The size of the particles and morphology of the synthesized ZnO nanoparticles are characterized by using SEM, FTIR and X-ray Diffraction. It was observed that the size of the nanoparticles ranged from 14-18 nm.. The present study confirms the eco-friendly green synthesis of the ZnO nanoparticles using Terminalia Mantaly Leaf extract.. Dielectric measurements have been carried out on the ZnO nanoparticles in the form of sintered pellet in the frequency range 1Hz-100MHz at room temperature. The value of dielectric constant ($/\!\!\!/\!\!\!/\!\!\!/ K$) and dielectric loss tangent (tan δ) were found to be decreasing with increase in frequency.

Keywords: Terminalia mantaly leaves; Green synthesis; ZnO Nanoparticles; Dielectric studies.

I. INTRODUCTION

Nanoparticles of different shapes have different applications in fields like chemical industries, biomedical, cosmetics, drug delivery, electronics, environment, health care, mechanics, optics, energy science, food, space industries, etc. due to their size, distribution and morphology. [1]-[7]. Metallic nanoparticles are used in the construction of fuel cells, piezoelectric devices, sensors, micro-electronic circuits, coatings against corrosion and as catalysts [8]-[10]. A number of research groups have carried out work on the synthesis and characterization of zinc oxide nanoparticles using various synthesis methods. Among all these methods, green synthesis is preferred because it is less expensive, readily available, and non-toxic[11]-[13] in general. In particular nanoparticles were synthesized using different parts of plants like leaves, fruits, flowers, stems, barks, roots, etc. Zinc oxide nanoparticles were synthesized using different leaf extracts from Pongamia pinnata, Agathosma betulina, Aloe vera, Tamarindus indica, Cassia fistula, tribulus terrestris, etc.[14]. In the present study, zinc oxide nanoparticles were synthesized using leaf extract of the Terminalia mantaly plant.

Plants have great genetic diversity in terms of biomolecules and metabolites such as proteins, vitamins, coenzyme-based intermediates, phenols, flavonoids, and carbohydrates. These plant metabolites have hydroxyl, carbonyl, and amine functional groups, which react with metal ions to reduce their size to the nano range. Flavonoids, in particular, have multiple functional groups, and it is believed that the OH group of flavonoids is primarily responsible for the reduction of metal ions into nanoparticles. Metals like silver (Ag), copper (Cu), gold (Au), and others have been widely used in the biosynthesis of nanoparticles using plant extracts from various plant species[15]. Zinc oxide nanoparticles are used in industrial applications like rubber, paint, coatings, and cosmetics. Because of its large band gap of 3.37 eV, large binding strength, and large excition binding energy (60 meV) at room temperature, ZnO is used for short wavelength optoelectronic applications[16]-[19]. Zinc oxide nanoparticles have drawn attention due to their unique antibacterial, antifungal, UV filter properties, and high catalytic and photochemical activity [20], [21].

Among the various plant species used in Cameroon's traditional medicine, Terminalia mantaly(TM) has gained scientific recognition for its reported antitumor, antidiabetic and antihypertensive drug properties[22]. Terminalia mantaly is a deciduous tree belonging to the family Combretaceae, a flowering plant. The genus Terminalia includes more than 100 species. Many scientific studies have examined antibacterial, antiprotozoal, antidiarrheal, anti-inflammatory, anti-tumor and wound healing properties[23]. Several studies have demonstrated the synthesis of biogenic Silver nanoparticles using different species of the genus Terminalia, which include Terminalia catappa, Terminalia chebula, Terminalia arjuna, and Terminalia bellirica. Terminalia mantaly leaf extract has been used to produce biogenic gold and silver nanoparticles [28]. Vindhya et al [29] have reported the Dielectric properties of zinc oxide nanoparticles using annona muricata leaf. As per the research survey, it was observed that this Terminalia mantaly has not been used till date for the synthesis of ZnO nanoparticles. This article reports the synthesis, characterization and dielectric properties of ZnO nanoparticles using Terminalia mantaly leaf extract and their antibacterial properties.

II. EXPERIMENTAL

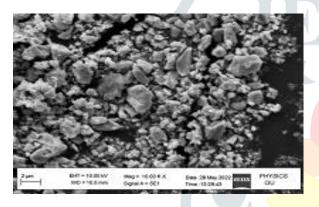
Zinc Nitrate hexahydrate, Deionized water were used throughout the process, Terminalia mantaly leaves were collected from Venkatapur, Ghatkesar and identified by Department of Botany, Kakatiya University, Warangal. Leaves were collected from Terminalia mantaly plants and washed thoroughly by using double distilled water to make them dust free. These leaves were dried at room temperature and then finely powdered by using a grinder. 10g of powder is added to the 200 ml of distilled water in a 500ml conical flask. Then it is stirred on a magnetic stirrer at 80°C for one hour. The aqueous leaf extract was filtered by using Whatmann No 1 filter paper and stored at 40° C for further experiments. 3g of Zinc Nitrate is added to 30 ml of leaf extract and stirred about 2 hours at 60° C. The yellowish paste that formed was carefully collected in crucible and heated in a furnace at 400°C for two hours. Finally white coloured powder was obtained. This was collected and preserved for characterization studies and antibacterial activities.

The Synthesized ZnO nanoparticles were characterized by using scanning electron microscope(SEM), EDX, X-ray diffraction(XRD) and Fourier Transform Infrared Spectroscopy(FTIR). The dielectric properties were performed on sintered and polished samples with Multi-frequency Hioki 3532-50 LCR Hi-Tester in the frequency range 1 Hz to 100 MHz at room temperature.

III. RESULTS AND DISSCUSSION

Scanning electron microscope(SEM)

Scanning electron microscope is used to investigate the microstructure. From the SEM image, it was observed that ZnO nanoparticles are spherical in shape. These ZnO nano particles with spherical shape were also agglomated with different shaped ZnO particles. Non-Spherical ZnO nanoparticles were also observed in the present SEM. The size of the Zinc oxide nanoparticles are in the range of 100-200nm. Fig.1 and 2 presents the SEM images of ZnO nanoparticles synthesized in this



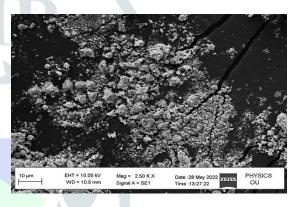


Figure-1 Figure-2

Fig.1 and 2. SEM images of ZnO nanoparticles at different magnifications.

Fig.3 shows the EDX of the ZnO nanoparticles. This reveals the presence of elements in the sample. The EDX shows the presence of Zn and O peaks exclusively. This confirms the purity of the sample, which was reflected as 80.11wt% of Zn and 19.89wt% of O.

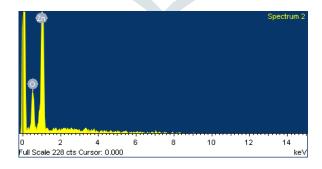


Fig.3 EDX spectrum of synthesized ZnO nanoparticles.

ii) X-RAY DIFFRACTION

XRD spectra of ZnO nanoparticles synthesized using Terminalia Mantely leaf extract was presented in Fig.4. It was observed that all peaks obtained in pattern were in close agreement with the reported peaks of ZnO in literature. [30] and confirms the formation of ZnO. The XRD data is presented in table-1. To find the Particle size Debye Scherrer equation is used and it is given by $D = K\lambda/\beta\cos\theta$ where D is the size of the particle, K is Debye Scherrer constant, λ is the wavelength of X-rays, and θ is the diffracted angle. The calculated particle sizes were found out to be in the range of 13-27nm.

Table-1 XRD data of ZnO nanoparticles

No. of Peaks	Planes	Pos. [°2Th.]	FWHM [°2Th.]	Particle
				Size(nm)
1	(100)	31.5126	0.4723	17.47
2	(002)	34.2042	0.4723	17.59
3	(101)	36.0124	0.4723	17.68
4	(102)	47.3407	0.4723	17.75
5	(110)	56.3785	0.4723	19.08
6	(103)	62.6794	0.3936	16.66
7	(200)	66.0579	0.6298	23.63
8	(112)	67.6931	0.9446	15.0
9	(201)	68.9302	0.4723	13.32
10	(004)	72.4328	0.4723	13.61
11	(202)	76.7895	0.4800	21.11
Particle average				15.38nm
		size		

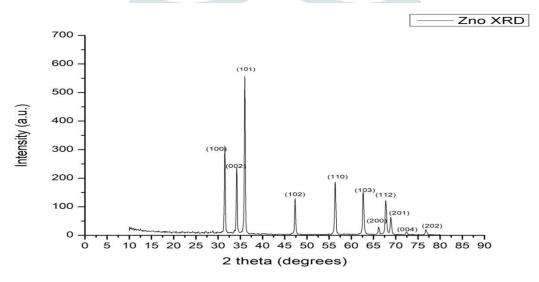


Fig. 4. XRD spectra ZnO nanoparticles from TM leaf extract.

iii) Fourier Transformation Infrared Spectroscopy

The FTIR spectrum of ZnO nanoparticles prepared by Terminalia mantaly leaves is recorded in the range of 173-4075 cm⁻¹ and is presented in fig.5 The observed spectral peaks were assigned to the different functional groups, modes and bonds, after comparing with the reported literature. FTIR the peaks formed at 470 cm⁻¹ and 436 cm⁻¹ confirm the presence of ZnO nanoparticles.

S.NO	WAVE NUMBERS		BOND TYPE	FUNCTIONAL GROUP
	Theoretical	practical		
1	3700-3640	3665,3644,,3632,3624, 3614	O-H Stretching	ALCOHOLS AND PHENOLS
2	3200-3500	3499,3478	O-H STRETCH,H- BOUNDED	ALCOHOLS AND PHENOLS
3	3350-3500	3443	N-H stretch	AMINES
	3300	3299	O-H Stretch	CARBOXYLIC ACID
	2850-3000	2889	C-H Stretch	ALKANES

	<u> </u>	•		, 0
4	1760-1690	1765	C=O stretch	CARBOXYLIC ACID
5	1735-1750	1735	C=O stretch,	ESTERS,SATURATED ALIPHATICS
6	1640-1680	1644,1651,1661	-C = C- stretch	ALKENES
7	1450-1470	1446	C-H BEND	ALKENES
8	1290-1360	1384	N-O Symetric stretch	NITRO COMPOUNDS
9	1020-1250	1044,1112	C-N stretch	ALIPHATIC AMINES
10	550-850	839,826	C-Cl stretch	ALKYL HALIDES

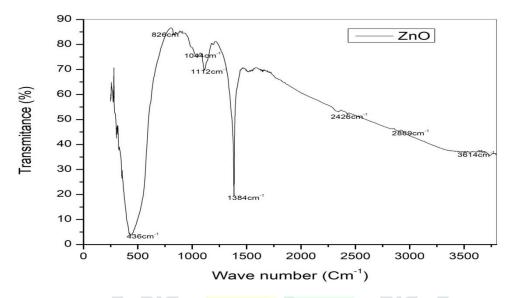


Fig.5. FTIR Spectrum of ZnO nanoparticles.

iv) DIELECTRIC PROPERTIES

Dielectric constant and loss tangent were studied in the frequency range of 1Hz to 100MHz at the room temperature. The variation of dielectric constant and loss factor with frequency are shown in fig. 6 and 7.. Both dielectric constant and loss factor decreases with increase of frequency. It is expected that at low frequencies orientation of space charges creates a dipole moment in the direction of field. This results the space charge polarization. It is also expected that the dipoles were able to align themselves resulting total polarization and high dielectric constant. Dielectric loss attains a constant value at higher frequency. This phenomenon was well explained by hopping model in the literature According to this model, the presence of low mobility carriers in the material cause space charge hopping between two localized levels where each level has space and charge. At higher frequencies the space charge polarization occurs very rapidly but medium responds at lower frequencies in comparison to the tunneling time. This produces strong conductivity dispersion at low frequency region of ZnO nanoparticles [31, 32]. The measured values of dielectric constant and loss tangent was found to nearly 10.8 and 2.83×10⁻³ respectively at room temperature in the frequency of 1MHz. These values are very close to that of reported by Vindhya et.al [29].

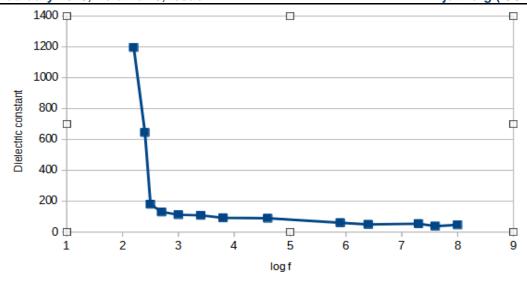


Fig..6. Variation of dielectric constant with frequency

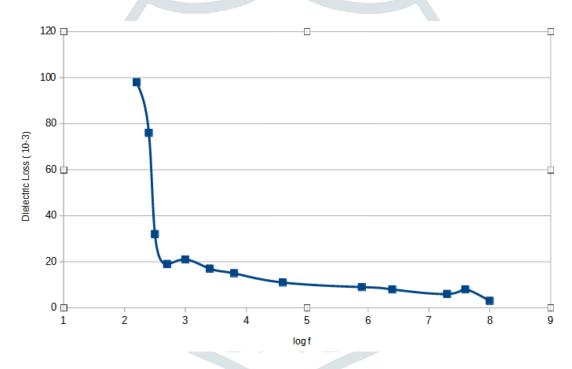


Fig..7. Variation of dielectric loss (tan δ) with frequency

IV. CONCLUSIONS

ZnO nanoparticles were successfully synthesized by using Terminalia mantaly leaf extract for the first time. Terminalia mantaly leaf extract was found be equally good when compared to other reported leaf meditated synthesis reported till date. It is observed that ZnO particles of the size as low as 13+5nm could be synthesized using Terminalia mantaly leaf extract. XRD analysis revealed that ZnO has hexagonal wourtzite structure This was evident from the characterization studies made on the synthesized ZnO particles by using XRD, SEM, EDX, and FTIR techniques. The range of the nanoparticles produced using Terminalia mantaly leaf extract was found to be almost uniform, within error limits, in the range 13nm to 23nm. This appears that Terminalia mantaly leaf extract was a potential candidate for bulk synthesis of ZnO nanoparticles using green methods. The synthesized ZnO nanoparticles show very good antibacterial activity against E-coil and B-coil. It can be concluded, based on the results and in comparision with the reported literature that Terminalia mantaly leaf extract can be best used in production of ZnO particles as this technique is eco-friendly, stable and in least time consuming and hence can be used in industrial bulk production. Frequency dependence dielectric properties have been studied. Dielectric constant and loss tangent was found to be ~11 and 360-10-3 respectively in the frequency of 1MHz at room temperature. The lower dielectric loss property of these ZnO nanoparticles can be used for the fabrication of low-loss microwave electronic circuits. The bulk production of ZnO nanoparticles and nanostructures using Trminalia Mantaly and the study of effect of concentration of leaf extract on the size and morphology of ZnO nanoparticles is under progress as the continuation of this work and will be reported soon.

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