A novel route for large-scale synthesis silver nanoparticles using *Uncaria tomentosa* leaves extract: Antibacterial activity

Nidá M. Salem[1], Saba H. Alkharabsheh[2], Akl M. Awwad[1,3]*

[1]*College of Agriculture, the University of Jordan, Amman, Jordan*
[2]*College of Engineering, Al-Balqaa Applied University, Salt, Jordan*
[3]*Royal Scientific Society, Amman, Jordan*

*Corresponding author

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Abstract

The present study deals with rapid and large-scale synthesis of silver nanoparticles (AgNPs) by *U. tomentosa* leaves aqueous extract at ambient temperature. Crystal growth of silver nanoparticles by this green route was formed within 5min at room temperature. Synthesized AgNPs were characterized by Ultra-violet visible spectrophotometer (UV-vis), Fourier transform infrared spectroscopy (FT-IR), Transmission electron microscopy (TEM) and X-ray diffraction (XRD). Silver nanoparticles synthesized by green route showed better antibacterial activity. Copyright © 2018 VBRI Press.

Keywords: Green large-scale synthesis, dilver nanoparticles, *U. tomentosa* leaves

Introduction

Silver Nanoparticles (AgNPs) have given much attention in research work due to its unique physical, chemical and biological properties. Eco-friendly and non-toxic plants extracts were used by different researchers for synthesis silver nanoparticles such as Belgian endive; *Cichorium intybus* L. var. *sativus*[1], aqueous extract of *Rubus ellipticus* leaves [2], *Cassia auriculata* flower extract [3], *Citrullus lanatus* fruit rind extract[4], *Eucommia ulmoides* bark [5], *Lycium barbarum* fruit extract [6], *Carcia papaya* peel extract [7], *Sesbania grandiflora* leaf extract[8], *Solanum muricatum* leaf extract [9], *Mussaenda erythrophylla* leaf extract [10], *Tamarix gallica* leaf extract [11],), *Ficus carica* leaf extract [12], *Tephrosia purpurea* leaf extract [13], leaf extract of *Grewia flaviscences*[14], *Atrocarpus altilis*leaf extract [15], *Ixora coccinea* leaves extract [16], *Arbutus Unedo* leaf extract [17], *Ocimum sanctum* (Tulsi) leaf extract [18], leaves of *Stevia rebaudiana* [19], *Ficus benghalensis* leaf extract [20], leaves and green berry extract of *Solanum nigrum* L.; *Solanaceae: Solanales* [21],*Olea europaea* leaves extract [22], carob leaves extract [23], loquat leaf extract [24] and leafy green extract of Belgian endive (*Cichorium intybus*L. var. *sativus*) [25]. *U. tomentosa* is deriving its name from hook-like thorns that resemble the claws of a cat. The leaves are elliptic with a smooth edge, and grow in opposing pairs. Cat's claw is planted at the campus of Royal Scientific Society, Amman, Jordan.

Herein, a novel green route is designed for synthesis silver nanoparticles using *U. tomentosa*leaves aqueous extract at room temperature at short time.

Experimental

Silver nitrate ($AgNO_3$, > 99.8%) was purchased from Riedel-de Haën, Germany and used without any further purification. In all experimental work, de-ionized water was used.

*U. tomentosa*leaves were collected from different places at the campus of Royal Scientific Society, Amman, Jordan. Leaves were washed with distilled water to remove dust and then left to dry in our laboratory for one week. Dried leaves were crushed and ground to fine powder.

Aqueous extract of *U. tomentosa*leaves was prepared by mixing 10g powder with 500ml de-ionized water and boiled for 10min. A yellow aqueous extract, **Fig. 1** obtained was separated by filtration on Whatman filter paper No. 1. An aqueous extract of *U. tomentosa* leaves was kept in a stopper bottle at room temperature for our experimental work.

Fig. 1. Leaves of *U. tomentosa*and the vaqueous extract

In a typical synthesis of AgNPS, 1.2g silver nitrate ($AgNO_3$) was dissolved in 100ml de-ionized water in 250ml conical flask with magnetic stirring at ambient temperature. Afterwards, *U. tomentosa* leaves extract was added drop wise to silver nitrate solution, the color changed from colorless to yellow and after 2 min to deep brown and after 5min to grey-black suspended particles. UV - vis absorption spectroscopy (UV - vis) analysis showed a strongSurface plasmon resonance (SPR) band at 432nm, indicating the formation of silver nanoparticles (AgNPs). This typical synthesis was repeated several times with changing the concentration of $AgNO_3$ and the volume of extract to develop large-scale synthesis.

Silver nanoparticles and *U. tomentosa* leaves extract were characterized by different techniques: UV-vis spectroscopy (Shimadzu UV-1601), X-ray diffraction (XRD-6000), Fourier transform infrared (FT-IR, IR-Prestige-21 Shimaduz), energy-dispersive X-ray spectroscopy (EDS) and Transmission electron microscopy (TEM, Hitachi 7600 machine).

Results and discussion

Silver nanoparticles synthesized by *U. tomentosa* leaves extract, which acts as reducing and stabilizing agent was monitored by the UV-vis spectrophotometry, **Fig. 2.** An absorbance spectrum was observed in each spectrum at 432nm which is characteristic of silver nanoparticles. No other peaks were observed in the spectrum, indicating the high purity of the synthesized silver nanoparticles by this fast and green method.

XRD pattern of synthesized AgNPs with the standard (JCPDS No. 04-0783) confirmed the form of nanocrystals, as evidenced by the peaks at 2θ values of 38.18°, 43.96°, 64.18°, and 77.22° corresponding to (111), (200), (220), and (311) Bragg reflections, **Fig. 2**. It was found that the average size from XRD data using Debye-Scherrer equation (Awwad et al., 2015) was about 8-12 nm. The peaks denoted by (*) indicated the presence of crystalline bio constituents of *U. tomentosa* leaves extract.

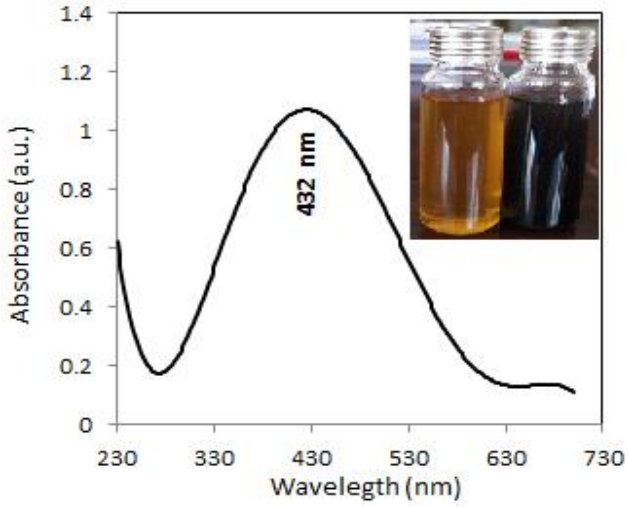

Fig. 2. UV absorption spectra of synthesized silver nanoparticles.

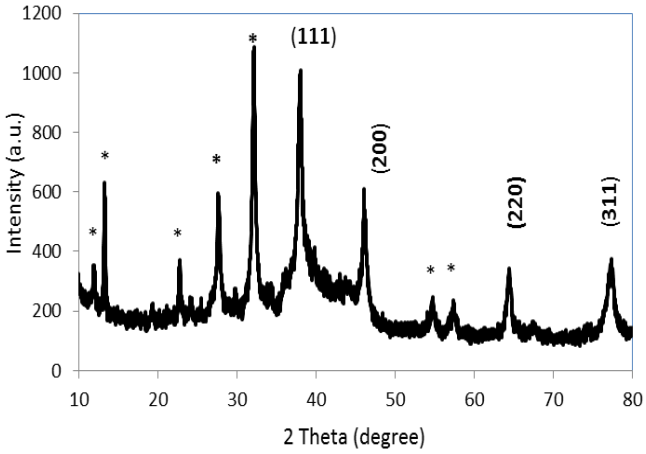

Fig. 2. XRD of synthesized silver nanoparticles by *U.tomentosa*.

FT-IR spectra of *U. tomentosa* leaves extract is shown in **Fig. 3.** The *U. tomentosa* leaves extract displays a number of absorption peaks, reflecting its complex nature. Peak at 3383 cm^{-1} results are due to the stretching of -OH groups of alcohol and phenol compounds. The strong absorption peaks at 2924 cm^{-1} and 2850 cm^{-1} could be assigned to –CH stretching vibrations of $–CH_3$ and $–CH_2$ functional groups in aliphatic and aromatic compounds. The shoulder peak at 1739 cm^{-1} and strong peak at 1612 cm^{-1} indicated the fingerprint region of C=O and N–H groups in amide (I) and amide (II) of protein. The intense band at 1442 cm^{-1} and 1230 cm^{-1} can be assigned to the C-N stretching vibrations of aliphatic amines. The peak at 1064cm^{-1} may be attributed to C-O-C stretching mode of aromatic ether linkage group. The peak at 621cm^{-1} indicates the –OH bending of the phenolic groups. FTIR study indicated that the free hydroxyl (-OH), carboxyl (-C=O), C-N and amine (N- H) groups present in the structure of *U. tomentosa* leaves extract are mainly involved directly in reduction of Ag^+ to Ag^o nanoparticles, **Fig.4.**

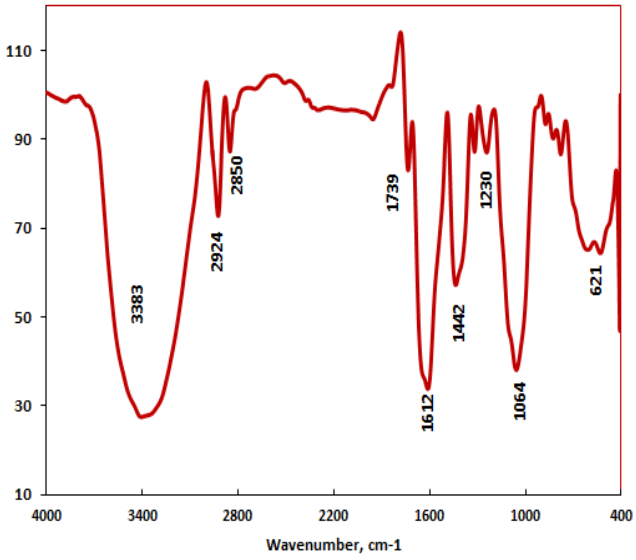

Fig. 3. FT-IR of *U. tomensota* leaves extract.

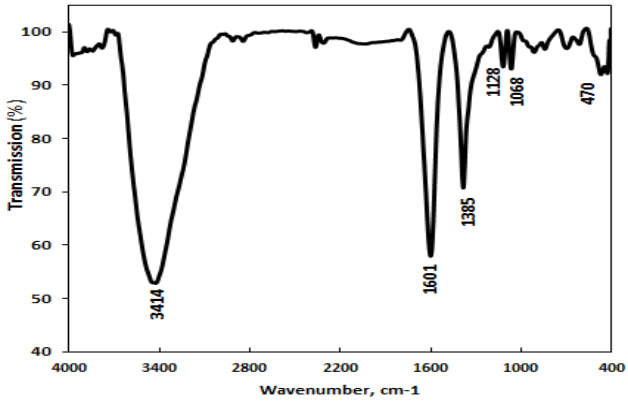

Fig. 4. FT-IR of synthesized silver nanoparticles.

Energy dispersive X-ray analysis (EDS) corroborated the presence of elemental Ag in synthesized AgNPs using *U. tomentosa*leaves extract, **Fig. 5.**

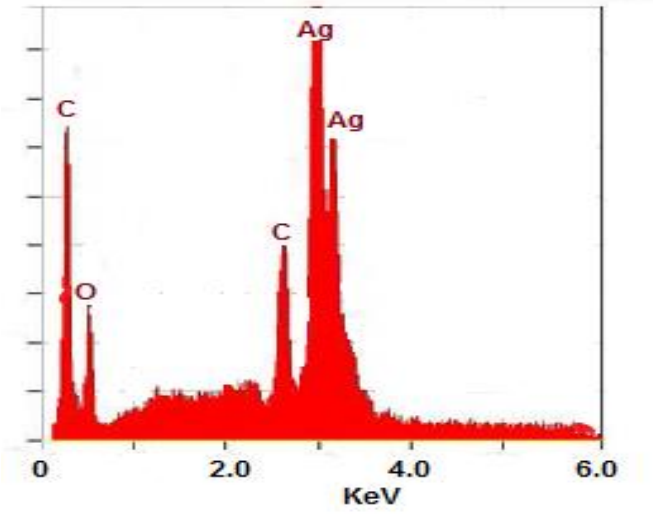

Fig.5. Energy-dispersive X-ray (EDS) spectrum of synthesized AgNPs

Transmission electron microscopy (TEM) analysis investigated the morphology and size distribution of synthesized silver nanoparticles. **Fig. 6** showed that the synthesized silver nanoparticles using *U. tomentosa* leaves extract at room temperature are mostly spherical. The particles size measured from the TEM images is from 5-20nm. However, relatively bigger particles are below 20nm. The average particles size of AgNPs is about 12nm. Stabilizing of silver nanoparticles was also observed under TEM micrograph. This stabilization might be due to of presence of bio-organic compounds present in the aqueous extract of *U. tomentosa* leaves.

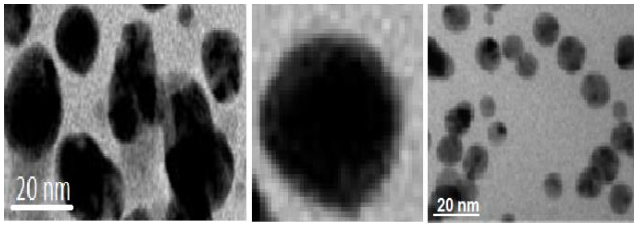

Fig. 6. Typical TEM image of the synthesized silver nanoparticles

Antibacterial properties of synthesized AgNPs tested against pathogenic bacteria, *E. coli and S.aureus*. It was observed that silver nanoparticles have antibacterial activities at concentration of 20µg/disc. Chloromphenical was used as a control antimicrobial agent. It was observed that an increase in AgNPS concentration increases the MZI and found that the MZI value of Gram negative bacterium *E. coli* showed lesser activity values by Gram positive bacterium *S. aureus*. Present study observed results reveal that bio-reduced AgNPs showed a significant antibacterial property compared with positive drug control, it could be explained by large surface area of AgNPs, which gives better contact with microorganisms thus alter the microbial metabolism. The nanoparticles attached to the cell membrane and penetrated inside the microorganisms.

U. tomentosa leaves extract can reduce silver ions to silver nanoparticles and also act as stabilizing agent for the formed silver nanoparticles and prevent them from aggregation. *U. tomentosa* leaves extract contains bioactive compounds having functional groups such as - C=O, -OH, -NH and C-N. The mechanism of biosynthesis silver nanoparticles proposed, **Fig. 7** on the presence of the bioactive constituents such as polyphenols, flavonoids, alkaloids, and amino acids in *U. tomentosa* leaves extract.

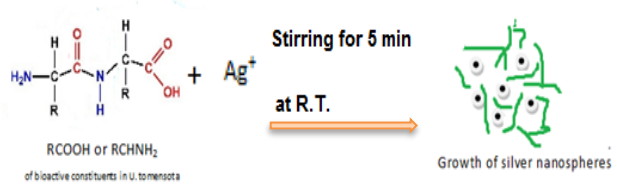

Fig. 7. Schematic illustration of the silver nanoparticles growth.

Conclusion

Facile and green method for synthesis silver nanoparticles using *U. tomentosa*leaves aqueous extract at room temperature (27ºC). *U. tomentosa*leaves aqueous extract act in the process of synthesis as reducing and stabilizing agent. Further, the above method, the concentration and molar ratio of U.t/Ag are critical for the formation and large-scale of silver nanoparticles synthesis at room temperature. AgNPs showed an effective antibacterial activity. Application of AgNPs based on these results may lead us to study the effect of these nanoparticles in field of agriculture (Banana plant growth)

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Author's contributions

NMS, SHA, AMA performed the expeirments, data analysis and wrote the paper

Supporting information

Supporting informations are available from VBRI Press.

References

1. Gallucci, M.N.; Fraire, J.C.; Maillard, A.P.V.F.;Masillard, P.L., Martinez, I.M.A.; Miner, E.V.P.; Coronado, E.A.; Dalmasso, P.R., Mater. Lett., **2017**, **197**, 98.
DOI: 10.1016/j.matlet.2017.03.141.

2. Al Qahtani, F.S.; Al Shebly, M.M.; Govindarajan, M.; Senthilmurugan, S.; Vijayan, P.; Benelli, G.G., J. Asia-Pacific Entomology, **2017**, **20**, 157. **DOI**: 10.1016/j.aspen.2016.12.004.
3. Muthu, K.; Priya, S., Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy, **2017**, 179, 66. **DOI**: 10.1016/j.saa.2017.02.024.
4. Ndikau, N.; Noah, N.M.; Andala, D.M.; Masika, E., 2017. Inter. J. Anal. Chem., **2017**, Article ID 8108504:9. **DOI**: 10.1155/2017/8108504.
5. Lǔ, S.; Wu, Y.; Liu, H.; Mater. Lett., **2017**, 196, 217. **DOI:** 10.1016/j.matlet.2017.03.068.
6. Dong, C.; Gao, C.; Zhang, X.; Zhan, Y.; Wang, X.; Yang, X.; K. Zhou, K.; Xiao, X.; Yuan, B.; Optik, **2017**, 130, 162-170 **DOI:** 10.1016/j.ijleo.2016.11.010.
7. Kokila, T.; Ramesh, P.S.; Geetha, D.; 2016. Ecotoxicology and Environ Safety, **2016**, 134, 467. **DOI:** 10.1016/j.ecoenv.2016.03.021.
8. Ajitha, B.; Reddy, Y.A.K.; Rajesh, K.M.; Reddy, P.S.;**2016**. Materials Today: Proceedings, 2016, 3, 1977. **DOI:** 10.1016/j.matpr.2016.04.099.
9. Gorbe, M., Bhat, R., Aznar, E.; Sancenón, F.; Marcos, M.D.; Herraiz, F.J.; Prohens, J.; Venkataraman, A.; Martínez-Máñez, R.; Materials, **2016**, 9, 325. **DOI:** 10.3390/ma9050325.
10. Varadavenkatesan, T.; Selvaraj, R. Vinayagam, R.;J. Molecular Liquids, **2016**, 221, 1063-1070. **DOI:** 10.1016/j.molliq.2016.06.064.
11. López-Miranda, J.L.; Vázquez, M.; Fletes, N.; Esparza, R.; Rosas, G.; Mater. Lett., **2016**, 176, 285. **DOI:** 10.1016/j.matlet.2016.04.126.
12. Ulug, B.; Turkdemir, M.H.; Cicek, A.; Mete, A., Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy, **2015**. 135, 153. **DOI:** 10.1016/j.saa.2014.06.142.
13. Ajitha, B., Reddy, Y.A.K.; Reddy, P.S.; Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy, 2014, 121, 164-172 **DOI:** 10.1016/j.saa.2013.10.077.
14. Sana, S.S.; Badineni, V.R.;Arla, S.K., Boya, V.K.N.; Mater. Lett., **2015**, 145 347. **DOI:**10.1016/j.matlet.2015.01.096.
15. Ravichandran, V.;Vasanthi, S.; Shah, S.A.A.; Harish, R.; Mater. Lett., **2016**, 180, 264. **DOI:** 10.1016/j.matlet.2016.05.172.
16. Karuppiah, M.; Rajmohan, R.;Mater. Lett., **2013**, 97, 141. **DOI:** 10.1016/j.matlet.2013.01.087.
17. Kouvaris, P.; Delimitis, A.; Zaspalis, V.;Papadopoulos, D., Tsipas, S.A., Mater. Lett., **2012**, 76, 18. **DOI:** 10.1016/j.matlet.2012.02.025.
18. Singhal, G.; Bhavesh, R.; Kasariya, K.; Sharma, A.R., Singh, R.P.; J. Nanopart. Res., **2011**, 13, 2981. **DOI:** 10.1007/s11051-010-0193-y.
19. Yilmaz, M.;Turkdemir, H.; Kilic,M.A., Bayram, E.; Cicek, A.; Mete, A., Ulug, B.; Mater. Chem. Phys., **2011**, 130, 1195. **DOI:** 10.1016/j.matchemphys.2011.08.068.
20. Saxena, A.; Tripathi, R.M.; Zafar, F.;P. Singh, P.; Mater. Lett., **2012**, 67, 91. **DOI:** 10.1016/j.matlet.2011.09.038.
21. Rawani, A.; Ghosh, R.; Chandraa, G.; Acta Tropica, **2013**, 128, 613-622. **DOI:**10.1016/j.actatropica.2013.09.007.
22. Awwad, A.M.; Salem, N.M.; Abdeen, A.O.; Nanosci. and Nanotechnol. J., **2012**, 2, 164. **DOI:** 10.5923/j.nn.20120206.03.
23. Awwad, A.M.; N.M.Salem, N.M.;A.O. Abdeen, A.O.; Inter. J. of Indust. Chem., **2013**, 4: 29. **DOI:** www.industchem.com/content/4/1/29.
24. Awwad, A.M.;Salem, N.M.;Abdeen, A.O.; Adv. Mater. Lett., **2013**, 4, 338 **DOI:** 10.5185/amlett.2012.11453.
25. M.N.; Gallucci, J.C. Fraire, A.P.V.F. Maillard, P.L.; I.M.A. Martínez, E.V.P. Miner, E.A. Coronado, P. R. Dalmasso, P.R.; Mater. Lett., **2017**, 197, 98. **DOI:** 10.1016/j.matlet.2017.03.141.

