



# Development of antimicrobial water filtration hybrid material from bio source calcium carbonate and silver nanoparticles



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## ABSTRACT

Biobased calcium carbonate and silver hybrid nanoparticles were synthesized using a simple mechanochemical milling technique. The XRD spectrum showed that the hybrid materials is composed of crystalline calcite and silver nanoparticles. The TEM results indicated that the silver nanoparticles are discrete, uncapped and well stabilized in the surface of the eggshell derived calcium carbonate particles. The silver nanoparticles are spherical in shape and 5–20 nm in size. The SEM studies indicated that the eggshells are in micron size with the silver nanoparticle embedded in their surface. The hybrid eggshell/silver nanocomposite exhibited superior inhibition of *E. coli* growth using the Kirby–Bauer discs diffusion assay and comparing the zone of inhibition around the filter paper disc impregnated with the hybrid particles against pristine silver nanoparticles.

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## 1. Introduction

The immobilization of distinct nanostructures on solid supports does not only overcome the inherent limitation of their individual nanoparticle structures but widen their scope of application [1–5]. The research and practical relevance of nanocomposite materials of noble metals particularly silver anchored on inorganic solid supports have been investigated. The increasing attention to nano base silver materials is mainly due to their broad antimicrobial activity and their potential application in a wide range of fields [6–10]. As a result, a variety of techniques have been used to reduce silver precursors to silver nanocomposite or nanoparticles. Some of these methods include photochemical techniques, sonochemical methods [11–14]. A variety of organic stabilizers or coating agents (such as polymers, ligands or surfactants) have been used to reduce aggregation and to stabilize the nanoparticles on their substrates [15]. These large organic compounds do not only increase the cost of the obtained material but also decreases the purity and accessibility of the active nanosites necessary for silver bioactivity. These polymeric surfactants could irreversibly bind to the nanoparticles and interfere with their surface physical and chemical properties and adversely affect the interaction of the nanoparticles with biological interfaces [16,17]. The synthesis of surfactant free and relatively uncapped and monodispersed metal silver nanoparticles

with improved properties is necessary. A number of attempts have been made to anchor silver nanoparticles on a variety of supports. Some of these solid supports include polymer matrices [18,19], carbon cages [20], natural fibers [21], glass and silica supports [22–24] and steel surfaces [25]. Particularly the synthesis of silver nanoparticles in the presence of porous calcium based material is a promising and beneficial approach. This technique is commonly used where  $\mu$ -CaCO<sub>3</sub> particles act as pH releasing templates for the fabrication of hollow polymeric host for the transport of protein and lower molecular weight drugs [26–28]. However few studies exist where silver nanoparticles are immobilized on  $\mu$ -CaCO<sub>3</sub> supports [29]. The use of porous calcium based materials as platforms for metallic nanoparticles are gaining attention because of their biological compatibility and sustain release properties [30]. In this regard calcium based materials have been used as anchors for silver nanoparticles. Literature exists on the successful use of porous calcium phosphate as biocidal silver composite material [31]. The attachment of silver nanoparticles to inorganic anionic clay matrix as nanostructured ensembles showed remarkable antibacterial activity [32]. While their synthetic techniques have avoided the use of organic surfactants, their use of chemical reduction agents and subsequent removal is labor intensive, costly, time consuming as the final products must be washed thoroughly to remove the residual reducing agents which also raises environmental concerns as residues are toxic. For instance calcium carbonate is extensively used in fabricating inorganic composite capsules due to its excellent environmental compatibility, bio-compatibility and biodegradability. In addition, the pH selective release property of

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porous calcium carbonate materials makes them important vehicle for drug or nanoparticles material [33]. Previous works have mainly focused on the use of synthetic calcium carbonate hollow materials where organic surfactants and stabilizers have been used extensively to confer the desired stability, shape and porosity onto the calcium carbonate material [34,35]. As discussed earlier, these organic stabilizers could be toxic and their presence could hinder the interaction of nanoparticles and their biological sites which results in the reduction of the effectiveness of the nanoparticles. The size reduction of waste chicken eggshells to porous nanometer size scale particles is significant in a number of ways. It increases the surface area for adsorption and exposes the inherent macromolecular proteins [36] functional groups in the eggshell [37]. This is evident in the stacking of the platelike particles that arises from the non covalent interaction of the hydroxyl, carboxyl and amino functional groups of the protein and other macromolecules in the eggshell. These exposed functional groups could therefore be used to stabilize and immobilize silver nanoparticles onto the surface eggshell particles leaving most part of the silver nanoparticles exposed. The objective of this study is synthesized and immobilizes monodisperse silver nanoparticles on the surface of eggshell particles using less toxic materials and test for its antimicrobial activity.

### 1.1. Materials and methods

Eggshell materials were received from American Dehydration Food Inc (Atlanta, GA). The silver nitrate and polypropylene glycol were purchased from Sigma–Aldrich (Saint Louis, MO). The *Escherichia coli* (# 11775) was purchased from ATCC (Manassas, VA). The bacteria growth media LB agar was purchased from Fisher Scientific (Waltham, MA).

### 1.2. Extraction of calcium carbonate from waste eggshells

The eggshells were boiled in water overnight using a rice cooker with the temperature adjusted to 100 °C. The shells were cooled down to room temperature and the water was removed. The shells were then grinded using coffee grinder. The grinded colloidal sample was suspended in a large volume of water, stirred and allowed to settle. The supernatant was decanted from the calcium carbonate sediment. This was repeated at least three more times to ensure thorough removal of all loose organic macromolecules. The sample was dried in room temperature and then in oven at 60 °C to remove all volatile components. The sample was further ball milled using 6 mm stainless balls each in polypropylene glycol (PPG) for 5 h to fine colloidal solution. The inorganic component is then extracted from the organic component of the eggshell and the PPG following this procedure. The slurry sample was mixed thoroughly with ethanol and water in a volume ratio of 1:2. The sample is transferred to a separatory funnel and shaken vigorously and allowed to settle on the bench and separate out. The extracted calcium carbonate is then dried in room temperature overnight followed by drying for 3 h in the oven at 60 °C. The extracted sample was characterized using X-ray diffraction spectra (XRD).

### 1.3. Synthesis of eggshell/silver nanoparticles

The as-prepared eggshell powder was used without further treatment. Two grams of the eggshell powder was magnetically stirred into a uniform dispersion in 20 ml of deionized water and ethanol (1:2 v/v) mixture for 10 min. Two grams of silver nitrate was dissolved in 10 ml of distilled water. The dissolved silver nitrate solution is then added at a rate of 1.0 ml per minute into the eggshell mixture while the content was being stirred at 150 rpm for a total time of 10 min. The dispersed mixture was divided into two and each part added to 5 ml of PPG and ball milled for 2 h using

6 mm stainless balls each as shown in the scheme. The product was washed with 20 ml of distilled water and each time the supernatant was removed by centrifuging at 12,000 rpm for 5 min. In the last step the product was washed with 10 ml of ethanol and then dried in vacuum overnight. In comparative study, silver nanoparticles were prepared using the same procedure as the composite as follows: five hundred milligram (500.0 mg) of silver nitrate was dissolved in 20 ml of deionized water and ethanol (1:2 v/v). The solution was transferred to a stainless steel canister and ball milled for 2 h. A blank experiment was performed to investigate the effect of ball milling on the production silver nanoparticles from AgNO<sub>3</sub> without chemical or organic reducing agents. 2 g of AgNO<sub>3</sub> was ball milled in stainless steel canisters for 2 h.

### 1.4. UV–vis spectroscopy

The UV–vis spectroscopic studies were carried out using HP-Agilent Technologies 95-03 8453X UV–vis spectrometer.

### 1.5. X-ray diffraction (XRD)

XRD analysis were carried out using Rigaku DMAX 2100 diffractometer with monochromatic CuK $\alpha$  radiation ( $\lambda = 0.154056$  nm) at 40 kV and 30 mA. The intensity data for the nanocomposite of eggshell/Ag powder were collected from 10 to 80° of  $2\theta$  at a scan rate of 0.5°  $2\theta$  per minute.

### 1.6. Transmission electron microscopy (TEM)

The size and shape of the nanoparticles were analyzed using a JEOL-2010 transmission electron microscope (TEM), operating at 80 kV. Samples were prepared by dispersing 5 mg of as synthesized eggshell/silver sample in ethanol by sonicating for 5 min and depositing a drop of the colloidal solution on a carbon grid (carbon coated copper grid, removing the excess solution using a tissue paper and drying at room temperature).

### 1.7. X-ray photoelectron spectroscopy (XPS) analysis

The surface chemistry of the composite eggshell/nAg powders was investigated using X-ray photoelectron spectroscopy (XPS) measurements. The XPS spectra were acquired with a load-locked Kratos XSAM 800 surface analysis system equipped with 127 mm radius double-focusing concentric hemispherical energy analyzer which was also fitted with an aberration compensated input lens (ACIL), using MgK $\alpha$  (1253.6 eV) radiation source exclusively. Wide and detailed XPS spectra which correspond to medium resolution to high signal-to-noise spectra respectively were collected using FAT (fixed analyzer transmission) mode operating at 80 eV. The magnification of the analyzer in the FAT mode was selected to collect electrons from the smallest allowable (5 mm<sup>2</sup>) area on the specimen. The samples were mounted on the sample holder using a double-sided adhesive carbon tape with specimens pressed into the double-sided carbon tape to a thickness which insured that the emitted photoelectrons would originate only from the specimen. The sample was transferred to the analysis chamber where the base pressure of the ion- and turbo-pumped system was  $8 \times 10^{-9}$  Torr as read on a nude ion gauge. The elemental composition was first determined using a wide spectra which was then followed by a detailed acquired spectra for quantitative (peak area) and chemical states analysis. The resolution of the instrument at the operating parameters was measured from FWHM of the Ag3d<sub>5/2</sub> peak to be 1.0 eV. The XPS energy scale was calibrated by setting the Ag3d<sub>5/2</sub> line on clean silver to exactly 368.3 eV referenced to the Fermi level. Due to specimen charging during X-ray irradiation, the energy axis of each XPS spectra has been shifted to make the C1s

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