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Preparation of Thai Silk Sericin/Poly (Vinyl Alcohol) Hybrid Films for Drug Delivery Control: Study on Amoxicillin Trihydrate

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ABSTRACT

Silk is a natural polymer, produced by silkworm *Bombyx mori*. Silk fibers are composed of sericin and fibroin protein. Sericin has unique properties in biomedical applications. This study aims to prepare sericin/Poly (vinyl alcohol) hybrid films by a solvent evaporation method for loading amoxicillin trihydrate. The sericin/Poly(vinyl alcohol) solution was mixed with amoxicillin trihydrate and placed on the 3.5 x 5 cm^2 polyethylene molds before drying in the oven to obtain hybrid films. The results showed that, sericin/ Poly(vinyl alcohol) without amoxicillin trihydrate was flexible and transparent, but the sericin/ Poly(vinyl alcohol) film loading with amoxicillin trihydrate was opaque and the film was not homogeneous. The structure of films was investigated by Fourier transform infrared (FT-IR) spectrometer. The releasing rate of amoxicillin trihydrate loaded in the film was high.

Key words: Thai Silk, Sericin, Amoxicillin trihydrate

INTRODUCTION

Silk has been hailed as the "Queen of Textiles". The best quality raw silk is obtained from silk worm, *Bombyx mori*, which feeds on mulberry leaves as food. The cocoons of mulberry silkworm *Bombyx mori* consist of two main proteins: sericin and fibroin. Fibroin is the core fibers which are coated with gummy layer of sericin. In silk textile processing, sericin is usually extracted from silk cocoon during the degumming process. The extracted sericin is considered as the waste of silk industry. Nowadays, sericin has been applied in various fields such as biomaterials, biomedical and pharmaceutical industries [1] due to their unique properties, including non-toxicity, biodegradability and biocompatibility[2]. Sericin is mainly consisted of serine and aspartic acid with strong polar side chain, so it can be blended with other polymers such as gelatin [3], collagen [4], polyacrylamide [5], and poly (vinyl alcohol) [6] to improve properties.

Poly (vinyl alcohol) (PVA) is a synthetic polymer which is obtained by the vinyl acetate polymerization in alcoholic solution followed by partial hydrolysis [7]. PVA shows biocompatibility with human body, thus it is applied to use for controlled delivery system of drugs, contact lens, artificial skins and biosensor.

In this research, we prepared the Thai silk sericin/poly (vinyl alcohol) hybrid film loaded amoxicillin trihydrate and then studied the releasing rate of amoxicillin trihydrate. The goal of this study is to evaluate the Thai silk sericin/poly (vinyl alcohol) hybrid film as a biomaterial for loading some medical drugs to apply in delivery system.

Materials and Methods

Silk sericin/PVA hybrid films preparation:

Silk cocoons were supplied from Nan Thailand. The overall preparation of film is shown in Fig 1. For clarity, the cocoons were cut into small pieces. The 2 g of cocoons were mixed with 20 ml of deionized water as a sample. The sample was boiled at 100^{o}C for 30 min. The solution was filtered by filter paper. The mixture of silk sericin/PVA solution at concentration of 0.45%w/v was prepared. The 4 ml of the mixture solution was poured in the 3.5×5 cm^2 molds. The molds were left at 50^{o}C in an oven for 3 days. The silk sericin/PVA film loaded with amoxicillin trihydrate was prepared by mixing 100

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mg of amoxicillin trihydrate in the 20 ml of silk sericin/PVA solution. The solution was molded into a film with the same condition of silk sericin/PVA hybrid film.

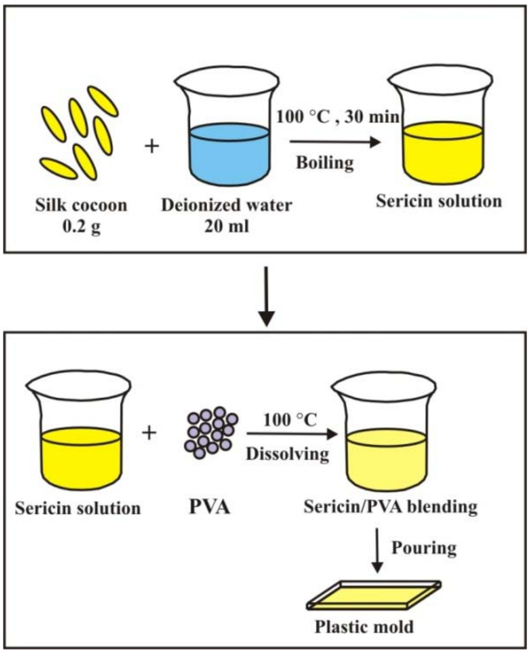

Fig. 1: Schematic of the extraction of sericin solution and the preparation of sericin/PVA hybrid film

Contact angle measurement:

Static contact angle of PVA film and silk sericin/PVA hybrid film were performed on dry films at room temperature using contact angle meter (Kyowa, DM-CE2). Distilled water droplets were used with a drop volume of approximately of 0.5 μL.

Structure characterization:

The structure of silk cocoon, PVA film, silk sericin/PVA hybrid film, silk sericin/PVA loaded with amoxicillin trihydrate films were analyzed by FTIR spectroscopy (Thermoscientific, Nicolet 6700) in the spectral region of 400 – 2000 cm^{-1}.

Drug release test:

The 2.5 × 3 cm^2 silk serici/PVA loaded with amoxicillin trihydrate film was immersed in a 20 ml of deionized water at room temperature. The absorbance at 272 nm of aqueous solution was measured. The amount of amoxicillin trihydrate was calculated by the absorbance according to the standard curve.

Results and Discussion

The prepared Thai silk sericin/ PVA hybrid film was smooth, transparent and flexible but the sericin/ PVA film loaded with amoxicillin trihydrate was opaque and the film was not homogeneous as shown in Fig. 2.

Fig. 2: sericin/PVA hybrid film (left) and sericin/ PVA film loaded with amoxicillin trihydrate (righ)

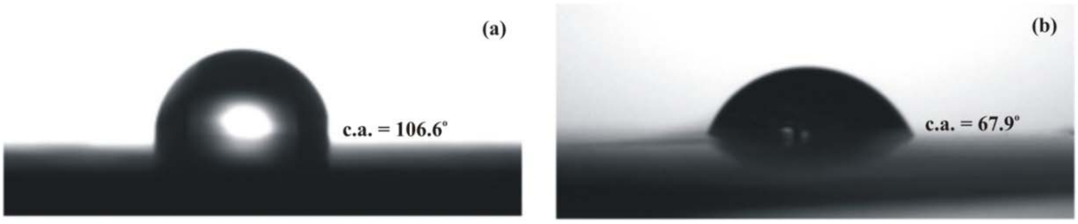

Fig. 3: Photographs of contact angle for (a) PVA film and (b) sericin/PVA hybrid film

From fig. 3, we found that the water contact angle of PVA film was about 106.6^o , while it was 67.9^o for sericin/PVA hybrid film. This phenomenon is due to sericin that has a highly hydrophilic macromolecular protein with strong polar side groups [6]. The surface of PVA film can be modified with a great number of hydrophilic group from sericin. The feature indicates that the sericin/PVA hybrid film can be used to release the drug.

FTIR spectroscopy was used to investigate the chemical structure of the films. Fig.4 shows the FTIR spectra of the prepared films. The peak positions of amide I (C=O stretching), amide II (N-H deformation), and amide III (C-N stretching) were determined for protein conformation. As shown in Fig. 4, amide I, amide II, and amide III exhibited at around 1632, 1520 and 1231 cm^{-1}. The amide I, amide II, and amide III bands in sericin/PVA hybrid film and sericin/ PVA film loaded with amoxicillin trihydrate have a reduced intensity. This result indicates that PVA affects on the bonding of amide group. Furthermore, the absorption peaks of sericin/PVA hybrid film and sericin/ PVA film loaded with amoxicillin trihydrate show specifically band of PVA and amoxicillin trihydrate. This result indicates that the chemical bonding between sericin, PVA and amoxicillin trihydrate are formed.

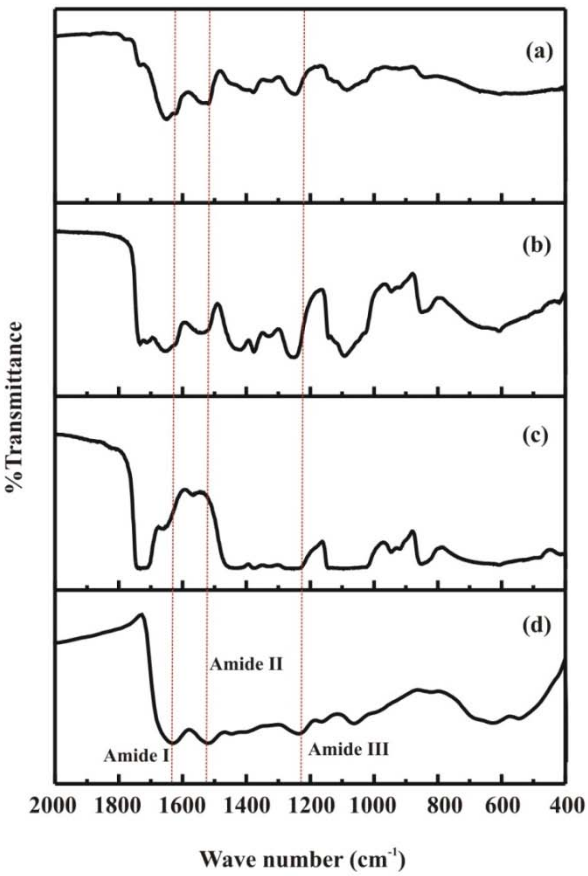

Fig. 4: FTIR spectra of (a) silk cocoon, (b) PVA film, (c) sericin/PVA hybrid film and (d) sericin/ PVA film loaded with amoxicillin trihydrate

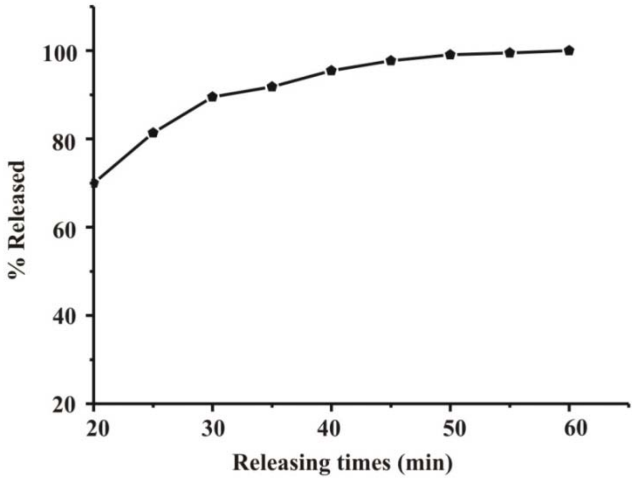

Fig. 5: Releasing of amoxicillin trihydrate from sericin/PVA hybrid film

From fig.5, amoxicillin trihydrate was rapidly released from sericin/PVA hybrid film. It is noteworthy that drug can be released from the film up to 70% within 20 minutes. The reason for this may be due to the good water solubility of sericin and PVA.

Conclusion:

Sericin/PVA hybrid film and sericin/ PVA film loaded with amoxicillin trihydrate could be prepared by molding method. A mixture of sericin and PVA causes the changes of chemical structure and contact angle of the film. A decrease in a contact angle of the film leads to sustain releasing of amoxicillin trihydrate from the film. The result of this study, drug release is preliminary and will be required further study and development.

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