Organic-free growth of gold nanosheets inside 3D bacterial cellulose as highly efficient and robust antibacterial biopolymers

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ABSTRACT

Without any chemical agent, gold nanosheets (AuNSs) were controllable synthesized through a facile photo-induced reduction within bacterial cellulose (BC) biopolymers. Compared with traditional polymers, AuNSs modified BC biopolymers (AuNS@BC) biopolymers exhibited similar levels of softness, ductility, and better tensile strength. The in situ constructing of AuNS@BC biopolymers was demonstrated to provide great reusability and antibacterial activities and towards both of Staphylococcus aureus (S. aureus) and Escherichia coli (E. coli). The optimized AuNS@BC biopolymers remain at least 95% antibacterial activities after three cycles. The facile and shape-controlled synthesis of AuNS@BC biopolymers is believed to be useful for the design and application of biomass-based medical dressing.

Introduction

During the outbreak of Coronavirus Disease 2019 (COVID-19), healthcare has aroused people’s considerable attention due to the infectious diseases which caused by bacteria or pathogenic microorganisms [33, 34, 37]. Furthermore, along with the rapid development of metallurgical industry [27], pharmaceutical industry [24, 31], and food industry [14, 47], manufactured antibacterial materials have been one of the most important projects during the past few years. Among various morphologies, two-dimensional (2D) antibacterial materials have been proved to be an ideal form for further the design of wound dressings [14], mobile phone screens [39], facial masks [1], etc.

Till now, most of the artificially synthesized antibacterial films are polymer-based products, such as polyvinylpyrrolidone (PVP) [16], polyethylene
glycol terephthalate (PET) [8, 40] and chitosan (CS) [15, 44]. To strengthen the antibacterial activity of polymers, the most common way is adding organic antibacterial agents (e.g., benzimidazoles [30], N-halamines [9], quaternary ammonium salt [23]. However, these organic agents have significant defects of poor-stability and low-safety in particular. As a result, inorganic antibacterial active-sites, e.g., CuO [41], ZnO [12], TiO₂ [25], and Al₂O₃ [36], have raised great interest in the antibacterial field. In order to improve further, noble metal (e.g., Ag, Au) nanomaterials have been introduced to antibacterial films. For instance, Ag nanoparticles (AgNPs) modified PET fibers [3, 7], AgNPs modified CS gelatin [43], and Au nanoparticles (AuNPs) modified cotton fabrics [38] all presented antibacterial enhancement. Recently, the requirement for comprehensive utilization of natural biomass is going up. Among various kinds of biopolymers, bacterial cellulose (BC) is well-known as polysaccharide polymer composed of glucan chains linked by β-1,4-glycosidic bonds, which has attracted significant interest owing to its favourable properties for its great bio-degradability, excellent bio-compatibility, inexpensive, non-immunogenicity [4], etc. Notably, BC is a hydrogel pellicle containing > 98% water due to its high swelling capacity. It has been proved that BC biopolymers could act as an ideal platform for the antibacterial aspect [11]. So far, several inorganic active-sites have been successfully modified based on this biomass, such as nanostructures of CuO [29], TiO₂ [32], Ag₂O [20], AgNPs [42], and gold nanoparticles [5]. Among all of these materials, Au nanomaterials have excellent biocompatibility and no significant cytotoxicity. As a broad-spectrum antimicrobial agent, Au nanomaterials mainly cause cell lysis and death by electrostatic adsorption, membrane damage and reactive oxygen species (ROS) damage to proteins and DNA. The antibacterial abilities of Au nanomaterials are closely related to the size, dispersity, and surface states which needs thorough analysis and investigation [13]. However, the morphological control of 2D gold nanosheets (AuNSs) and its effect on the antibacteria have not been extensively studied [19, 45]. In particular, the following issues will be addressed: (1) whether the 2D surface-clean Au structures can be synthesized inside 3D bacterial cellulose, (2) whether the Au modified BC biopolymers have outstanding antibacterial activities.

Based on our previous work, photo-induced reduction method was proved to be an efficient route for the in situ synthesis of gold nanosheets within the organic liquid crystals (LCs) templates [48]. So far, the fabrication of AuNSs without organic reagent still remains a great challenge. Besides, BC biopolymers were used in our laboratory as a novel biomass template to prepare different nanostructures including 2D CdS nanosheets [49] and 1D ZrTiO₄ nanotubes [6]. To the best knowledge of the authors, the combination of photo-induced reduction and biomass template is far less investigated. To confirm the hypotheses mentioned above, we attempted to design and explore a new route for the organic-free growth of gold nanosheets inside 3D bacterial cellulose. Compared to the traditional Au@BC composites with organic agents that hard to remove [5], our surface-clean AuNSs-contained BC would be a promising active material in various antibacterial applications, including water purification, medical dressing, and food packages.

Materials and methods

Materials

HAuCl₄·4H₂O (≥ 45%, molecule weight = 411.85) was obtained from Aladdin (Shanghai, China). The chemical characterization information of bacterial cellulose (BC) was carefully analyzed in this work (50 ~ 100 mm diameter; > 20 μm length; 80 GPa young’s modulus; 4000 MPa tensile strength; ~ 7% elongation; type I polymorph, 1 ~ 2% solid content). BC membranes were purchased from Guilin Qihong Technology Co., Ltd (China). The cryopreservation freeze-dried of the strain is obtained from the American Type Culture Collection (ATCC) including Escherichia coli (E. coli, Gram-negative bacteria, ATCC-25922) and Staphylococcus aureus (S. aureus, Gram-positive bacteria, ATCC-6538).

Purification of BC biopolymers

The preparation of BC biopolymers was preformed according to our previous work [17]. Briefly, the preparation of BC aerogel is listed as follows. First of all, BC biopolymers were purified by soaking in NaOH aqueous solution (~ 2 wt%) at 90 °C for 3 h and washed by deionized water before adjusting the
pH value to 7 by acetic acid. Secondly, pretreated BC biopolymers were washed by deionized water several times to get rid of impurities. Finally, BC biopolymers were collected after freeze-drying for 24 h. All water used in this work was deionized. All reagents used were without any additional treatment.

**Preparation of Au modified BC biopolymers**

In a typical preparation, low-pressure mercury lamp of 254 nm (8 W) was used as an ultraviolet irradiation source. 10 mL solution of 60 mmol/L HAuCl₄ together with 0.1 g bacterial cellulose (BC) were placed ~ 20 cm away from the mercury lamp in a petri dish. After turning on the light, the reaction system was irradiated at 25 °C for 0.5 h, 2 h, and 12 h, respectively. After time-equally illumination for both sides, the biopolymers were washed with deionized water to remove the excess chemicals three times, after freeze-drying for 24 h, which were named as AuNPs@BC, AuNSs-S@BC, and AuNSs-L@BC.

**Characterizations**

The bacterial cellulose biopolymers loaded with gold nanostructures were examined by X-ray diffraction (XRD) under a Rigaku D/Max-2500PC X-ray diffractometer (Rigaku Co., Ltd., Japan) employing Cu Kα radiation, λ = 1.54056 Å operated at 40 kV and 100 mA. A JEM-2100 transmission electron microscope (TEM, JEM-2100, Japan) and a JSM-6360LA scanning electron microscope (SEM, JEOL, Japan) were used to characterize the morphology of the samples. Mechanical properties of biopolymers were measured by CMT6103-MTS. The maximum force is 200 N with instrument force of 1000 N.

**Measurements of antibacterial activity**

Different strains concentrations (1.5 × 10³ CFU/mL, 1.5 × 10⁴ CFU/mL, and 1.5 × 10⁵ CFU/mL) were carried out to test the best zone of inhibition on the sample. Blank BC, AuNPs@BC, AuNSs-S@BC and AuNSs-L@BC were cut into round pieces in diameter of 10 mm. The zone of inhibition recorded under the same experimental condition. The zones of inhibition (ZOI) bacteria were estimated and compared by disk diffusion method. The antibacterial effect was selected at bacterial concentrations of 1.5 × 10⁵ CFU/mL for both two bacterial strains. Round pieces were placed on the nutrient broth agar that contains *S. aureus* or *E. coli* which incubated at 37 °C for 24 h. Before the test of cycle stability, the cleaning process for recycle use contained alcohol washing three times, deionized water washing three times, and oven-drying under 60 °C overnight.

**Measurements of the cytotoxicity**

Cytotoxicity of the Au modified BC biopolymers were tested with the Hela using the CCK-8 assay method [22]. Firstly, Hela were cultured in MEM and incubated at 37 °C with 5% CO₂. Subsequently, the Hela cells were extracted counted at log phase. Hela cells were added into 96-well plates with a density of 6 × 10⁵ cells/well and cultured at 37 °C overnight. The samples were divided into 5 groups according to the control group. Sample 0 (BC), sample 1 (AuNPs@BC), sample 2 (AuNSs-S@BC), and sample 3 (AuNSs-L@BC) were incubated with 0, 1, 6, 8, 16, 32, 100, and 200 mg/L. After cultured for 24 h, remove the medium before washing each well with PBS for three times and add 100 μL/well to medium containing 10% CCK-8, 5% CO₂, and incubate in a constant temperature incubator at 37 °C for 2 h. The cytotoxicity was measured by CCK-8 assay and detected by Tecan SPARK 10 M multimode reader at 450 nm. Relative cell activity% = (OD value of sample–OD value of background)/(OD value of control group–OD value of background).

**Results and discussion**

**In situ growth of AuNSs within BC biopolymers**

The schematic illustration of AuNSs construction within BC biopolymers is illustrated in Fig. 1. As a natural biomass, the surface of BC is massively covered by hydroxyl (–OH) groups. It has been proved in our previous work that these groups possess enough reduction capability under the irradiation of ultraviolet (UV) light [48]. Compared with traditional thermal annealing process, this photo-reduction method was milder and more controllable. However, due to the totally different shape and size of fibers, not all the plant cellulosics could provide an ideal platform for the growth of 2D Au structures. For example, on
the surface of cotton fiber, the morphology of Au was large particles under the same condition [10]. Thus, after 0.5 h irradiation, small Au\(^0\) nanoseeds emerge and grow to be AuNPs on the fibers of BC substrates during the first step. Furthermore, after 2 h irradiation, most of the AuNPs gradually grow up and become 2D-shaped small Au nanosheets (AuNSs-S@BC) in the scaffold during the second step. Finally, after 12 h continuous reduction of the third step, a great number of large AuNSs are anchored to form AuNSs-L@BC. Particularly remarkable that during the whole process, no protectant or reductant is used and the synthesis is organic-free.

**Characterization analysis of Au modified BC biopolymers**

**SEM characterization**

As shown in Fig. 2, the growth process of AuNSs has been confirmed scientifically by scanning electron microscope (SEM) and transmission electron microscopy (TEM). Original BC biopolymers exhibited a typical 3D cross-linked feature and the average diameter of fibers are about 80 nm (see Fig. 2a). After been irradiated for 0.5 h in 60 mmol HAuCl\(_4\), it was found in Fig. 2b that small Au particles were formed and bonded upon the surface of BC (AuNPs/BC). After driven by continuous photo-reduction, the crucial growth to small AuNSs can be observed in Fig. 2c. Similar evolution from the AuNPs to the AuNSs has been reported by Li [21]. The AuNPs (~ 60 nm of diameter) and AuNSs (~ 1 \(\mu\)m of diameter) were coexisted with in the third step. Finally, after irradiation for 12 h, these small AuNSs constantly grew up to ~ 2 \(\mu\)m of average side length in Fig. 2d.

The enlarged photograph of Au nanostructures and the detailed relationship between irradiation time and the morphology can be checked in Fig. 3. Interestingly, these 2D AuNSs appeared flexibility. Compared with the current reports [2, 7, 46], the curled or ruffled side of AuNSs were morphological different and appeared like the margins of certain leaves (see supplementary materials). Notably, these AuNSs were highly dispersed in the system of BC because of the BC fibers were adsorbed on the top and bottom of the flexible AuNSs structures that can prevent the aggregation. Actually, the morphology characterization also proved that small AuNSs were closely contacted with the BC fibers. Similar mechanism was reported by Boomi et al. [2].

**TEM characterization**

Figure 4 shows the typical transmission electron microscopy (TEM) images of AuNPs, AuNSs-S, and AuNSs-L. Interestingly, AuNSs-S@BC contains several different structural morphologies of (a) quadrilateral, (b) isosceles trapezoid, (c) rounded equilateral triangle, and (d) equilateral triangle. Besides, the average sizes of these geometries were about 50 ~ 100 nm (see supplementary materials). All the angles contained by two adjacent edges laid within 57 ~ 63° or 115 ~ 121°. It is worth mentioning that the moment of “dynamic growing” in Fig. 5a, b around the rounded equilateral triangle was observed clearly for the first time. It is consistent with the theory that nano-twin of Au nanoseeds can provide active growth sites for the assembling of Au nanoseeds [50]. BC fibers provided ideal platform for the slowly growth of Au nanoparticles and small triangular/trapezoid nanoplates. A possible schematic illustration of the growth process is listed in the supplementary information. As displayed in Fig. 4e,
a typical high-resolution TEM (HRTEM) image of the obtained AuNSs-S revealed the single crystals structure with a fringe spacing of ~ 0.235 nm, which corresponds to {111} reflection. In Fig. 4f, the hexagonal diffraction spots further confirm that these AuNSs are single crystals with highly oriented (111) planes. Based on the above results, the growth mechanism of AuNSs within BC under UV irradiation is shown in Fig. 5c. More TEM images of the evolution from particles to small nanosheets and finally large AuNSs can be seen in the supplementary materials.

XRD characterization

The X-ray diffraction (XRD) patterns of the blank BC, AuNPs@BC, AuNSs-S@BC, and AuNSs-L@BC are listed in Fig. 6. Three intensive diffraction peaks of BC appeared at 14.2°, 16.7°, and 22.3° which corresponding to the crystallographic planes of (1 1 0), (1 1 0), and (2 0 0) diffraction planes. Besides, diffraction peaks of (1 1 1), (2 0 0), (2 2 0), and (3 1 1) can be clearly seen which prove the face-centred cubic (fcc) crystal of Au. As indicated by the strong (1 1 1) diffraction peak, the {111} facets are the dominating facets. The (2 0 0)/(1 1 1) relative diffraction intensity value of the obtained AuNSs-S is 0.103 which is much lower than Au bulk value of 0.539 (JCPDS 04-0784), suggesting a
preferred orientation of nanosheets in the (111) direction. Particularly, the (200)/(111) relative diffraction intensity values of AuNPs@BC, AuNSs-S@BC, and AuNSs-L@BC are 0.099, 0.103 and 0.282, respectively. The relative diffraction intensities of (200)/(111) increase upon prolonging the irradiation time of UV light, indicating that the preferred orientation of the (111) direction in larger size AuNSs is further intensified.

**Performance analysis of Au modified BC biopolymers**

**Antibacterial activities**

Together with blank BC biopolymers, the antibacterial activities of AuNPs@BC, AuNSs-S@BC and AuNSs-L@BC biopolymer were determined by bacterial strains of Staphylococcus aureus (*S. aureus*, Gram-positive bacteria, ATCC-6538) and Escherichia coli (*E. coli*, Gram-negative bacteria, ATCC-25922). The antibacterial experiment was selected at bacterial concentrations of $1.5 \times 10^5$ CFU/mL for both two...
bacterial strains. All films were cut into round pieces in diameter of 10 mm before placed on the nutrient broth agar at 37 °C for 24 h. As shown in Fig. 7a, d, pure BC does not show any antibacterial activity; however, after the construction of Au nanostructures inside BC, the incubation of *E. coli* and *S. aureus* were inhibited obviously. The zones of inhibition (ZOI) results of bacteria were estimated and compared by disk diffusion method. As for *S. aureus*, the ZOI diameters of blank BC, AuNPs@BC, AuNSs-S@BC and AuNSs-L@BC are about 10.0 mm, 12.5 mm, 16.5 mm, and 15.0 mm, respectively. For comparison, the ZOI diameters for *E. coli* are about 10.0 mm, 10.0 mm, 17.0 mm, and 11.5 mm (see Fig. 7a, b). The maximum value of ZOI is measured around AuNSs-S@BC against *E. coli* (~ 17 mm). This phenomenon is consistent with the result which explained by Lee et al. and Shan et al. that the two belongs to different types of cell wall [18, 35]. Besides the above agar disk diffusion method, bacterial incubation data were also recorded by the optical density of the liquid culture medium (see Fig. 7c, d). The results of optical density at 600 nm (OD600) were tested for 24 h with 2-h interval. As for the AuNPs@BC, the values of OD600 reach the peaks at 12 h for *S. aureus* and at 14 h for *E. coli* (black lines). The OD600 curve of AuNPs@BC with platform pattern indicate very weak antibacterial activity. Besides, the OD600 values of AuNSs-L@BC are different in two bacterial strains (red lines, *S. aureus* < *E. coli*). It means antibacterial activities of AuNSs-L@BC for *S. aureus* is better than that of *E. coli*. Instead, the OD600 of AuNSs-S@BC restricted in a low level of 0.2 ~ 0.28 for both *S. aureus* and *E. coli* (blue lines). All the results above showed that AuNSs-S@BC biopolymers have the best antibacterial activity for both gram-positive and gram-negative bacterial strains.

![Figure 6](image_url)  
**Figure 6** X-ray diffraction pattern of blank BC (black line), AuNPs@BC (green line), AuNSs-S@BC (blue line) and AuNSs-L@BC (purple line).

![Figure 7](image_url)  
**Figure 7** Bacterial growth inhibition zone of (a) *S. aureus* and (b) *E. coli*; and bacterial growth curve against (c) *S. aureus* and (d) *E. coli* in the presence of AuNPs@BC (black lines), AuNSs-S@BC (blue lines) and AuNSs-L@BC (red lines).
bacteria. A possible reason is small AuNSs can efficiently generate holes in the cell walls which causes the leakage of the cell contents. Furthermore, compared with small AuNPs and large AuNSs, small AuNSs present the highest percentage of exposed facets, which favors the direct interaction of small Au nanosheets with bacterial surface and leads to the enhanced surface binding, cell uptake, and efficiently killing of bacteria [28].

**Mechanical properties**

The mechanical properties of membranes are important for many applications especially medical dressing. Supplementary materials indicate the Young’s modulus, tensile strength and elongation of as-prepared biopolymers. Compared with the blank BC, Young’s modulus of the AuNSs-S@BC greatly decreased from 8.14 to 3.78 GPa, which represents a better malleability [26]. Moreover, after introducing AuNSs, the tensile strength of AuNSs-S@BC was promoted from 2072 to 3180 MPa and the elongation also grew up from 45 to 56%. Furthermore, with the comparison of traditional polymer-based PET and cellophane films, AuNSs-S@BC biopolymers exhibited similar level of softness, ductility, and better tensile strength. Briefly, the mechanical properties of blank BC biopolymers have been actually improved after the in situ growth of AuNSs. On this basis, we further proved the recycling performances of AuNSs-S@BC biopolymers, which remained 97% activity of S. aureus and 95% activity of E. coli after three times circulation (see Fig. 8). As a result, AuNSs-S@BC biopolymers exhibited outstanding mechanical property and excellent cycling behavior.

**Cytotoxicity test**

It is really true that cytotoxicity test is in the future design and application such as biomedicine materials. Furthermore, the cytotoxicities of the Au modified BC biopolymers were tested with the Hela using the CCK-8 assay method. As shown in Fig. 9, the AuNSs@BC biopolymers present the highest relative cell activity of 98.7% at 100 mg/L, which may benefit from the growth-promoting effect of BC.

![Figure 8](image.png)

**Figure 8** Reusability of blank BC, AuNPs@BC, AuNSs-S@BC, AuNSs-L@BC before and after three times antibacterial circulation under (a) S. aureus and (b) E. coli, which evaluated by the inhibition zone diameters (mm).

**Conclusion**

A series of AuNSs were creatively fabricated with biomass template of BC biopolymers via a simple photo-induced reduction method. The Au nanoseeds were proved dynamic assembled to form uniform Au nanosheets with size from 60 to 2 μm. With anchored AuNSs, BC biopolymers exhibited enhanced mechanical properties and excellent antibacterial activity against both S. aureus and E. coli bacteria. After three cycles, AuNSs-S@BC film remains at least 95% activity. This in situ process of photo-induced reduction by BC biomass template is hopeful to be expanded in the synthesis of other noble metal-contained cellulose composites. The novel system of surface-clean AuNSs@BC biopolymers has great potential in biomedical fields.
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Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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