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Studying the Effect of Zn_2SnO_4 Nanoparticles on Antibacterial Activity Using the Sol-Gel Method

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Abstract

In this study, zinc stannate Zn_2SnO_4 (ZTO) nanoparticles were prepared using the sol-gel method. X-ray diffraction technique was used to investigate the Zn_2SnO_4 nanoparticles, which showed that they have a polycrystalline structure and were of the cubic type. Field emission scanning electron microscopy (FE-SEM) measurements showed that the grains were nanostructures with regular shapes, and their average atom dimensions were 162.01 nm. Transmission electron microscopy (TEM) showed the appearance of nanosheets and others resembling nanoscale tilts. Zn_2SnO_4 nanoparticles showed biological activity against pathogenic bacteria and showed a difference in the average diameter of inhibition for all types of used bacteria, and the average diameter of inhibition ranged from 11 to 17 mm.

Keywords: Zn_2SnO_4, Sol-Gel, XRD, TEM, FE-Inhibition bacteria.

Introduction

Zinc stanate (Zn_2SnO_4) is an n-type semiconductor that has been prepared by many researchers. It has cubic structures and has an energy gap of (3.7eV) compared to other semiconductor oxides. (Zn_2SnO_4) has exceptional qualities, including chemical stability that make it useful as a foundation material in a variety of applications, high electron mobility (15^{-10} $cm^2/v.s$) as well as high electrical conductivity [1]. Recent years have seen a surge in interest in zinc stannate because of its potential applications in a wide range of fields,

including filters, solar cells, humidity and gas sensors, and more [2]. Zinc stannate, a prominent transparent conducting oxide, is a wonderful functional material. Pure forms aren't always intriguing [3]. Adding impurities can change material characteristics by creating localised states and flaws. Similarly, blending two materials creates a new compound with unique features [4]. Industrial TCO materials are made by mixing zinc oxide (ZnO) with tin oxide (SnO_2) [5].N-type semiconductors made of metal oxides are the main focus of research on ultratransparent and conductive thin films [6]. Many techniques were used to create ZnO/SnO_2 films, powders, nanowires, and nanorods [7], whose optical and electrical characteristics have been extensively studied [8].

Sol -gel technique is a process of deposition of a wet chemical solution, and it takes place in several stages, such as hydrolysis, gelation, polymerization, condensation, and drying [9]. The colloidal solution method is considered one of the important methods and it is significant in the manufacture of nanomaterials, and work includes the transition from the chemical solution phase (Sol) to the solid phase (Gel) [10]. The benefit of this method currently is the preparation of nanomaterials of various shapes, such as powders with very small particles, as well as porous materials that are easy to penetrate, called (Aero-Gel) [11]. There are many factors that must be taken into consideration in this method, such as controlling the pH, which is an important factor to avoid precipitation, as well as its benefit in forming a homogeneous gel that can be manufactured by adding a base or acid to the solution [10.].

At the end product of the hydrolysis of zinc stannate nanoparticles. By utilising a variety of synthetic techniques, including hydrothermal, co-precipitation, solvo-thermal, and others, it is possible to enhance their characteristics with exceptional microstructures [12]. The wet chemical method, which has low processing temperatures and great molecular homogeneity, is one of these sol-gel procedures. Customised final product form and microstructure are possible by controlling the forced hydrolysis process [13].

The past several years have seen an increase in microbial contamination as a result of the expanding scientific knowledge base. There have been many different materials employed to stop their development. Zinc stanate (Zn_2SnO_4) is among these materials [14]. The antibacterial effectiveness of ZTO NPs (Zn_2SnO_4), which were made using the hydrothermal

technique with zinc and tin chloride salts as the precursors, has been documented by Pandimurugan and Sankaranarayanan [15]. Gram-positive (S. aureus, Bacillus subtilis) and gram-negative (K. pneumonia, E. coli) bacteria have both shown potential antibacterial action when exposed to the NPs. A study conducted by Krishnasamy *et al*. [16] focused on preparing Zn_2SnO_4 and characterizing its structural properties, which developed a wide range of optical behavior for the Zn_2SnO_4 nanomaterial and tested its antibacterial activity using microwave technology. They came to the conclusion that these compounds have the potential to curb the spread of infectious illnesses by acting as effective antibacterial agents. The purpose of the study is to know the effect of the substance on the resistance of disease-causing germs.

Material and Method

1- Preparation of Zn_2SnO_4 by Sol-gel Method

A mixture of aqueous zinc nitrate ($Zn(NO_3)_2.6H_2O$) at a concentration of 0.1 M was mixed with aqueous tin chlorides ($SnCl_4.5H_2O$) at a concentration of 0.05 M, and then citric acid was added at a molar concentration of 0.5 M ($C_6H_8O_7$) with a molecular weight of 19.2123 g/mol. After completely dissolving the solution, the pH was measured and found (pH = 1.4). In order for the solution to be neutral, a solution of ammonia (NH_4OH) with a concentration of 25%, prepared by the SIGMA company, was added in droplets to the prepared solution at intermittent intervals while the solution was stirred on the magnetic mixer until the solution became neutral (pH = 7±0.05) and the solution turned white. After the solution becomes neutral, the temperature of the magnetic mixer is turned on until the solution temperature reaches 80 °C. The temperature is stabilized, and the solution is left on the magnetic mixer for (50-60) minutes in order for the liquid to gradually evaporate until it turns into (gel). The compound (Zn_2SnO_4) was obtained as a result of the following chemical reaction [17]:

$2Zn\ (NO_3)_2.6H_2O + SnCl_4.5H_2O \rightarrow Zn2SnO4 + 3NO_2 + 4HCl + 11H_2O + O_2$

After that, the reaction products are slowly dried using an electric dryer for 24 hours in order to remove all water molecules and suspended liquids. The resulting minutes are then placed inside an electric oven at a temperature of 550 degrees for two hours, and the material is left inside the oven for 24 hours. An hour until its temperature reaches room temperature, after which the powder (Zn_2SnO_4) was obtained.

2- Characterization of Zn_2SnO_4

- **X-Ray Diffraction (XRD)**

To study the nature of the crystalline structure of the prepared sample and to identify the effect of doping on films [18], an X-ray diffraction device was done in the service laboratory - University of Technology in Baghdad, Iraq. TYPE: XRD-6000, SHIMADZU.

- **Transmission Electron Microscope (TEM)**

A transmission electron microscope represents a microscopic technique in which a beam of electrons is used to examine and test samples. The image was formed by the electrons penetrating through the sample and was magnified and focused by an objective lens, then displayed on an imaging screen. This screen in most transmission microscopes was in the form of a monitoring screen, or the image was displayed on photographic film, or the image was detected by a sensitive detector such as a CCD camera. The A-STEM device equipped with a 3rd-order spherical aberration corrector was used in this test, and the examination was conducted at the University of Tehran, Iran [18].

- **Field Emission Scanning Electron Microscopy Measurements (FE-SEM)**

(FE-SEM) provides Morphological and elemental information at magnifications of 10x to 300,000x, with virtually unlimited depth of field. Both types of devices (Model TE-SCAN) were used. (MIRA3)). These tests were carried out at the University Of Tehran, Iran [19]

- **Biological effectiveness against gram positive and Gram bacteria**

1- Samples Collection

Collection, isolation, and diagnosis of bacteria, gram-positive (*S.epidermidis* and *S.aureus*) and gram-negative (*E.coli*, *P.aeruginosa*), that cause diseases by specialists in educational laboratories, Baqubah Teaching Hospital, Diyala, Iraq

2- Culture of bacterial isolates

Staphylococcus aureus and *Staphylococcus epidermidis* isolates were cultured on blood agar, mannitol and salt isolates, *Escherichia coli* and *Pseudomonas aeruginosa* on MacConky agar and Pseudomonas agar isolates, as the turbidity standard (Mac-Farland) was the preparation solution from the company (Biomeriex) and was used to calibrate a number of bacterial cells, which gives an approximate number of cells (1.5×10^8 cells/ml) [20].

3- Antibacterial activity of Zn_2SnO_4 against Bacteria

In order to make Muller Hinton Agar medium, you need to dissolve (38 g) in a litre of distilled water, sterilise it in an autoclave at 121 °C and 15 P for 15 min, cool it, and then transfer it to sterile plates [21]. The diffusion method was adopted to determine antimicrobial activity, as follows [22]:

Several bacterial colonies were transferred by loop into tubes containing brain herat infusion broth to activate the suspended bacteria. Incubation of the tubes occurred at 37 degrees Celsius for 18 to 24 hours. After comparing the suspended bacteria to the normal Mac-Farland solution (1.5108 cells/ml), the germs were distributed using a sterile brush over Mueller Hinton agar plates and let to dry. Four wells were made with a drop (5 ml) in the culture medium using a sterile cork drill. 100 (µL) of Zn_2SnO_4 was added at concentrations (100, 75, 50, and 25 g/ml), each of which was individually perforated using a micropipette. By measuring the size of the inhibition zone surrounding each well, it was possible to assess how effectively each concentration worked.

1. Structural Measurements of Zn_2SnO_4

- **X-ray Diffraction (XRD)**

The results of the diagnosis using X-ray diffraction technology for the Zn_2SnO_4 nanoparticles showed that they have a polycrystalline structure and are of the cubic type, as shown in Figure1. By analyzing these curves, the locations of the peaks became clear, and we noticed the appearance of the levels (311, 222, 420, 422, 531) and that the prevailing growth trend was (311, 222). The current study found that these results were largely consistent with the standard card (ICDD) with serial number (0381-014-00), and this was consistent with the study done by Taşer *et al*. [23] and as shown in the table (1), as well as the appearance of the plane (100) belonging to the compound ZnO at the angle (31.9°), which matches the values in the card (01-079-0205), and the plane (110) belonging to the compound SnO_2 at the angle (26.8°), which matches the values in Card (01-072-1147).

The size of the grains was calculated, and it was found that it was within the nanoscale, as its lowest value was (10 nm) at the Plane (311), then it increased until it reached the highest

value at the plane (420), and then it decreased until it reached the lowest value at the level (531) using relation (1) [24] .

$$D = \frac{k\lambda}{\beta cos\theta} \qquad (1)$$

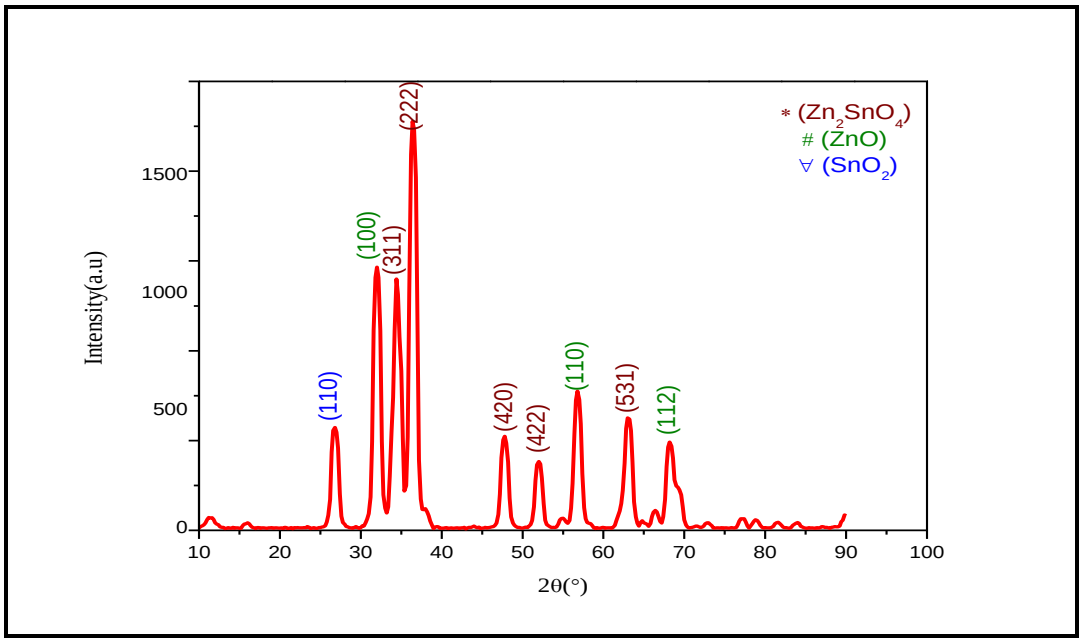

Figure 1: X-ray diffraction of Zn_2SnO_4 nanoparticles

Table 1: Values of structural parameters of Zn_2SnO_4 nanoparticles prepared by Sol-Gel method.

2θ (Deg.)	FWHM (rad.)	d_{hkl} (Å) Exp.	d_{hkl}(Å) Stand.	C.S (nm)	hkl	No.Card
26.8	**0.421**	**3.356**	**3.349**	**20**	**110**	**01-072-1147**
31.9	**0.290**	**2.798**	**2.807**	**30**	**100**	**01-079-0205**
34.52	**0.848**	**2.599**	**2.590**	**10**	**311**	**00-014-0381**
36.48	**0.510**	**2.466**	**2.490**	**17**	**222**	**00-014-0381**
47.7	**0.200**	**1.903**	**1.930**	**45**	**420**	**00-014-0381**
51.9	**0.245**	**1.760**		**38**	**422**	**00-014-0381**
56.83	**0.266**	**1.621**	**1.620**	**35**	**110**	**01-079-0205**
63.11	**0.484**	**1.473**	**1.463**	**20**	**531**	**00-024-1470**
68.20	**0.296**	**1.373**	**1.089**	**34**	**112**	**01-079-0205**

- **Field Emission Scanning Electron Microscopy Measurements (FE-SEM)**

FE-SEM pictures and a grain distribution diagram of the Zn_2SnO_4 nanopowder made with the Sol-Gel method can be seen in figures (2) and (3). The electron microscope images indicate

the formation of what resembles nanostructured sheets (Nano Plate) with an average dimension of 162.01 nm, and these results match previous studies [25].

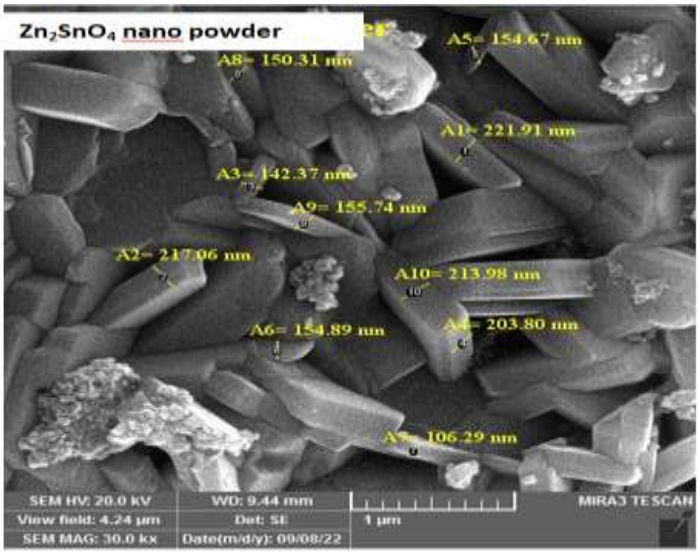

Figure 2: FE-SEM image of Zn_2SnO_4 nanoparticles

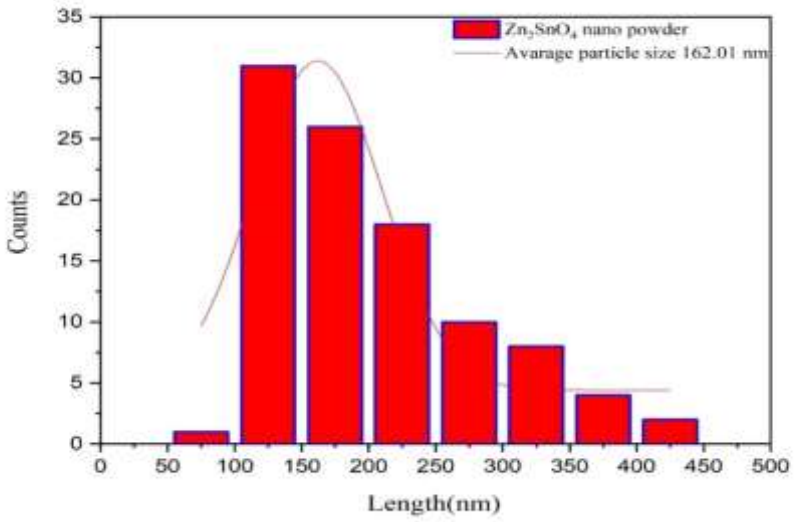

Figure 3: Particle dimensional distribution chart for nanopowder (Zn_2SnO_4)

- **Transmission Electron Microscope (TEM)**

Further investigations were conducted to study the formation of Zn_2SnO_4 nanopowder using transmission electron microscopy. Figure 4 shows TEM images of Zn_2SnO_4 nanopowder,

which was prepared by the sol-gel method. It shows the appearance of nanosheets resembling a structure and others resembling nanoscale tilts. This indicates that the results match the scanning electron microscope (SEM) images and that the nanopowder was formed from nanoplates [26].

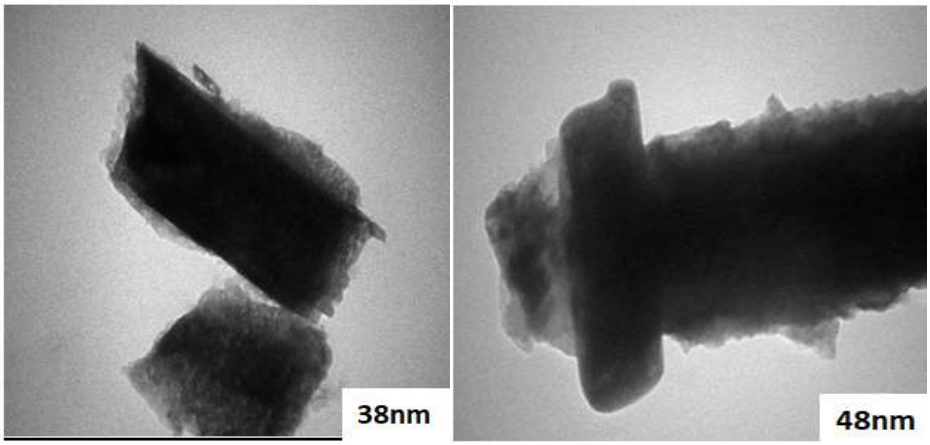

Figure 4: TEM image of Zn_2SnO_4 nanoparticles prepared by Sol-Gel method

- **Testing Zn_2SnO_4's antibacterial efficacy**

The well diffusion method was used to quantitatively assess the antibacterial activity of Zn_2SnO_4 nanoparticles, which had an average crystal size of 20.95 nm. The four distinct Zn_2SnO_4 nanoparticle concentrations that were tested against Gram-positive bacteria (S. aureus and S. epidermidis) are displayed in Table 2, and Gram-negative bacteria (*E.coli*, *P.aerugionsa*). Based on the observations, Figure 5 shows a biocidal inhibition zone for Zn_2SnO_4 nanoparticles. The inhibitory zone grows dramatically with nanoparticle concentration. The highest zone of inhibition for Gram-positive S.aureus bacteria is 17 mm, and Zn_2SnO_4-NPs shows modest antibacterial efficacy for pneumonia. S. epidermidis exhibits strong resistance to all Zn_2SnO_4-NP addition concentrations. Even though E. coli was shown to have a smaller inhibition zone (13 mm), Zn_2SnO_4-NPs' toxicity varies with concentration; at low concentrations, these NPs are moderately hazardous. The location, size, surface modification, inherent features, and kind of bacteria all affect a nanoparticle's toxicity mechanism [27]. Through electrostatic contact, NPs attach to the bacterial cell membrane and cause damage to the cell's integrity. This, in turn, increases the permeability of the cell and

ultimately results in cell death. The inherent poisonous qualities of heavy metals determine the toxicity potency of nanoparticles, whereas the toxicity resulting from dissolved Zn and Sn ions in the NPs is negligible [28].

Table 2: Bacterial species with concentrations and inhibition zone of Zn_2SnO_4 granules.

Type of bacteria	Concentrations and Inhibition zone			
	%25	%50	%75	%100
S.aureus	R	11mm	14mm	17mm
S.epidermidis	R	R	R	R
E.coli	E	R	11mm	13mm
P.aeruginosa	R	R	R	R

R= Resistance

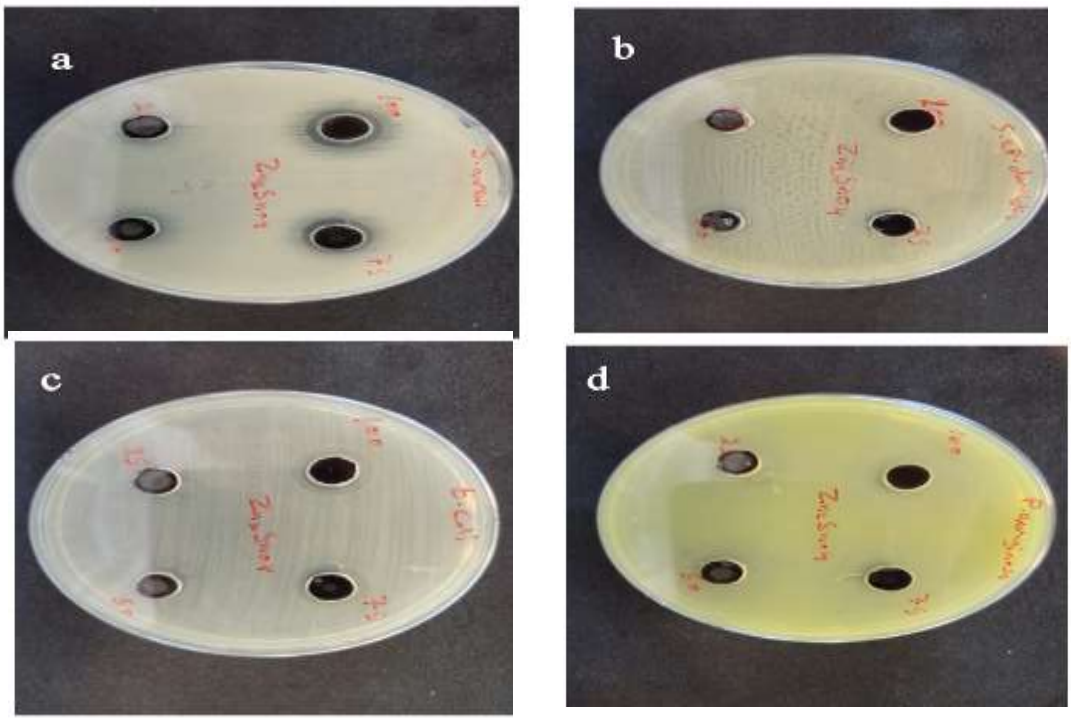

Figure 5: inhibition zone of bacteria by Zn_2SnO_4 nanoparticles.
a- *Staphylococcus aureus* b- *Staphylococcus epidermids* c- *Escherichia coli*
d- *Pseudomonas aeruginosa*

Conclusion

Zinc-stannate Zn_2SnO_4 (ZTO) nanoparticles were prepared using the sol-gel method. X-ray diffraction measurement showed that they have a polycrystalline structure and are of the cubic type. The size of the grains was calculated, and it was found that it was within the nanoscale. Field Emission Scanning Electron Microscope (FE-SEM) images indicate the formation of

what resembles nanostructured sheets (Nano Plate) with an average dimension of 162.01 nm. The biological effectiveness of ZTO nanoparticles against pathogenic bacteria showed that there was a difference in the diameter of inhibition values when testing the samples and that they had maximum antibacterial activity against Gram-positive *S. aureus* bacteria with a maximum zone of inhibition of 11-17 mm and moderate antibacterial activity against pneumonia.

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