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Assessment of antibacterial properties of novel silver nanocomposite

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ABSTRACT

Recently, developed nanocomposite films with highly efficient antimicrobial properties are considered. This study investigated the antibacterial effects of tetraethyl orthosilicate–3-Tri-methoxysilyl-1-propanethiol–silver nanoparticle (TEOS–TMSPT@AgNPs) composite and pure silver nanoparticles (AgNPs). Antibacterial activities of AgNPs and TEOS–TMSPT@AgNPs were examined against the test bacteria *Escherichia coli, Salmonella typhimurium, Staphylococcus aureus and Bacillus cereus*, using disk diffusion, minimum inhibitory (MIC) and bactericidal (MBC) concentration methods. Comparing the antibacterial activity of AgNPs and TEOS–TMSPT@AgNPs according to the zone of inhibition in millimeter (mm) showed that TEOS–TMSPT@AgNPs in addition to keeping the antibacterial activity of AgNPs, increase this activity against different bacterial strains. Composite containing AgNPs showed the lower Ag^+ release than pure AgNPs when the storage time exceeds 21 days. These results suggested that TEOS–TMSPT@AgNPs could be a very interesting slow releasing nanocomposite film with antibacterial activity.

1. Introduction

Silver, a strong antimicrobial agent, has been used since the ancient times while silver nanoparticles have been used for more than one century [1]. Silver nonomaterials refer to any materials containing silver with an enhanced activity due to their nanoscale structures. Different forms of silver nanoparticles are often synthesized by chemical reduction from silver salt as a precursor [2]. Although the mechanism of nanosilver on bacteria is not fully understood, the four mechanisms of bactericidal activity suggested to date include: (1) direct attack and damage to cell membrane, (2) generation of reactive oxygen strains (ROS), (3) disruption of ATP production and (4) cessation of DNA replication [3].

Today, the antimicrobial properties of silver nanoparticles make it suitable for use in different kinds of researches and industries such as food packaging [4], implants and medical devices [5], textile [6], *etc*. Many factors can influence the bactericidal effect of silver nanoparticle, such as size, shape, dose, surface charge, surface coating, *etc*. In the past few years, the production of silver nanoparticles in combination with other materials, especially the production of novel silver based composite materials with new physical, chemical and biological properties, has been considered [7–11].

Kubacka et al. produced nanocomposite films consisting of ethylene–vinyl alcohol copolymer (EVOH) embedded with Ag–TiO_2 nanoparticles, exhibiting antimicrobial activity. This activity can be enhanced by irradiation with ultraviolet (UV) light [10]. According to [12], silver complexes of poly (amidoamine) (PAMAM) dendrimers displayed considerable antimicrobial activity without the loss of solubility and activity. Sanpui et al. synthesized a novel chitosan Ag nanoparticle composite against *Escherichia coli.* Chitosan has been used in many applications due to its inherent biodegradability and biocompatibility properties. This composite was more efficient than either AgNPs or chitosan alone for inactivating bacteria, possibly due to the synergistic effect of both AgNPs and chitosan in the composite [13]. Therefore, development or modification in silver nanoparticle compounds to improve bactericidal activity and reduce the risk for human and the environment is a new area of research in nanotechnology field.

A serious problem in using nanomaterials, especially nanosilver, is the release of nanoparticles into the environment. It is obvious that the release of Ag^+ from silver nanoparticles contributes to the bactericidal efficacy of these nanoparticels [14–16], but its release needs to be under control. AgNPs could be engineered with different surface coatings to release Ag ions at the desired rate [17]. Thus, new compounds which show antimicrobial effects without releasing toxic biocides are of strong interest. Some studies have been conducted to make slow release of nanosilver into its environment; for example polyelectrolyte multilayer films that sustained the release of Ag^+ [18]. Also, numerous studies have been directed toward improving the biocidal properties of nanosilver in addition to the slow release

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of particles. According to [19], nanocomposites of iron oxide and silver nanoparticles showed very significant antibacterial and antifungal activities. Due to the magnetic properties of iron oxide and the controlled release of Ag^+, these new nanocomposites could be used in medicinal and disinfection applications. As regards, there are several ways to make antimicrobial nanomaterials. Creating nanocomposite films is one of these approaches that can help to slow the release of nanosilver. Kumar et al. prepared polyamide/silver (PA/Ag) composite materials. They showed that matrix crystallinity was very decisive in determining the silver ion releasing properties and hence, the antimicrobial efficacy of silver–based antimicrobial polyamides [20]. On the other hand, making the antibacterial films is a suitable strategy for the prevention of initial adhesion and then colonization of bacteria onto the surfaces. Chiang et al. showed that the silver–palladium surfaces inhibited biofilm formation by generating microelectric fields [21]. Therefore, biocidal activities at the low concentration of silver and the good distribution of silver on the surface of composite films could be an appropriate approach for increasing the antibacterial properties and maximizing the slow release of nanosilver.

Aggregation or agglomeration of nanoparticles is the main problem in nanoparticles synthesis and stabilization, limiting their application. Therefore, synthesis and stabilization of non–aggregated nanoparticles remain a goal. Different inert matrices, titania [10,20] and silica [22], as a carrier for silver nanoparticles, can be used to disperse them, but these modifications may alter the antibacterial behavior of AgNPs. Composites consisting of inert matrices coated with metallic silver have been shown to be potentially useful for infection control [23,24]. Silica makes an appropriate substrate for silver–silica nanocomposites. These composites are stable at high temperature and useful for photonic devices [25,26]. TEOS is mainly used as a silica source in silicone polymers and also, as a crosslinking agent for sol–gel methods [27]. The formation of sol–gel from hydrolysis and the condensation of silane coupling agents is a potential process which can be applied to strengthen the boundary of films, modify surfaces, and improve the properties [28]. However, silanol groups from TEOS do not lead to the good bonding of silane molecule on metal [29]. TMSPT has been shown to form a strong covalent bond with different metals such as copper, silver and gold [30,31].

In this study, by preparing the highest surface nanomaterials with many reactive surface sites and therefore, enhanced reactivities, tetraethyl orthosilicate-3-Tri-methoxysilyl-1-propanethiol-silver nanoparticle (TEOS–TMSPT@AgNPs) was evaluated by Fourier-transform infrared (FTIR) spectroscopy, X–ray diffraction (XRD) and Field emission scan electron microscope (FESEM). Finally, the resulting nanocomposite film was tested for releasing Ag^+ in water and evaluated against both gram positive, *Staphylococcus aureus* and *Bacillus cereus*, and gram negative, *Escherichia coli* and *Salmonella typhimurium,* bacteria.

2. Experimental

2.1. Materials

Chemical Tetraethyl orthosilicate, 3-Tri-methoxysilyl-1-propanethiol and ethanol were purchased from Merck (Darmstadt, Germany). Other reagents were of analytical grade also purchased from Merck (Darmstadt, Germany).

2.2. Nanoparticle preparation

The synthesis of silver nanoparticles was adapted from a previously reported study [32]. To prepare silver nanoparticles, 50 ml of 5 mM of $AgNO_3$ was added drop–wise to 50 ml of 10 mM of sodium borohydrate into a beaker and allowed for 2 h to reduce the silver ions into AgNPs.

2.3. Preparation of TEOS–TMSPT@AgNPs nanocomposite and TEOS @AgNPs nanocomposite

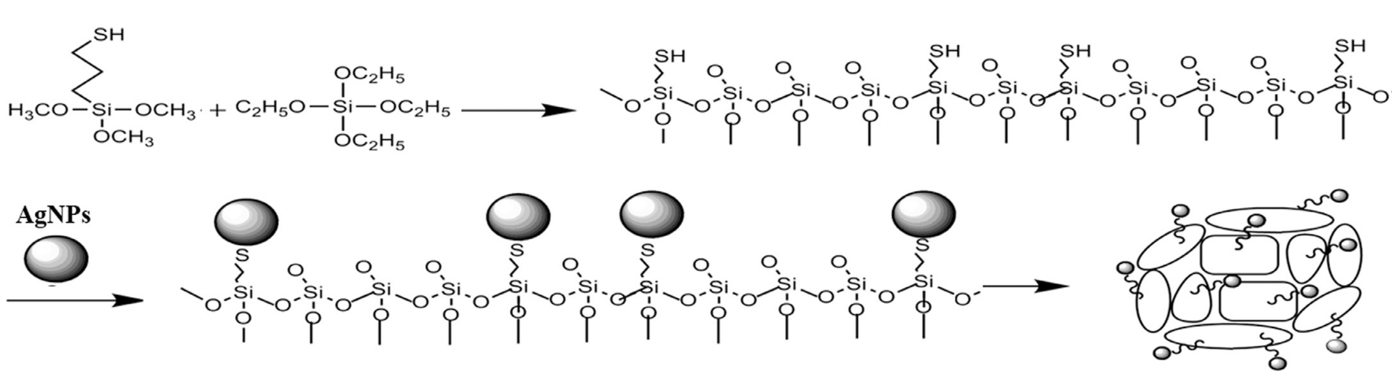
First of all, to prepare TEOS–TMSPT@AgNPs nanocomposite, TEOS–TMSPT sol gel solution was prepared as follows. TEOS in absolute ethanol was placed in an experimental beaker, with the amount of TEOS being 50% by weight relative to the total weight of the resulting solution. This solution was stirred until the TEOS was dissolved. Next, TMSPT was added to the solution with the amount of TMSPT being 5% by weight relative to the weight of TEOS. The resulting solution was stirred for 30 min. The catalyst solution was prepared by adding 0.15 g of NH_4OH (3.1 wt%) into the water (4.85 g). When the TMSPT was completely dissolved in the solution, the catalyst solution was added. The weight ratio of catalyst solution to TEOS–TMSPT solution was kept at 1:10. The hydrolyzed solution was stirred magnetically for 3 h, and then left at room temperature without stirring for 21 h. After being at room temperature for 21 h, the solution was diluted with absolute ethanol to around 5% by the weight of SiO_2 equivalent.

To prepare TEOS–TMSPT@AgNPs nanocomposite, silver nanoparticles were deposited on TEOS–TMSPT sol-gel solution as follows; 1.0 mg of silver nanoparticles were dispersed in 10 ml of TEOS–TMSPT sol-gel. The mixture was sonicated at room temperature for 30 min using a low–intensity ultrasonic cleaner bath. The schematic of the achieved TEOS–TMSPT@AgNPs nanocomposite is shown in Scheme 1.

TEOS@AgNPs nanocomposite was maked same method for preparation of TEOS–TMSPT@AgNPs nanocomposite.

2.4. Characterization

To evaluate the silver nanocomposite film formation, Ag embedded percentage and distribution, low release effect and biocidal property, some analysis was conducted. For surface morphology



Scheme 1. Schematic representative for the preparation of TEOS–TMSPT@AgNPs nanocomposites.