Advanced Powder Technology xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Advanced Powder Technology

journal homepage: www.elsevier.com/locate/apt

Rapid Communication

Bio-mechanochemical synthesis of silver nanoparticles with antibacterial activity

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ARTICLE INFO

Article history:
Received 20 July 2017
Received in revised form 14 September 2017
Accepted 26 September 2017
Available online xxxx

Keywords:
Mechanochemistry
Ag nanoparticles
Antibacterial activity
Green synthesis

ABSTRACT

By using a bio-mechanochemical approach combining mechanochemistry (ball milling) and green synthesis for the first time, silver nanoparticles (Ag NPs) with antibacterial activity were successfully synthesized. Concretely, eggshell membrane (ESM) or *Origanum vulgare* L. plant (ORE) and silver nitrate were used as environmentally friendly reducing agent and Ag precursor, respectively. The whole synthesis took 30 min in the former and 45 min in the latter case. The photon cross-correlation measurements have shown finer character of the product in the case of milling with *Origanum*. UV–VIS measurements have shown the formation of spherical NPs in both samples. TEM study has revealed that both samples are composites of nanosized silver nanoparticles homogenously dispersed within the organic matrices. It has shown that the size and size distribution of the silver nanoparticles is smaller and more uniform in the case of eggshell membrane matrix implying lower silver mobility within this matrix. The antibacterial activity was higher for the silver nanoparticles synthesized with co-milling with *Origanum* plant than in the case of milling with eggshell membrane.

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1. Introduction

Silver nanoparticles (Ag NPs) belong among the mostly studied topics nowadays, as can be seen from numerous recent review papers [1–3]. The most attractive seems to be their antibacterial activity [4–8]. The synthetic approaches towards the synthesis of silver nanoparticles are very numerous [9]. One of the very popular topics today is the so-called green synthesis using natural materials, most often plants [10–12]. The compounds responsible for the reduction of silver ion are most often extracted in water or some non-toxic solvent and then silver nitrate, as a source of silver, is introduced. Sometimes, also a reducing agent is required in addition to substances present in those natural materials.

Eggshell membrane is a unique natural biomaterial with multidisciplinary applications [13], mainly because of its biocompatibility with human body. Namely, its use as a biotemplate [14], as a part of biosensor [15], bio-sorbent [16] and in medicine [17] is of particular interest. Also the ability of the eggshell membrane to reduce silver is known and this property has been used for the synthesis of silver nanoparticles in more papers [18–20]. However, in most of them, further reducing agent was used.

Very different plants have been used for the synthesis of silver nanoparticles including *Origanum vulgare* L. [21]. This plant is very beneficial for human health [22], belongs among the most favorite culinary herbs [23], is quite common in nature and contains substances with antibacterial and antioxidant activity [24].

Mechanochemistry is an expanding branch of chemistry which is used for the wide variety of synthesis of various materials, including organic [25] and inorganic ones [26]. The concept of applying this method for the synthesis of metallic nanoparticles is not new [27,28]. The mechanochemical approach was used also for the synthesis of Ag NPs, using only mortar and pestle [29,30], or high-energy milling [31,32].

In the present study we demonstrate a simple one-pot solid-state method for the preparation of silver nanoparticles with various shapes using ball milling of two natural materials exhibiting reducing properties with silver nitrate. To our best knowledge, the combination of bio-approach with mechanochemistry for the synthesis of silver nanoparticles was not reported until now.

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https://doi.org/10.1016/j.apt.2017.09.028
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Please cite this article in press as: M. Baláž et al., Bio-mechanochemical synthesis of silver nanoparticles with antibacterial activity, Advanced Powder Technology (2017), https://doi.org/10.1016/j.apt.2017.09.028

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2. Experimental

2.1. Materials

Silver nitrate (99.8%, Mikrochem, Slovakia) was used as chemical without further purification. *Origanum vulgare* L. plants were collected from nature. Eggshell membrane was collected in a similar way as already reported in [33].

2.2. Mechanochemical synthesis

In a typical procedure, 1.5 g of $AgNO_3$ and 1.5 g of biomaterial (*Origanum vulgare* L. plant or eggshell membrane) were milled in Pulverisette 7 Premium line planetary ball mill under the following conditions: air atmosphere, 18 tungsten carbide milling balls with the diameter 10 mm, ball-to-powder ratio 47, rotation speed of the planet carrier 500 min^{-1}, milling time up to 45 min. The final milled samples are referred to as follows in further text: Ag/ESM for the mixture of $AgNO_3$ milled with the eggshell membrane for 30 min and Ag/ORE for the mixture of $AgNO_3$ milled with the *Origanum vulgare* L. plant for 45 min. The samples were washed with the distilled water after the milling, in order to remove residual silver nitrate.

2.3. Characterization methods

The XRD patterns were obtained using a D8 Advance diffractometer (Bruker Germany) with CuKα (40 kV, 40 mA) radiation. All samples were scanned from 15° to 85° with steps 0.03° and 10 s counting time. The approximate crystallite sizes were calculated using the Scherrer's equation.

UV–Vis spectra were collected using the UV–Vis spectrophotometer Helios Gamma (Thermo Electron Corporation, Great Britain) working in the range 200–800 nm nm in a quartz cell by dispersing of synthesized particles in absolute ethanol by ultrasonic stirring for 10 min.

The zeta potential was measured using a Zetasizer Nano ZS (Malvern, Great Britain). The ZP values were obtained by applying the Helmholtz–Smoluchowski equation built into the Malvern zetasizer software. Prior to the measurement, 10 mg of the sample was sonicated in distilled water for 10 min. The measurements were repeated 3 times for each sample.

Scanning electron microscopy (SEM) images were recorded using a MIRA3 FE-SEM microscope (TESCAN, Czech Republic) equipped with the EDS detector (Oxford Instrument, United Kingdom). Prior to the analyses, the samples were dried at 70 °C.

Transmission electron microscopy (TEM) analyses were performed using a 200-kV microscope JEM 2100 (JEOL, Japan) with LaB_6 electron source and equipped with energy dispersive X-ray spectrometer (EDS) for chemical analyses. For TEM studies, the samples were ultrasonically dispersed in ethanol for 5 min and applied onto the lacey carbon-coated nickel grid and dried. Prior to the TEM analyses, the samples were coated with a thin layer of carbon to prevent charging under the electron beam.

The average diameter of the Ag NPs was determined by encircling the NPs in the TEM images and transforming the irregularly-shaped area of each grain into a circle with equivalent surface area. Between 100 and 300 NPs were measured altogether. The particle size analyses were performed using a dedicated microstructure analysis program Image Tool™ (University of Texas Health Science Center, San Antonio, Texas, USA).

2.4. Antibacterial activity

The antibacterial properties of the samples were evaluated by the agar well diffusion method by slight modification of the process reported in [34]. The tested bacteria (*S. aureus* CCM 4223, *B. cereus* CCM2010, *L. monocytogenes* CCM 4699, *E. coli* CCM 3988, *Salmonella enterica ser. Typhimurium* CCM 7205, and *P. aeruginosa* CCM 3989) were obtained from the Czech collection of microorganisms (CCM).

In brief, bacteria were cultured aerobically at 37 °C in nutrient broth (Oxoid, United Kingdom) with agitation, or on standard plate count agar (Oxoid, United Kingdom). Frozen stock cultures were maintained at -20 °C. Before the experimental use, cultures were transferred to liquid media and incubated for 24 h. Cultures were then subcultured in liquid media, incubated for 24 h, and used as the source of inoculum for each experiment.

Agar media was cooled to 42 °C after autoclaving, inoculated with liquid overnight bacterial culture to a cell density of 5×10^5 cfu/mL and 20 mL of this inoculated agar was pipetted into a 90-mm diameter Petri dish. Once the agar was solidified, five millimeters diameter wells were punched in the agar and filled with 50 μL of samples. The four different concentrations of samples were prepared by immersing the given amount of washed samples after milling in distilled water and subsequent sonication. Concretely, 3.5, 8.7, 17.7 and 35.5 mg were used in order to obtain 1, 2.5, 5, 10 mM concentrations, respectively. Distilled water was used as negative control and gentamicin sulfate (Sigma-Aldrich, USA) with the concentration 10 mM as a positive control. The plates were incubated for 24 h at 37 °C. After the incubation, the plates were photographed and the inhibition zones were measured by the ImageJ software, the values used for the calculation were mean values of 3 replicate tests.

The antibacterial activity was calculated by applying the formula:

$$\%\text{RIZD} = [(\text{IZD sample} - \text{IZD negative control})/\text{IZD gentamicin}] \times 100$$

where RIZD is the percentage of relative inhibition zone diameter and IZD is the inhibition zone diameter (mm). The statistical analysis was performed using one-way ANOVA with Dunnett's post hoc test in a GraphPad Prism5 software (GraphPad Software, USA).

3. Results and discussion

3.1. Mechanochemical synthesis

The formation of Ag NPs was pursued by powder X-ray diffractometry. The XRD patterns are presented in Fig. 1.

XRD patterns reveal that elemental silver forms in both systems already after a short milling time of 15 min. In the presence of eggshell membrane, the process is slightly faster, as the peaks corresponding to the silver nitrate are not observed already after 15 min of milling time, whereas in the case of *Origanum vulgare* L., they are still present in the sample milled for 30 min. The intensity of the peaks corresponding to elemental silver (JCPDS 01-087-0717) increases with longer milling time, their intensity is quite high after 30 min of milling in both samples. This indicates progressive reduction of initial silver nitrate to elemental silver during high-energy milling in the presence of ESM and ORE. Longer milling of $AgNO_3$/ORE system (45 min) results in a complete consumption of $AgNO_3$. In the case of $AgNO_3$/ESM mixture, longer milling times resulted in a slight contamination by the milling media (tungsten carbide, qusongite, WC). For this reason, the samples milled for 30 min with ESM and 45 min with ORE were selected as final. In further text, they are referred to as Ag/ESM and Ag/ORE. Mechanochemical syntheses of Ag NPs reported in previous works [31,32] lasted for much longer time. The reaction

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