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## Preparation of Nanoparticles in an Eco- friendly Method using Thyme Leaf Extracts

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### Abstract:

Colloidal silver nanoparticles were prepared by single step green synthesis using aqueous extracts of the leaves of thyme as a function of different molar concentration of  $\text{AgNO}_3$  (1,2,3,4 mM). The Field Emission Scanning Electron Microscopy (FESEM), UV-Visible and X-ray diffraction (XRD) were used to characterize the resultant AgNPs. The surface Plasmon resonance was observed at wavelength of 444 nm. The four intensive peaks of XRD pattern indicate the crystalline nature and the face centered cubic structure of the AgNPs. The average crystallite size of the AgNPs ranged from 18 to 22 nm. The FESEM image illustrated the well dispersion of the AgNPs and the spherical shape of the nanoparticles with a particle size distribution between 13 to 50 nm.

**Key words:** Biosynthesis, Friendly method, Nanoparticles (NPs), Silver, Thyme.

### Introduction:

Nanotechnology is a modern branch of research aimed to synthesize or to manipulate the particle structure to be in the ranging of 1 to 100 nm in order to make a pivotal change in the chemical, physical, and biological properties. The novel properties of nanoparticles or nanomaterials such as the morphology, particle size and their distribution spread out their usage rapidly on various potential applications. It is becoming more modernized for tremendous disciplines such as health care, drug delivery, biomedical, cosmetics, environmental applications, chemical industries, catalysts, electronics, optics and other applications (1–7). The ratio of the surface/volume of the metallic nanoparticles is quite big which in turn promoted it to be among the most valuable materials in the antibacterial application because of their microbial resistance towards the antibodies (6). AgNPs are unique nanoparticles with a good chemical stability, unique antibacterial effect and anti-inflammatory activity (8,9). Because of the antiseptic and biocidal effect against microorganisms of the AgNPs, it is suitable to be used for wound dressings, topical creams, antiseptic sprays and fabrics. It operates through disrupting the unicellular membrane of the microorganisms which consequently disturbs their enzymatic activities.

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In addition, recently, synthesis of AgNPs for cancer diagnosis and treatment grabbed the interested of many scientists (10, 11).

However, synthesis of nanoparticles via chemical or physical methods is either expensive or associated with an environmental hazard due to inclusion of some toxic chemicals during synthesizing. The biological inspired synthesis of nanoparticles is an alternative and promised environment friendly process to synthesis the nanoparticles because it does not involve using toxic chemicals during synthesizing. The biological synthesis of nanoparticles is an ecofriendly method that includes using microorganism (12–13), enzyme (13), and plant or plant extract (14). Among all biological methods, the advantages of synthesis nanoparticles using plants is to eliminate the elaborate process of maintaining the cell cultures (14). In addition, it is a scalable process to synthesize the nanoparticles. However, in order to replace the chemical methods, the biological methods need to be developed in term of accelerating the rate of synthesis. On the other hand, due to the hazardous issues, the dedicated research developed eco-friendly synthesis of metal nanoparticles (15). The plant sources viz such as leaves, bark, fruit extracts (16, 17) and bio-organisms (18, 19) are commonly used in that approach. The plant extract method is a cost effective, time effective and efficient green method provides synthesis of crystalline nanoparticles of

wide range of sizes and shapes. Herein, the thyme leaf extract was used for the biosynthesis of silver nanoparticles. The composition of the nanoparticles is compared as a function of the concentration of the metal ions.

## Materials and Methods:

### Plant material

Leaves of thyme were obtained from local markets. The plant leaf extract was typically prepared by mixing 10 g of the leaves powder with a 100 ml of DW (distilled water) in a round flask and refluxed at 50 °C for 45 mins before being cooled down into the room temperature. The Whatman No. 1 filter paper was used to filter the obtained extract which then stored at 4 °C.

### Green synthesis of AgNPs:

Typically, 10 ml of 1 mM AgNO<sub>3</sub> (Shanghai Jiuling Chemical Co., Ltd.) aqueous solution, was added to 1 mL of plant extract and left at room temperature for one hour. The change in color was observed indicating the presence of silver nanoparticles. These procedures were repeated with other molar concentrations of AgNO<sub>3</sub> (2, 3, 4 mM)

### Characterization of AgNPs

The AgNPs were characterized by an UV-vis spectrum (Shimadzu UV-Vis 1800 spectrophotometer. A double beam UV-visible spectrophotometer (PD-303 UV) is used to detect the surface Plasmon resonance property (SPR) of AgNPs at room temperature.

The structural evolution of the green synthesized AgNPs using thymus vulgaris aqueous extract was executed by X-ray diffractometer [Shimadzu XRD-6000, AS (3k.NOPC)] using Cu-K $\alpha$ -radiation of wavelength ( $\lambda = 0.15418$  nm) operating at 40 kV and 30 mA. Debye-Scherrer formula ( $D = k\lambda/\beta\cos\theta$ ) was used to measure the average crystallite size of the AgNPs, where D is the size of crystalline, k is a constant = 0.94,  $\lambda$  is wavelength of X-ray source,  $\beta$  is the full width at half maximum (FWHM) and  $\theta$  is the Bragg angle). The field effect scanning electron microscopy (FESEM) type Jeol JSM-6460 LV microscope was used to study the morphology and microstructure of the AgNPs.

## Results and Discussion:

UV-Vis absorption spectra were used to monitor the surface plasmon in AgNPs spectra. The intensive absorption peak reveals the formation of Ag colloid. Fig. 1 shows the absorption peaks of UV-visible spectra of aqueous media of AgNPs as a function of silver nitrate salt's concentration. In

UV-visible spectra of AgNPs, the absorption peak is observed at 437 nm. It was further confirmed by other characterizations that this peak indicates the formation of spherical shape of AgNPs. The shifting in the wavelength (from 437 to 444 nm) of the AgNPs spectra indicates an increase in the concentration of metal ions. The higher is the metal ion concentration, the higher is the particle size of the AgNPs (20). This result is in agreement with the result reported by Edreese Alsharaeh et al (21), where they utilized lemon juice under microwave irradiation (MWI) and UV light irradiation to synthesize AgNPs. The peak of maximum absorption AgNPs spectra was between 420 to 450 nm. Pandian Bothi Raja et al (22) employed a rapid green method to synthesize silver nanoparticles using verity sources of tannin such as mangrove (MG), chestnut (CN) and quebracho (QB) as a reducing agent. Their results were very similar to what has been found. These result is in agreement with Jae Yong Song et al (20), who reported using five plant leaf extracts for green synthesis of silver nanoparticles. The UV spectrum showed an absorbance band recorded at 430 nm. Our reports are also correlated with (23) in term of using fungus *Fusarium oxysporum* to synthesize of AgNPs. The UV spectra stated that the absorbance band was from 437 to 444 nm. That single SPR peak refers to the spherical shape of the our synthesized AgNPs where the absorbance spectra demonstrated only a single SPR band (Mie's theory). In addition, presence of SPR is a characteristic of formation of the small size and spherical shape of AgNPs .

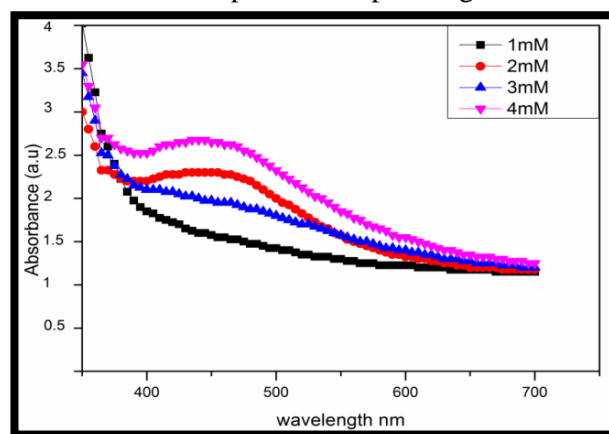


Figure 1. UV-visible spectra of silver Nanoparticles synthesised as a function of molar concentration of AgNO<sub>3</sub>.

XRD patterns of dried AgNPs (Fig. 2) shows the fcc structure of silver nanoparticles which were synthesized using thyme extract as a function of metal ion concentration. Consequently, the peaks at  $2\theta$  of 38.45°, 44.39°, 64.57°, and 77.54° are corresponding to the crystallographic

planes 111, 200, 220 and 311, respectively (21). No extra peaks were detected as belonging to other crystalline phases indicating the pure synthesis of fcc structural Ag nanoparticles with no impurities (24). The peaks of XRD pattern of our synthesized AgNPs agreed with the JCPDS pattern's peaks of the standard gold (file no. 04-0784). The peak at  $38.18^\circ$  plane (111) exhibited stronger intensity than the intensities of the peaks at  $44.39^\circ$ ,  $64.57^\circ$  and

$77.54^\circ$ . The number of peaks and the intensities are affected by the change of the concentration of the metal ion. At the low concentration (1mM and 2mM) the intensity of the four peaks were higher with comparison to the high concentrations (3 mM and 4 mM). The crystalline size was calculated using Scherrer equation which was between 18-22 nm and the average crystallite size was 20 nm.

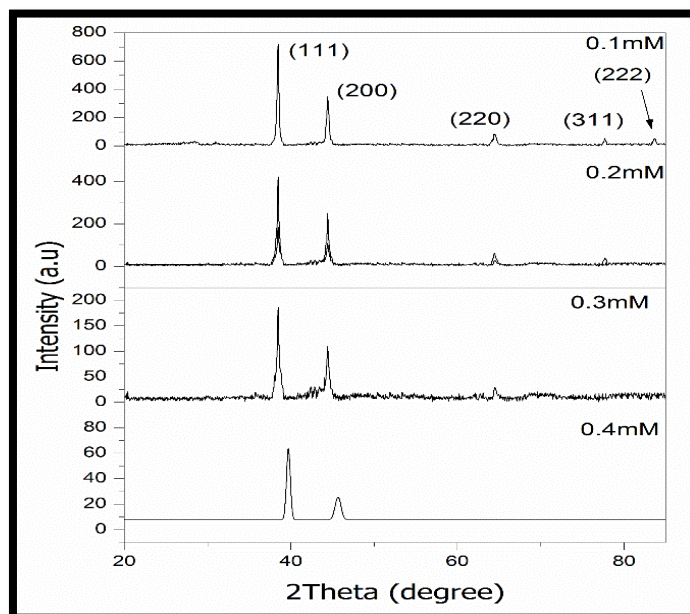


Figure 2. AgNPs XRD pattern as a function of molar concentration of AgNO<sub>3</sub>( 1, 2, 3, 4 mM).

FESEM imaging was performed to investigate the shape and the size of the AgNPs. (Fig. 3) illustrates the FESEM images of the biosynthesized nanoparticle which were deposited on silicon substrates. The majority of the particles were distributed in a nanoscale range of (1-100 nm). In addition, as can be observed from the images, the spherical shape of the particles ,the particle size is

distributing from 13-50 nm (Fig 4), which are suitable for the medical application usage. However, some particles of different shapes are existed which was expected due to the aggregation of Ag nanoparticles during the deposition process (23). In addition, it has been observed that the temperature of synthesizing affected the shape of the nanoparticles

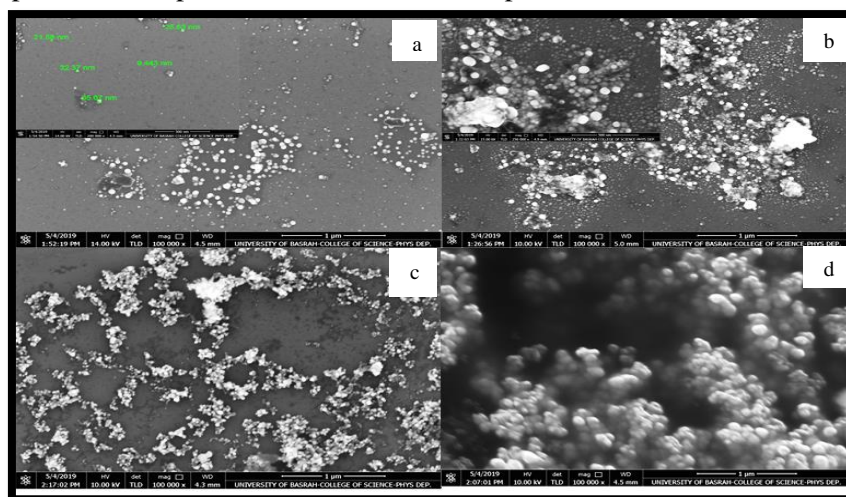
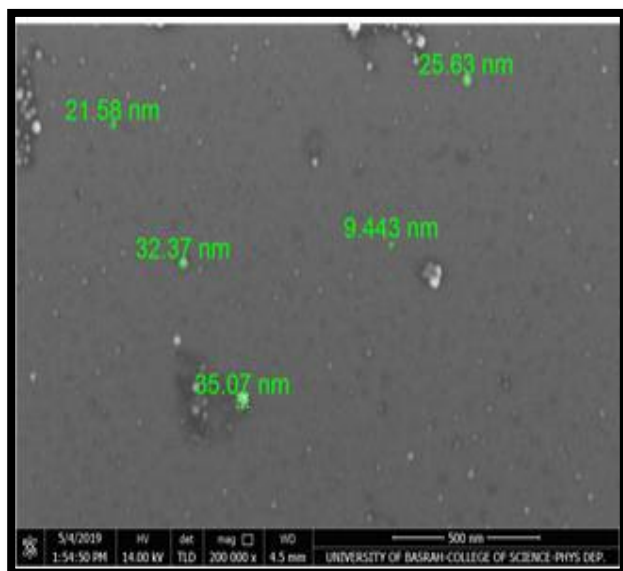


Figure 3. FESEM image of the Ag-NPs forms using as a function of molar concentration of AgNO<sub>3</sub>: a (1mM),b(2mM),c(3mM), and d(4mM)



**Figure 4. FESEM image of the Ag-NPs forms using as a function of molar concentration of AgNO<sub>3</sub> (1mM)**

### Conclusion:

Thyme leaf extracts have been adopted as a bio-reduction agent of silver nitrate solution for AgNPs green synthesis. The particle size of AgNPs is found to be correlated with the metal ion concentration. Where concentration is low, the particle size of the AgNPs becomes smaller. The result of UV-vis spectroscopy states that the surface Plasmon resonance of AgNPs is located at the wavelength of from 437 to 444 nm, while FESEM analysis reveals the well dispersion of AgNPs with a spherical shape and the average particle size is calculated using Scherrer equation and distributed between 18- 22nm.

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### Authors' declaration:

- Conflicts of Interest: None.
- We hereby confirm that all the Figures and Tables in the manuscript are mine ours. Besides, the Figures and images, which are not mine ours, have been given the permission for re-publication attached with the manuscript.
- Ethical Clearance: The project was approved by the local ethical committee in University of Anbar.

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## تحضير الجسيمات النانوية بطريقة صديقة للبيئة باستخدام مستخلصات أوراق الزعتر

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### الخلاصة

تم تحضير الجسيمات الفضة النانوية الغروية بواسطة التخليق الأخضر ذو الخطوة الواحدة باستخدام المستخلصات المائية من أوراق الزعتر. كعامل الحد وتغطية. أجريت خصائص جسيمات الفضة النانوية الناتجة باستخدام حيود الأشعة السينية (XRD) والأشعة فوق البنفسجية والميكروسكوبية المسح الضوئي للانبعاثات الميدانية (FESEM)، وكان صدق Plasmon السطحي لـ AgNPs عند 440 نانومتر. تشير قمم نموذج XRD إلى الطبيعة البلورية والبنية المكعبة المتمركزة على الوجه لـ AgNPs. يتراوح متوسط الحجم البلوري للجسيمات بين 13 إلى 22 نانومتر، وتوضح صورة FESEM التشتت الجيد للبقايا النانوية والشكل الكروي للجسيمات النانوية مع توزيع حجم

الكلمات المفتاحية: تخليق حيوي، طريقة صديقة للبيئة، الجسيمات النانوية، الفضة، زعتر.