



Contents lists available at ScienceDirect

Journal of the Taiwan Institute of Chemical Engineers

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

Biocompatibility polyelectrolyte coating with water-enabled self-healing ability

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ARTICLE INFO

Article history:

Received 28 March 2018

Revised 5 June 2018

Accepted 8 June 2018

Available online xxx

Keywords:

Biocompatibility

Self-healing

Loading

LbL self-assembly

ABSTRACT

A biocompatibility polyelectrolyte coating capable of self-healing damages was developed via Layer-by-Layer self-assembly, a synthetic strategy that alternatively assembles layers of Chitosan (CS) and hyaluronic acid (HA). The reversible interactions of the hydrogen bonding formed between CS and HA can be dynamically associated/dissociated with water, imparting the CS/HA polyelectrolyte coating with substantially improved self-healing property and significantly decreasing micro-cracks and their associated concerns. Also, through structural changes induced during the freeze-drying and self-healing processes, not only can different kinds of drug molecules be easily loaded into CS/HA polyelectrolyte coating but also a variety of nanoparticles. These features provide the CS/HA polyelectrolyte coating with extend lifespan, enhanced reliability, and considerable promise for applying in novel functional biomedical materials.

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1. Introduction

Biocompatible polyelectrolyte coatings are a class of co-polymer made from readily available biomaterials [1]. These coatings have excellent biocompatibility and are hypoallergenic [2], as such, hold significant promise as functional coatings on medical implants and tissue scaffolds for patient care [3]. However, the deterioration and damage to the coating during application and use are dangerous [4]. The designation and preparation of durable functional coatings are sorely needed. Most recent strategies for enhancing the service time of polyelectrolyte coatings have focused on chemical polymerization and crosslinking [5,6], yet the special cross-linking agents in these composites dramatically limits their applications in biocompatible polyelectrolyte coatings [7].

An ideal biocompatible polyelectrolyte coating design would include self-healing properties similar to those seen in organisms across nature [8–10], which would effectively prolong the service

life of the coating on these biomedical materials and significantly improve their reliability [11–15]. Recent studies have demonstrated that a Layer-by-Layer (LbL) self-assembly technique might be particularly suitable for fabricating such self-healing polyelectrolyte coatings [16–18], as it can be driven by different types of molecular interactions [19–21], such as electrostatic interaction, π - π interaction, coordination interaction, hydrophilic/hydrophobic interaction, and, most importantly for the present study, hydrogen-bonding [22,23]. Over the past two decades, different styles of functional self-healing polyelectrolyte coatings have been prepared via LbL assembly technique [24–27]. bPEI/PAA functional self-healing polyelectrolyte coatings, for instance, have been designed and applied in many different fields [28–30]; however, the cytotoxicity of the PEI in these polyelectrolyte films can limit the applicability for biomedical applications [31]. Combining biocompatibilities and self-healing properties into a functional unit to construct stable biomedical materials with LbL self-assembly has not yet been reported and remains a challenge.

Herein, a highly efficient and technically simple approach is developed to efficiently fabricate Chitosan/Hyaluronic acid (CS/HA) polyelectrolyte coatings that are capable of self-healing damage. In addition, different kinds of medicines and specific targets can be easily loaded into the CS/HA polyelectrolyte coating during the

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<https://doi.org/10.1016/j.jtice.2018.06.009>

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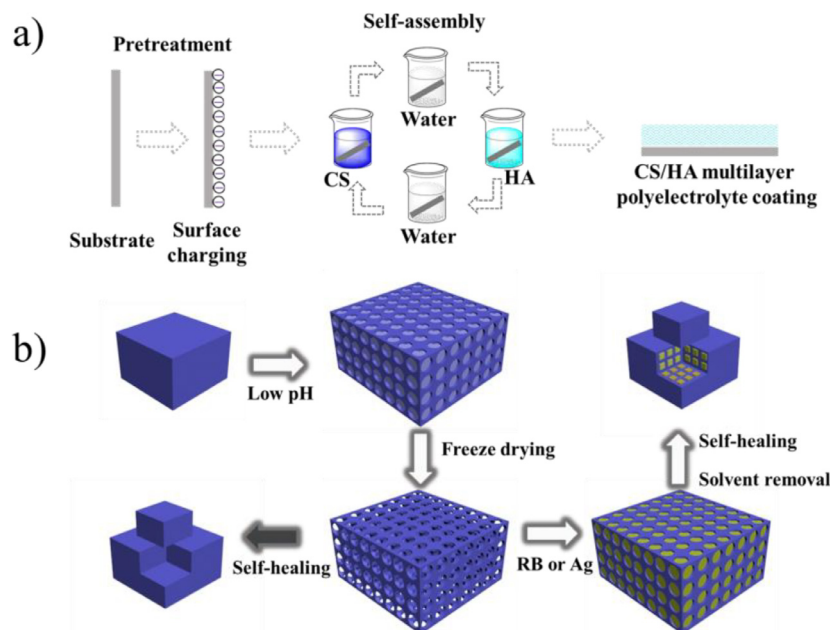


Fig. 1. (a) Schematic diagram of the synthetic process of the LBL assembly technique to fabricate the CS/HA polyelectrolyte coating; (b) fabrication of polyelectrolyte coating with microporous structures based on freeze-drying and integration of a specific target into the polyelectrolyte coating during self-healing.

structure changes induced during the free-drying and self-healing processes. These features extend the lifespan of CS/HA polyelectrolyte coating and make it a promising application for biomedical materials.

2. Experimentation

2.1. Chemicals and materials

Hyaluronic acid (HA, Mw \approx 403) and rhodamine B (RB) dye were purchased from Aladdin Industrial Corp., whereas Chitosan (CS, Mw \approx 375,000, with the degree of N-deacetylation at 85%), acetic acid (CH₃COOH), sodium hydroxide (NaOH), and silver nitrate (AgNO₃) were purchased from Sinopharm Chemical Reagent Co., Ltd. The concentration of the HA solution used for assembling the biomedical polyelectrolyte coating was 4 mg mL⁻¹ and the pH was maintained at 7.0; the concentration of the CS solution was the same as the HA solution, but the pH value of CS was kept at 3.0, the pH value of CH₃COOH was kept at 2.0. To obtain the negatively charged substrates, the used substrates in the experiment were all treated with plasma processing apparatus. The silver nanoparticles were synthesized using the UV irradiation method: firstly, 1.7 g AgNO₃ was added into 100 mL CS solution; the mixed solution was then exposed to UV light at room temperature; and, after 1 h of irradiation, the Ag nanoparticles were obtained.

2.2. Fabrication of the CS/HA and bPEI/PAA polyelectrolyte coating via LbL assembly

The CS/HA polyelectrolyte coatings were fabricated via LBL assembly (Fig. 1(a)). To summarize, the treated substrate was first immersed in the CS solution for 20 min and then the resulting substrate-CS was rinsed for 3 min to remove any excess CS solution. The substrate-CS was subsequently deposited in the HA solution for 20 min, after which the substrate-CS/HA*1 was then washed for 3 min to remove the excess HA solution. This sequence was repeated 50 times, with the expected CS and HA alternately deposited on the substrate. Following this, the substrate was removed and, the self-healing CS/HA polyelectrolyte coating was ob-

tained. The bPEI/PAA polyelectrolyte coatings were also fabricated via LBL assembly technique like the CS/HA polyelectrolyte coating just involves alternating assemble bPEI and PAA (see Fig. 2).

2.3. Integrating the RB dye and Ag nanoparticles

To load specific target compounds or particles, the self-healing CS/HA polyelectrolyte coating were immersed in an acid solution (pH 2.0) for 2 h. The polyelectrolyte coating, which absorbed some of solution, was then freeze-dried for 24 h to produce a polyelectrolyte coating with a more porous surface. The sample was then soaked in an RB or an Ag nanoparticle solution for 10 min to integrate RB or Ag nanoparticles. The RB-loaded or Ag-loaded CS/HA polyelectrolyte coatings were then set to dry at 60 °C. In the experiment, the CS/HA coatings were all deposited on the substrates.

2.4. Characterization

A Nicolet iS5 (Thermo Electron Scientific Instruments Corp.) was used to analyze the FT-IR spectrum of the CS/HA polyelectrolyte coating. A U-4100 spectrometer (Hitachi High-Tech Corp.) was used to examine the loading and release of the RB dye and Ag nanoparticles. To observe the self-healing process and surface behavior of the CS/HA polyelectrolyte coatings, we used a MVX10 Macroview Microscope (Olympus Corp.). Electrochemical impedance spectroscopy (EIS) was measured with a CHI 660d electrochemical analyzer system (CHI Instruments Inc.) to further investigate the self-healing process of the coatings. The CS/HA polyelectrolyte coating was used as working electrode, the counter electrode was a piece of platinum sheet and the reference electrode was an Ag/AgCl electrode. The experiment was carried out at open circuit potential with a superimposed 5 mV sinusoidal voltage in the frequency range from 10⁴ Hz to 10⁻² Hz.

The antibacterial effect of the CS/HA polyelectrolyte coating was determined by the sticking membrane method. Briefly, this method involved preparing a CS/HA and an Ag-loaded CS/HA polyelectrolyte coating (2 cm \times 2 cm), labeled (b) and (c), respectively. The polyelectrolyte coatings were placed on LB petri-dishes that were inoculated with *E. coli*. One LB petri-dish inoculated with bacteria

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