Characterization and antibacterial property of epsilon-poly-L-lysine grafted cellulose films

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Research Article

Keywords: Cellulose, Epsilon-poly-L-lysine, Film, Antibacterial activities

Posted Date: July 8th, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-680996/v1>

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Abstract

In order to obtain a stable, biodegradable and cost-effective antibacterial food package materials, this study developed a facile route to immobilize epsilon-poly-L-lysine (EPL) on films. Cellulose films were prepared via a sol-gel transition method and activated by epoxy chloropropane (ECH) to form epoxide group. EPL was covalently immobilized on the surface of the cellulose films (ECFs) by reacting the amino groups of the EPL molecules with the epoxy groups of the epoxidized cellulose films. The structure, morphology and properties of films were characterized by scanning electron microscope, Fourier transform infrared spectroscopy, X-ray diffraction, thermogravimetric analysis, universal material testing machine, ultraviolet-visible spectrophotometer and static water contact angle. The results showed that EPL was immobilized successfully in the cellulose films. ECFs exhibited 80% transmittance through 300 nm to 1100 nm and the excellent thermal degradation temperature of 240°C. Moreover, good antimicrobial activity against Gram-negative *Escherichia coli*, Gram-positive *Staphylococcus aureus* and *Alicyclobacillus acidoterrestris* were confirmed within 12 h, which is expected to provide a novel material for potential applications in the food industry.

Introduction

In recent years, the threat of harmful bacteria become one of the major issues that people are concerned. The common food-borne pathogens include *S. aureus* and *E. coli* which can transmit through food and water, cause many disease and death in developing countries (Koujima I, 1978; Neil K P et al., 2012). Meanwhile, *A. acidoterrestris* that are resistant to commercial pasteurization, as a kind of gram-positive acidophilic heat-resistant spoiling bacteria, is a major reason of huge economic losses in juice industry (Pornpukdeewattana S et al., 2020). Therefore, the quality controlling of food become a target that people have to comply.

Packaging has the function of preventing food biological, chemical, and physical external factors from being damaged and maintaining the stable quality of the food during the distribution process of food from the factory to the consumer. However, with the rapid development of synthetic plastic packaging materials (Tian H F et al., 2017), the hazards to the environment, ecosystems and human health have caused anxious due to its non-renewability, non-degradability and plasticizer migrated from the package (Cazon P et al., 2017; Wang S et al., 2016). Nowadays, more attention focus on the development of environment-friendly, renewable packaging materials by using natural polymers to be an alternative of conventional plastic packaging. Cellulose, the most abundant natural polymer on the earth (Yang Z et al., 2015), is a linear polymer polysaccharide with excellent properties such as renewability, availability, high mechanical strength, environmental friendliness, biocompatibility and biodegradability (Li B G et al., 2020; Liu L J et al., 2019). In order to overcome the problem of low solubility of cellulose in general solvents (Qin X Z et al., 2013), the alkali/urea system is proposed to dissolve cellulose for forming a transparent solution by driving hydrogen bonds of the small molecules such as alkali and urea in the solvent to self-assemble with cellulose macromolecules obtaining a worm-like inclusion compound at low temperature (Cai J and L Zhang, 2005). The glycosyl ring of cellulose has 3 hydroxyl groups, which

can undergo various reactions such as oxidation, etherification, crosslinking, esterification and graft copolymerization to realize the functional modification and application of cellulose (Guo L F et al., 2019; Kakibe T et al., 2019; Kim U J et al., 2019; Liu S L et al., 2021; Wang C P et al., 2020). Therefore, it could be widely used in biomedicine, food, cosmetics, textiles, papermaking and other field (Bianchet R T et al., 2020; Fotie G et al., 2020; Liu S J et al., 2020; Lourenco A F et al., 2019; Yang J S and J F Li, 2018), as different forms including film, bead, fiber, etc. (Orelma H et al., 2020; Wu H et al., 2018; Wu H et al., 2020).

Original regenerated cellulose films lack antimicrobial properties and are easily contaminated by microorganisms (Khattak S et al., 2019). As the incidence of foodborne diseases and the threat to social economy caused by microorganisms are increasing (Cui F C et al., 2020; Song Z H et al., 2019), it is critical to endow antibacterial properties for the cellulose films as food package materials. EPL that is approved as a food additive by the U.S. Food and Drug Administration in 2003 (Zahi M R et al., 2017), a homologous monomer polymer containing 25 ~ 35 L-lysine residues (Yoshida T and T Nagasawa, 2003), shows a highly effective broad-spectrum antibacterial activity and stability under both acidic and alkaline conditions (Zhang L M et al., 2015). Realizing the covalent immobilization of EPL with the matrix provides the possibility of novel antibacterial materials.

In this study, we have proposed new avenue to prepare EPL grafted cellulose films connected by the amino groups of EPL and the epoxide group on films with the antibacterial properties. The morphology, structure, thermodynamics, mechanical properties, optical transmittance and hydrophobicity of the films were characterized by scanning electron microscope (SEM), Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), thermogravimetric analysis (TGA), universal material testing machine, ultraviolet-visible spectrophotometer (UV-VIS) and static water contact angle (CA). The antibacterial properties against *E. coli* (ATCC 25922), *S. aureus* (ATCC 29213) and *A. acidoterrestris* (DSM 3922) are explored.

Materials And Methods

Materials

Cotton linter cellulose (α -cellulose $\geq 95\%$, viscosity average molecular weight ($M\eta$) approximately of 1.07×10^5) was purchased from Hubei Chemical Fiber Group Co., Ltd. (Xiangyang, China), Epsilon-poly-L-lysine ($M_w < 5000$) was purchased from Macklin Biochemical Co., Ltd. (Shanghai, China). $\text{LiOH} \cdot \text{H}_2\text{O}$ and Urea were purchased from Aladdin Biochemical Technology Co., Ltd. (Shanghai, China). Sodium hydroxide, Epoxy chloropropane and Anhydrous alcohol were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). All reagents were analytical grade and used without further purification.

A. acidoterrestris (DSM 3922) was purchased from the German Collection of Microorganisms. *S. aureus* (ATCC 29213) and *E. coli* (ATCC 25922) were purchased from the American Type Culture Collection.

Preparation of regenerated cellulose film (RCF)

The regenerated cellulose films were prepared according to the previous work with a minor modification (Wu H et al., 2018). Briefly, 4 g pure cellulose was placed in 100 g urea/alkali solution system ($\text{LiOH}\cdot\text{H}_2\text{O}/\text{urea}/\text{H}_2\text{O} = 8 \text{ wt\%/}15 \text{ wt\%/}77 \text{ wt\%}$), stirred violently and stored at -20°C overnight. After thawed at room temperature, the solution was stirred using mechanical agitation and centrifuged at 4000 rpm for 2 min to obtain a transparent cellulose solution at the concentration of 4 wt%. The viscous solution was cast on a glass plate and spread using a miniature scraping machine (SX-5000AX, Tianjin Boxue Machine Co. Ltd., China). Then, the glass plate with cellulose solution was immersed into anhydrous ethanol to regenerate for 3 min and obtained films. Subsequently, the films were washed and then dialyzed against distilled water for 2 days. The regenerated cellulose films were obtained by drying at ambient conditions, which was marked as RCF.

Preparation of EPL grafted cellulose films (ECFs)

100 ml cellulose solution was prepared by using the freeze-thaw method according to 2.2. Then, 5 mL ECH was added dropwise to the cellulose solution with mechanical stirring. The mixed solution was centrifuged at 4000 rpm for 2 min to remove air bubbles after continuously stirred for 0.5 h. The centrifugal solution was equally poured into molds (20 g per dish) and heated for 1 h at 60°C after capping. Subsequently, the solution was immersed into anhydrous ethanol to regenerate and form films. The films were washed and dialyzed for 2 days to remove ECH and other impurities. The cross-linked cellulose films were obtained by drying naturally at room temperature, which was marked as CCF.

5% (v/v) ECH was added to NaOH solution (1M), and the mixed solution was equally divided into four groups. EPL with different initial amount (0, 1, 5, 10 mg/ mL) were dissolved in the above solution and poured into the reaction vessel respectively. The cross-linked cellulose films that have not been dried were immersed reaction vessel and heated at 60°C for 2 h. EPL grafted cellulose films were obtained by drying at ambient conditions after washed thoroughly with deionized water and dialyzed for 2 days, which were marked as ECF-0, ECF-1, ECF-5, ECF-10, respectively.

Characterization

After the films were sputter coated with gold and fixed on the sample stage with conductive glue, the surface and cross-sectional topography of films were observed by an SEM (Sigma 500, Carl Zeiss, Germany) under an acceleration voltage of 20 kV. FTIR spectrometer (Vetex 70, Bruker Instruments Ltd., Germany) was used to determine the chemical structure of the films. The films were ground and pressed with potassium bromide (1:100) into pellet by the tableting method. The infrared spectrum was obtained by scanning 16 times in the range of $4000 \sim 400 \text{ cm}^{-1}$ with a resolution of 4 cm^{-1} . X-ray diffractometer (Ultima IV, Rigaku, Japan) was used to measure the crystal structure of the films and pure cellulose (PC) in the scattering range of $2\theta = 5^\circ \sim 80^\circ$ with Cu K α target ($\lambda = 0.15406 \text{ nm}$) at 40 kV and 40 mA. A thermogravimetric analyzer (TG209F3, NETZSCH, Germany) was used to study the thermal performance of films under nitrogen atmospheres in the temperature range of $30 \sim 800^\circ\text{C}$ at a heating rate of $10^\circ\text{C}/\text{min}$. Ultraviolet-visible spectrophotometer (UV2700, Shimadzu Corporation, Japan) was used to test

the optical transmittance of the films from 200 to 1100 nm using air as a blank controlled. The universal material testing machine (fatigue test system 8802, Instron, USA) were used to measure mechanical properties of the films at the clamping distance of 30 mm with a speed of 1mm/min. Contact angle tester (JY-PHb, Chengde Jinhe Instrument Co., Ltd., China) was used to test the surface hydrophobicity of the films. The films (2 cm · 2 cm) that were stuck on a glass slide were tested on the measuring table in the test solution of 10 μ L.

Antimicrobial activity

The antibacterial properties of the samples were determined via turbidimetric method according to previous work with some modification (Tavakolian M et al., 2018). *A. acidoterrestris* was cultured for 12 h at 45°C in *Alicyclobacillus* spp. medium (AAM) containing yeast extract (2.0 g/L), glucose (2.0 g/L), (NH₄)₂SO₄ (0.4 g/L), MgSO₄·7H₂O (1.0 g/L), CaCl₂ (0.38 g/L), KH₂PO₄ (1.2 g/L). *S. aureus* and *E. coli* was cultured in LB medium containing tryptone (10 g/L), yeast extract (5 g/L), NaCl (10 g/L) for 12 h at 37°C respectively. ECFs (6 cm·3 cm) were added in 50 mL the bacterial suspension (10⁴ CFU/mL), and incubated for another 12 h respectively. The OD₆₀₀ of each treatment was measured by an UV-VIS spectrophotometer (N600, Shanghai Yoke Instrument Co., Ltd, China) every 2 h.

Results And Discussion

Fabrication of EPL grafted cellulose films

The schematic pathway for formation of EPL grafted cellulose films is displayed in Fig. 1. The cellulose was dissolved in the urea/alkali solution system by the freeze-thaw method. The ECH was added dropwise to the cellulose solution under stirring, then poured into the mold and heated to promote cross-linking of cellulose molecules. After regeneration using anhydrous alcohol, the cellulose films were undergone nucleophilic reaction via hydroxyl groups on the cellulose with ECH, and covalent immobilization by the amino groups on the EPL with epoxide groups of epoxidized cellulose films. Finally, ECFs were obtained by washing, dialysis and drying at ambient conditions.

Morphology analysis

The optical transmittance spectra of RCF, CCF and ECFs is displayed in Fig. 3. Films all showed good optically transparent. The transmittances of RCF were 87.8%, while for the CCF and ECFs, it ranged from 87.1–84.6% at 580 nm. This might result from the process of activated treatment and grafted EPL had an effect on the microstructure of the membrane surface, which could be shown in SEM (Wu H et al., 2018). The optical transmittance of ECFs was higher than 80% in the visible wavelength range of 400-760nm, and was expected to broad the application of antibacterial films in the packaging market.

FTIR analysis

FTIR spectra of RCF, CCF and ECFs are showed in Fig. 4. In all films, the broad peak at 3400 cm^{-1} was caused by the stretching vibration of the hydroxyl group (Yan J A et al., 2015). The peaks at 2926 cm^{-1} and 2860 cm^{-1} were related to asymmetric and symmetric CH_2 stretching (Huang H L et al., 2016). For CCF and ECFs, the absorption peak at 1378 cm^{-1} and 1321 cm^{-1} were ascribed to the C-O-C band produced by cellulose with ECH, and C-C band respectively (Salleh K M et al., 2019). In the case of ECFs, the characteristic bands at 1631 cm^{-1} and 1060 cm^{-1} were exhibited, which were assigned to $-\text{NH}_2$ stretching and the bending vibration of C-N respectively (Yang B et al., 2019). The results demonstrated that the cellulose molecules were cross-linked through ECH, and EPL was installed into cellulose matrix via covalent coupling reactions between amine groups of EPL and epoxide group of epoxidized films.

XRD analysis

The XRD patterns of PC, RCF, CCF and ECF-10 are displayed in Fig. 5. The diffraction peaks of PC at $2\theta = 14^\circ, 16^\circ, 23^\circ$ and 34.5° were correspond to the (1-10), (110), (200) and (004) planes of typical cellulose I crystal (French A D, 2014). The peaks of RCF at $2\theta = 12^\circ, 20^\circ$ and 22° were attributed to the (1-10), (110) and (020) crystal planes of typical cellulose II (He M et al., 2018), which showed a typical conversion from cellulose I to cellulose II as a result of dissolution and regeneration of cellulose (Cai J and L Zhang, 2005). Compared with RCF, the peak intensity increased for CCF and ECFs, indicative of increased crystallinity degrees. We could find similar conclusions in the literature after the crosslinking effect of ECH on cellulose molecular (Fei G et al., 2019).

TGA analysis

Performance analysis

The hydrophobicity and mechanical property of RCF, CCF and ECFs were measured in Fig. 7. Commonly, the more hydrophilic a material is, the lower its CA value (Song H Z and L W Zheng, 2013), CA of RCF was 49.8° , exhibiting as hydrophilicity due to the cellulose contained a large number of hydroxyl functional groups. In CCF and ECF-0, CA was 68.6° and 68.9° respectively, which was probably resulted from the ECH combined with the hydroxyl sites of the cellulose. CA of ECFs had increased from 82.7° to 106.9° with increasing EPL contents, which exhibited as outstanding hydrophobicity. The reason may lie in EPL contained a large number of hydrophobic alkyl groups. Good hydrophobicity will help to improve the water vapor barrier properties of the film.

It can be seen from Fig. 7(B) that the elongation at break values (E) of RCF were lower than the films with EPL. Meanwhile, the E of grafted cellulose films with 0 mg/ mL, 1 mg/ mL, 5 mg/mL was lower than those of the films with 10 mg/ mL, indicating the E values of the grafted films increased with the increase in the EPL content. The reason may be explained in two aspects: the hydrogen bond force of cellulose was weakened when the modification happened, and EPL cross-linked cellulose films absorbed water and swelled in the process of dialysis, in which water molecules combined with the polar hydrophilic -OH of the films to reduce the interaction of cellulose molecules (Peng N et al., 2016).

Conclusions

In this work, a series of EPL grafted cellulose films with different grafting amount were prepared. The morphology, structure, thermodynamics, mechanical properties, optical transmittance, water-resistance and antibacterial properties of the grafted film materials were studied. The results showed that the ECFs had good optical transmittance, thermal stability, hydrophobicity and surface compactness. ECFs at a certain grafting amount were resistant to *A. acidoterrestris*, *S. aureus* and *E. coli*. ECFs combined the outstanding advantage of cellulose and EPL, which provided a new choice for the industrialization and marketization of antibacterial films.

Declarations

Acknowledgments

This work was financially supported by the project of the National Natural Science Foundation of China (NSFC) (NO.31501436); The Fundamental Research Funds for the Central Universities (Z1090220122); Key Research and Development Program of Shaanxi Province, China (No. S2020-YF-YBNY-0142); Natural Science Foundation of Jiangsu Province (BK20201480), the Open Research Fund Program of Beijing Key Laboratory of Quality Evaluation Technology for Hygiene and Safety of Plastics, Beijing Technology and Business University, Beijing 100048, China (QETHSP 2020008) and the Fund for Enterprise Technological Innovation in Haixi Prefecture of Qinghai Province (2018-N-502), the Open Project of Key Laboratory of Green Chemical Process of Ministry of Education (GCP20200206), Hubei Key Laboratory of Novel Reactor and Green Chemical Technology (Wuhan Institute of Technology) (K202002).

Conflict of interests

The authors declare that they have no conflict of interest.

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Figures

Figure 1

Schematic diagram for the preparation and mechanism of ECFs.

Figure 2

SEM images of RCF (a-d), ECF-1 (e-h) and ECF-10 (i-l).

Figure 3

The optical transmittance spectra of the films.

Figure 4

FTIR spectra of RCF, CCF and ECFs.

Figure 5

XRD patterns of PC, RCF, CCF and ECF-10.

Figure 6

TGA (a, b) and DTG (c, d) curves of PC, RCF, CCF and ECFs.

Figure 7

Water static contact angles (CA) of RCF (a), CCF (b), ECF-0 (c), ECF-1 (d), ECF-5 (e) and ECF-10 (f).

Figure 8

Antibacterial activities of RC, CCF and ECFs treatment.