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Mussel-inspired nano-multilayered coating on magnesium alloys for enhanced corrosion resistance and antibacterial property

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ABSTRACT

Magnesium alloys are promising candidates for load-bearing orthopedic implants due to their biodegradability and mechanical resemblance to natural bone tissue. However, the high degradation rate and the risk of implant-associated infections pose grand challenges for their clinical applications. Herein, we developed a nano-multilayered coating strategy through polydopamine and chitosan assisted layer-by-layer assembly of osteoinductive carbonated apatite and antibacterial sliver nanoparticles on the surface of AZ31 magnesium alloys. The fabricated nano-multilayered coating can not only obviously enhance the corrosion resistance but also significantly increase the antibacterial activity and demonstrate better biocompatility of magnesium alloys.

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1. Introduction

Magnesium (Mg) alloys are promising candidates for load-bearing orthopedic implant materials due to their biodegradability, mechanical compatibility and biological safety which can avoid second removal and reduce the stress-shielding effect [1–3]. However, the rapid degradation of Mg alloy in the physiological environment does not match the healing or growth of new bone tissues which will often cause hydrogen accumulation and local alkalization [4]. Various surface modification strategies have been proposed to increase the corrosion resistance of Mg alloy [5–12]. Among them, alkaline-heat treatment [13] and calcium phosphates coatings [8,14,15] are two typical effective methods.

On the other hand, for implant biomaterials, the possibility of bacterial adhesion to the surface of implant and subsequent biofilm formation at the implantation site can lead to implant failure [16,17]. The above mentioned two typical coating strategies usually lack of antibacterial activity. In order to prevent such implant-associated infections, various techniques including thermal spraying [18], electrochemical deposition [19–21] and magnetron co-sputtering [22], have been used to incorporate antimicrobial agents, such as silver, copper and zinc ions, into the surface coating [17,18,23–26]. These coatings have demonstrated good osteoconductivity and enhanced antibacterial activity.

Inspired by the composition of adhesive proteins in mussels, dopamine self-polymerization has been found to be able to form a thin, surface-adherent polydopamine (PDA) coating on a variety of material [27,28] and such PDA coating can further enhance the surface bioactivity [29] and promote cell adhesion and viability [30]. PDA-induced nanocomposite Ag/calcium phosphates coatings on the surface of titania nanotubes has demonstrated excellent antibacterial and osteointegration functions. In our previous work, with the aid of PDA, strontium substituted hydroxyapatite nanocrystals and bisphosphonate alendronate have been co-immobilized on the surface of AZ31 Mg alloy for osteoporotic repair [31]. Considering that carbonated apatite (CAp) nanoparticles have superior biocompatibility, bioactivity and osteoconductivity due to chemical and structural similarity to biological apatite [32], and that chitosan has excellent biocompatibility and biodegradability and has been used as antimicrobial agent [33], to simultaneously increase the corrosion resistance and antibacterial activity while enhance good biocompatility of magnesium alloys, herein, we developed a facile but effective nanocomposite coating strategy through PDA and chitosan assisted layer-by-layer assembly of biomimetic carbonated apatite (CAp) and sliver nanoparticles on the surface of AZ31 magnesium alloys, as schematically illustrated in Fig. S1. After alkaline pre-treatment, a PDA transition layer was deposited on the AZ31 Mg alloy substrate. Then, a chitosan intermediate layer was grafted through Michanael addition

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and/or Schiff-base reactions between the amino groups of chitosan and the catechol/quinone groups in PDA, followed by immobilization of a layer of PDA coated CAp (CAp@PDA) nanoparticles, another chitosan intermediate layer and a layer of PDA coated Ag (Ag@PDA) nanoparticles. Such chitosan-CAp@PDA-chitosan-Ag@PDA periodic unit can be repeated for the required number of cycles.

The proposed coating strategy have effectively combined the advantages of the antibacterial activity of silver nanoparticles, the excellent bioactivity and osteoconductivity of biomimetitic CAp nanoparticles, the strong adesion of PDA and the biocompatibility of chitosan. The obtained nano-multilayered coating thus have multifunctions: (1) obviously increase the corrosion resistance of the magnesium alloy substrate; (2) significantly increase the antibacterial activity of the implants; and (3) effectively improve the biocompatility. Such coating strategy overcomes the conflicts between antibacterial property and biocompatibility, provides deep insights into surface functionalization of magnesium alloys and can potentially be extended to design of many other orthopedic devices.

2. Materials and methods

2.1. Materials

AZ31 Mg alloy ingot was supplied by National Engineering Research Center for Magnesium Alloys, P. R. China and the as-received ingot was cut into discs with 10 mm in diameter and 2 mm in thickness using a wire-cut electrical discharge machine. Dopamine hydrochloride, tris(hydroxymethyl) aminomethans (Tris) (99.0%), chitosan (low molecular weight, deacetylation degree = 98%), silver nitrate ($AgNO_3$, ≥99.8%), $NaHCO_3$, Na_2HPO_4, $Ca(NO_3)_2{\cdot}4H_2O$, and 3-(4,5-dimethylthiazol-2yl)-2,5-diphenyltetrazolium bromide (MTT) were purchased from Sigma–Aldrich (USA). Fetal bovine serum (FBS), a-minimum essential medium (α-MEM), and 1% penicillin−streptomycin solution were purchased from HyClone (USA) and used without further purification. The water was purified before use in a three stage Sartorius arium® pro purification system and has a resistivity higher than 18.2 MV cm^{-1}.

2.2. Alkaline treatment of AZ31 Mg alloy substrates

The AZ31 Mg alloy discs were ground with SiC emery paper down to a 1200-grit and then polished using 1 μm size alumina suspension. After ultrasonically cleaned in double distilled water, ethanol and acetone for 10 min, respectively, the discs were immersed in 100 mL of 0.5 M $NaHCO_3$ solution for 24 h, rinsed with double distilled water three times and dried in air.

2.3. Preparation of CAp@PDA nanoparticles

First, biomimetic CAp nanoparticles (with the obtained carbonate content of 3.7 wt.%) were synthesized via a hydrothermal route, as detailed in our previous work [32]. The hydrothermal reaction was conducted at pH = 10 and temperature of 200 °C for 24 h. The obtained CAp nanoparticle were dispersed into 2 mg/mL dopamine in a Tris-HCl buffer (10 mM, pH = 8.5) under ultrasonication for 30 min. The mixture was stirred vigorously for 12 h at 60° C, and then subjected to centrifugation (speed at 8000 rpm for 20 min) and finally washed with double distilled water and ethanol three times, respectively.

2.4. Preparation of Ag@PDA nanoparticles

Silver nanoparticles were prepared by the reaction between 20 mL $AgNO_3$ (1 mM) aqueous solution and 2 mL (40 mM) sodium citrate aqueous solution at 100 °C for 1 h under vigorous stirring. Silver nanoparticles were formed when the color of the solution changed into yellow-green. Then, the precipitates were collected by centrifugation (8000 rpm, 20 min) and washed with double distilled water three times. The Ag@PDA nanoparticles were prepared following the identical procedure with that of CAp@PDA.

2.5. Assembly of CAp@PDA and Ag@PDA on AZ31 magnesium alloy substrates

The alkaline pre-treated magnesium alloy discs were sequentially immersed into the following solutions: (1) 2 mg/mL dopamine solution for 24 h; (2) 4 mg/mL chitosan solution (pH = 5.0) for 15 min; (3) 4 mg/mL CAp@PDA suspension for 15 min; (4) 4 mg/mL chitosan solution (pH = 5.0) for 15 min; and (5) 2 mg/mL Ag@PDA solution. All the treated specimens were cleaned ultrasonically for 2 min with double distilled water before moving to the next step. Such chitosan-CAp@PDA-chitosan-Ag@PDA periodic unit can be repeated for the required number of cycles. The corresponding nano-multilayered coatings are denoted $(\text{chitosan-CAp@PDA-chitosan-Ag@PDA})_n$, where n is the number of cycles. Here, we use $n = 3$. For the antibacterial activity and biocompatibility evaluations, we also switched the order to assemble Ag@PDA and CAp@PDA layers. The coated sample with CAp@PDA as the outmost layer ($(\text{chitosan-Ag@PDA-chitosan-CAp@PDA})_3$) was also prepared as control.

2.6. Materials characterization

The size and morphology of CAp, CAp@PDA, Ag and Ag@PDA nanoparticles were examined by transmission electron microscopy (TEM) and high resolution TEM (FEI Tecnai F30). The Ag nanoparitcles were confirmed by UV–vis spectroscopy (Lambda 25, PerkinElmer) and their size distrution was measured by dynamic light scattering (DLS, Horiba Scientific) technique. The surface composition was analyzed by X-ray photoelectron spectroscopy (XPS, Kratos Axis Ultra DLD, Japan) using monochromatic Al Kα radiation. All binding energies were calibrated with reference to the C 1s hydrocarbon peak at 284.6 eV. Surface morphology of the coatings was examined by scanning electron microscopy (SEM, Zeiss Ultra 55). The cross-section of the multilayered coating was prepared by focused ion beam (FIB, FEI Helios 600i). To protect the original surface from Ga ions damage, a platium cap layer with thickness of ~1 μm was first deposited before milling the trench.

2.7. Static immersion in the simulated body fluid (SBF)

In vitro degradation and ions release were determined by immersing the mechanically polished AZ31 and coated AZ31 discs with Ag@PDA as the outmost layer into 5 mL of SBF at 37 °C for 14 days. The SBF was refreshed at each pre-defined intervals. The concentrations of Mg^{2+} and Ag^{+} ions in SBF were measured by inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7700X). The pH evolution was investigated by immersing the two groups of samples into 110 mL SBF (with sample surface area/volume of solution = 1:50) at 37 °C in a water bath for 14 days. During the immersion period, the pH of the SBF was monitored by a pH meter. For each group, three individual measurements were performed.

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