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SHORT COMMUNICATION

Biosynthesis of Cu and CuO nanoparticles using aqueous leaves extract of *Sambucusnigra* L.

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Abstract

In this research we are synthesized CuO nanoparticles (NPs) using water extract of. Also the total phenolic content of *Sambucusnigra* L. leaves water extract was measured by the Folin-Ciocalteu method. For confirmation the structure of synthesized bio-CuO-NPs we are employed X-ray Diffraction (XRD), Ultraviolet-visible (UV–vis), Fourier Transform Infrared (FTIR) spectroscopies, Scanning Electron Microscopy (SEM), and Transmission electron microscopy (TEM) methods that indicated that synthesized bio-CuO NPs have crystalline face-centered cubic (*fcc*) CuO phase with spherical morphology and with size about 14 nm. It should be mentioned that two different temperatures (100 and 400 °C) caused to Cu and CuO NPs respectively.

Keywords: *Aqueous extract; Bio-CuO nanoparticles; Sambucusnigra L.; Total phenolic contents; Transmission electron microscopy (TEM).*

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INTRODUCTION

Owing to their size and morphology dependent noble physicochemical properties, nanomaterials have received tremendous attention by researchers over the last 4 decades. In particular, transition metal oxide nanomaterials have been of due interest as a result of their optoelectronic, magnetic, catalytic, and medicinal properties [1]. Among metal oxide, copper oxide has gained more attention in the last decade, due to its distinctive properties such as high temperature superconductivity, spin dynamics and electron correlation effects, which those increase widely its applications as heterogeneous catalysts, antioxidants, drug delivery agents, and imaging agents in field of biomedicine [2, 3]. Furthermore, copper oxide is inexpensive antimicrobial agent when compared to inorganic bactericidal agents such as silver and gold, and has longer shelf life as compared to other organic antimicrobial agents [2]. All the mentioned properties of copper oxide drastically increase when it fabricates in nano-size scale. In fact, copper oxide nanoparticles (CuO NPs) due to their extremely unusual crystal morphologies and high surface area to volume ratio, have unique physical, chemical and biological properties [2, 4]. Development of a non-toxic, clean, reliable, cost effective, eco-friendly and biocompatible processes to NPs fabrication is one of the great interests of researchers in the last decade to replace them with common physical and chemical synthesis methods, which those are high energy consuming with intensive capital cost and utilize toxic chemicals and non-polar solvents in their processes [5]. Green synthesis of NPs with plants and their derivatives extract, have gained more attentions during last year's, due to the presence of the numerous natural reductants (i.e., polyphenols, flavonoids, tannins, ascorbic acids and sugars) and stabilizers (i.e., proteins, carbohydrates, gums and pectic substances) [6-8]. Numerous researches have been done on green synthesis of CuO NPs with plant extract such as

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tea leaf, coffee powder and Eichhornia crassipes extracts [9]. CuO is the transition metal oxide and an intrinsically p-type semiconductor with a band gap value ranging between 1.2 eV in the bulk to up to 3.57 eV in the nanoscale size regime [10]. As a result of its narrow band gap and chemical stability, it has been used in a multitude of application including in magnetic storage media [11], in high efficient thermal conducting materials [11, 12], as an electrode in dye sensitized solar cells [13], in lithium ion batteries [14] and solid oxide fuel cells [1], in chemical and gas sensors [15–17], as catalyst for degradation of organic contaminants in waste water [18, 19], in medicine as antimicrobial agent [20].

CuO NPs have been synthesized by physical, chemical, and biochemical methods. In the physical method, it is synthesized using mechanical ball-milling [21] and laser ablation [22]. It has also been synthesized chemically using such methods as solvothermal [23, 24], micro emulsion [15], microwave irradiation [25], sonochemical [26], precipitation [27], sol-gel [28], and electrochemical methods [29]. The biosynthesis method has been of recent interest for its economical, eco-friendly approach, and less work upinvolved in the preparation of the desired nanomaterials [30]. As such, CuO NPs has been synthesized from bacteria [31] and variousplant extracts including Aloe vera [32], *Thymus vulgaris* [33], Olive leaves [34], *Carica papaya* [18], banana peels [35], and brown algae [36-38].

Sambucusnigra L., or European elderberry, a member of the Caprifoliaceae or honeysuckle family is a well-known plant, which grows in the shade and moist region in Europe, Asia, North Africa, and North America. It is a deciduous shrub with 5–9 leaflets, pinnate leaves, which is maximum 6 m high. This plant has small, white or cream clusters of flowers in late spring, and early summer. It bears fruits with dark purple color and about 6 mm diameter each, ripen in late summer. Historically, the leaves, bark, flowers, and fruits have all been used medicinally, and most of the medical studies have been done on the therapeutic uses and properties of the elderberry. Elderberries are highly regarded due to biologically active compounds. Due to the existence of antioxidants (anthocyanins, phenolics), proteins, sugars, organic acids, vitamin A, vitamin C, bioactive compounds in different parts of the elderberry, it is very important for the pharmaceutical, biotechnology, food technology and medicine [39-41].

In this work, CuO NPs is synthesized using *Sambucusnigra* L. leaves extract without adding any baseor stabilizing agent. The synthesized CuO NPs were subjected to different characterizations.

EXPERIMENTAL

In this research, all chemicals are purchased from Fluka (Buchs, Switzerland) and employed with any purification. For measuring infrared spectroscopy and melting point, a Shimadzu IR-460 spectrometer was utilized. The scanning electron microscopy (SEM) employing a Holland Philips XL30 microscope was used for determination of CuO nanoparticles morphology. X-ray diffraction (XRD) analysis at room temperature using a Holland Philips Xpert X-ray powder diffractometer, with CuK_{α} radiation (λ=0.15406 nm), with 2θ ranging from 20 to 80° was employed for characterization of crystalline structure of Cu and CuO nanocomposites. Scherrer's formula; $D= 0.9\lambda/\beta \cos\theta$ was employed for calculating the average crystallite size where D is the diameter of the nanoparticles, λ (CuKα) =1.5406 Å and β is the full-width at half-maximum of the diffraction lines. The reagents used in this study, such as gallic acid, Folin–Ciocalteu's phenol, $CuCl_2.2H_2O$ (99%purity), and solvents were purchased from Sigma and Merck Chemical Co.

Biosynthesis of CuO NPs

The *Sambucusnigra* L. is collected from Sari (Iran) in May 2016 and its leaves was separated, washed and dried. The dried leaves of *Sambucusnigra* L. were crushed into finest powder. The powder (15g) was mixed with distilled water (150 ml) for 2 hours at 100°C using soxhlet apparatus. The extract was then filtered through Whatman No. 1 filter paper. For the synthesize of CuO nanoparticles, 1.023 g of $CuCl_2.2H_2O$ was poured in 40 mL of *Sambucusnigra* L. leaves extract in Erlenmeyer and the mixture was heated in at 85-90 °C for 3 h. After 10 min the color of the solution was converted from yellow to dark brown during the heating process. In this step two methods were done. The resulting solution was centrifuged at 4000 rpm for about 20 minutes of removing the useless organic matters and was filtered. For further investigation, the resulting solution was studied in two methods. In the first method, the obtained sample was dried at 100°C for 2 h and in another method it was heated at 400 °C for 2 h.

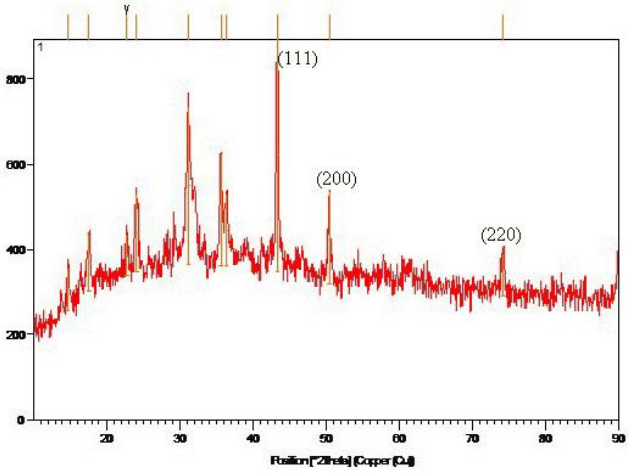

Fig 1. XRD pattern of bio-Cu NPs.

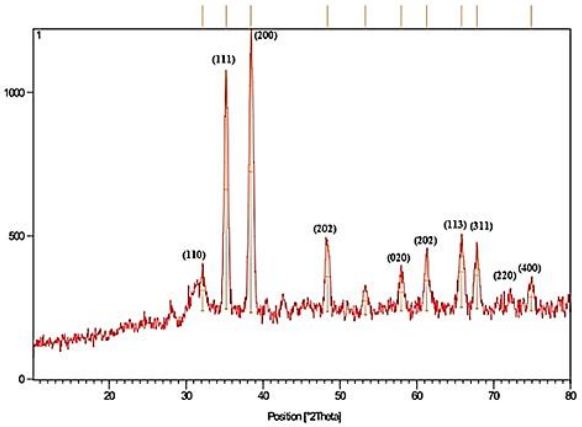

Fig 2. XRD pattern of bio-CuO NPs.

Determination of total phenolic contents [42]

The ethanolic leaf extract content of *Sambucusnigra* L. were prepared by the mixing of 10 g *Sambucusnigra* L. powders with 100 mL of 95% ethanol in a flask for 24 h. Then, the extract was filtrated by filtered paper Whatman No.1 to eliminate the residue. Total phenolic content (TPC) was determined using the Folin-Ciocalteu colorimetric method. First, extract diluted to 100 mL by ethanol, and then 1 mL of this solution mixed with 20 mL ethanol. In the next step, to a 0.5 mL of diluted extract, 3 mL of the Folin-Ciocalteu reagent (Folin-Ciocalteu reagent was prepared by diluting it with distilled water at the ratio of 1 : 10) was added and stirred for 5 min. Then 2.5 mL of 20% (w/v) Na_2CO_3 solution were added and the mixture was allowed to stand for 30 min at room temperature. The absorbance of solution was recorded at 761 nm against a blank. The blank consisted of all reagents and solvents, except the extract (instead of the extract was added distilled water). The total phenolic content was assigned from the standard calibration curve and expressed as gallic acid equivalents per dry mass of *Sambucusnigra* L. sample (mg GAE/g dried sample) [42-44].

RESULTS AND DISCUSSION

The phase identification was carried out with the XRD data. According to the XRD pattern, in the first method, the drying of the sample at 100 °C leads to the synthesis of Cu NPs as the dominant phase (Fig. 1). From calcinations of sample at 400 °C, CuO nanoparticles are prepared (Fig. 2). The

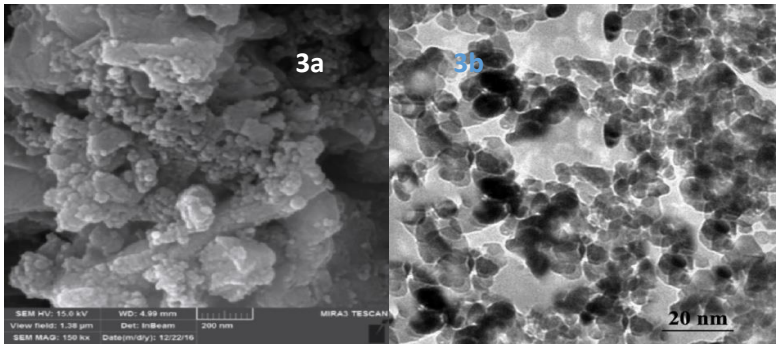

Fig 3. SEM (3a) and TEM (3b) images of biosynthesized CuO NPs.

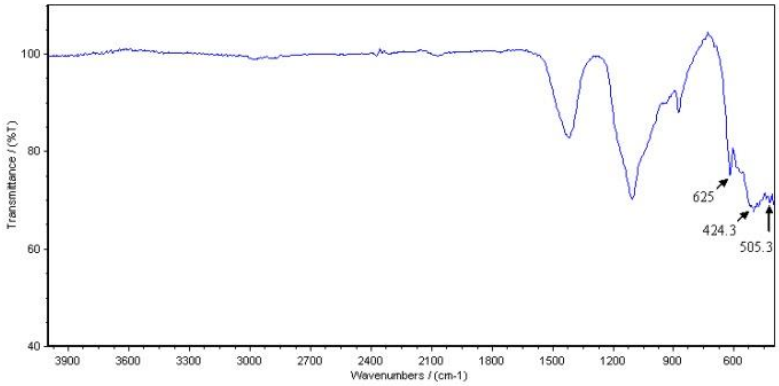

Fig 4. FT-IR spectra of biosynthesized CuO NPs.

XRD pattern of the CuO NPs is consistent with face-centered cubic (*fcc*) structure (Fig. 2). Lack of impurity, no detection secondary phase and narrow diffraction peaks in XRD demonstrates that a single phase of CuO is produced by this method and product has good crystalline structure. The XRD pattern corresponded to face-centered cubic (*fcc*) CuO NPs are found in the lattice planes (h, l, k) of (110), (111), (200), (202), (020), (202), (113), (311), (220) and (400). Based on the Scherer's equation, the average crystallite size of the CuO NPs is 14.00 nm. Though the exact mechanism of metal oxides synthesis using plant extracts without alkali is not yet fully understood, one proposed mechanism of synthesized CuO reported [35] involves first the reduction of Cu^{2+} into elemental Cu andthen reaction of Cu with air/O_2 to give CuO. Hence, the calcination step is important for obtaining CuO phase and accordingly the following mechanism is proposed [37, 45]:

$$Cu^{2+} + Extract \rightarrow Cu—Extract\ (complex) \quad (1)$$

$$Cu{-}Extract\ (complex) \xrightarrow{400^{\circ}C} CuO \quad (2)$$

These results are represented that the *Sambucusnigra* L. leaves extract act as reducing agent and the Cu^{2+} ion is reduced to Cu^{0} and calcination at 400 °C, convert Cu NPs to CuO NPs according Eq. (1) and (2).

Scanning electron microscopy (SEM) was employed to predict the morphology of the CuO nanoparticles. The SEM images of synthesized CuO NPs are shown in Fig. 3a. It is observed that most of the nanoparticles are spherical with agglomeration. The results were confirmed by the TEM analysis as shown in Fig. 3b.

Fig. 4 shows the FTIR spectra of CuO NPs biosynthesized by leaves extract of *Sambucusnigra L.* The stretching bands of the M-O NPs were found at around 600-1000 cm^{-1}. The stretching bands at 505.3, 424.3 and 625 cm^{-1} assigned to the stretching vibration of Cu−O bond in monoclinic CuO [46-50].

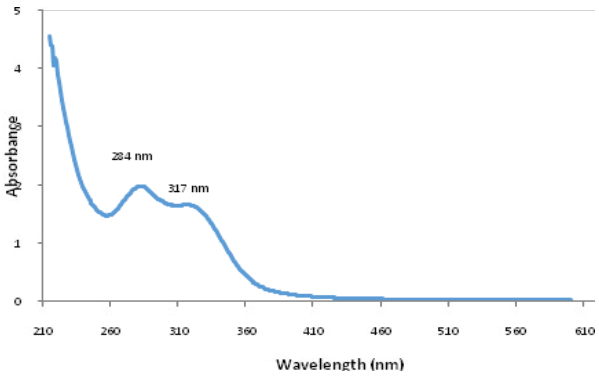

Fig 5. The UV–vis spectra aqueous leaf extract of *Sambucusnigra* L.

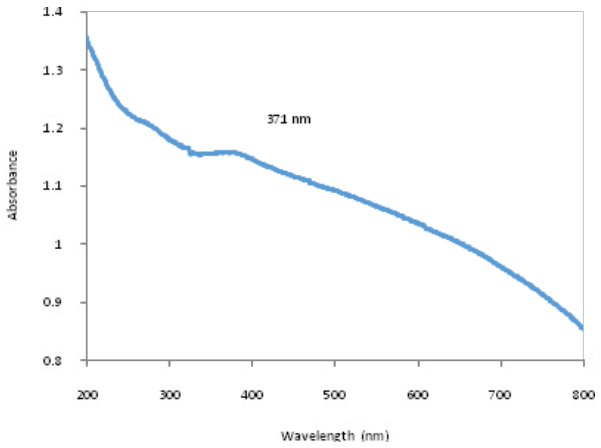

Fig 6. The UV–vis spectra of bio-CuO NPs.

The UVvisible spectrum of aqueous extract of *Sambucusnigra* L. leaves has been shown in Fig. 5. This spectrum demonstrates specified signals of phenolic within the plants as the bonds at λ_{max} 317 nm (bond I) due to the transition localized within the ring of cinnamoyl system; whereas the one around 284 nm (bond II) is for absorbance of ring related to the benzoyl system [40, 51]. They are related to the π→π* transitions and these absorbent bonds show the presence of polyphenolic as antioxidant source for green synthesis of nanoparticles.

The UV–vis spectrum of the CuO NPs showed maximum peak at 371 nm, Fig. 6. The maximum absorbance peak of CuO NPs depends on factors such as temperature, type of extract and precursor, and the synthesis method employed but a value as small asabout 219 nm to about 500 nm is reported in the literature [26, 52].

The band gap (E_g) value can be estimated to be 3.34 eV, on the basis of the corresponding absorptionedges according to Eq. (3) [53, 54] and Fig. 6:

$$E_g = 1240\ \lambda^{-1} \quad (3)$$

This blue shift as compared to the direct bulk band gap of 1.2 eV [37] is due to the quantum confinement effect associated with the reduced crystallite size [55].

Total phenolic content

Ethanolic leaf extract of *Sambucusnigra* L. had a good reducing power. The total phenolic content of the ethanolic leaf extract of *Sambucusnigra L.* was determined using a linear gallic acid standard curve (y = 0.004 x; R^2= 0.992) and expressed as mg of gallic acid equivalents per 100 gram of dry mass of sample (mg GAE/g DM). The TPC of ethanolic extract of *Sambucusnigra* L. was 38.5 mg GAE/ g DM.

CONCLUSION

In this study, a simple, ecofriendly and economic method has been investigated for the preparation of face-centered cubic (*fcc*) CuO NPs *via* green method using *Sambucusnigra* L. leaves extract. The morphology and size of synthesized bio CuO NPs were determined by XRD, SEM, TEM and UV spectra. Also, the TPC of ethanolic extract of Sambucusnigra *L*. was 38.5 mg GAE/g DM.

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CONFLICTS OF INTEREST

The authors do not have any personal or financial conflicts of interest.

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