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Chitosan and gelatin based biodegradable packaging films with UV-light protection

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ABSTRACT

Biopolymers are polymers obtained from biological origins and used for various biological and industrial applications. A biopolymer should be non-toxic, non-antigenic, non-irritant, non-carcinogenic, sterilisable and adequately available for their widespread applications. In this study, chitosan (CS) and gelatin (GL) based films were prepared to be used as biodegradable packaging films. CS was blended with GL to improve various physicochemical properties. The blended CSGL films were crosslinked with boric acid (BA) to improve various properties viz. light barrier properties, Water Vapour Permeability (WVP), moisture content (%), Total Solubility Matter (TSM), most important to improve the strength. The studies of transparency, X-ray Diffraction (XRD), Scanning Electron Microscopy (SEM), and optical microscopy confirms that the synthesized films were found to be transparent and homogenous indicating good compatibility among different components. The synthesized CS and GL based films showed UV-light barrier properties as supported by data. The tensile strength of films increases, decreases water solubility, moisture content (%) and WVP on crosslinking. In order to make the crosslinked films more flexible, Polyethylene glycol was used as plasticizer, making the films more flexible and transparent. This study indicates that these biodegradable CS and GL based films are potent to be used as packing films.

1. Introduction

Biopolymers are polymers obtained from biological origins and used for various biological and industrial applications. A biopolymer should be non-toxic, non-antigenic, non-irritant, non-carcinogenic, sterilisable and adequately available for their widespread applications [1]. Chitosan (CS) and gelatin (GL) both the natural polymers appear to offer most of these features to serve in various applications. Both of these are non-toxic, biodegradable and biocompatible polymers based on natural and renewable resources, used as coatings and films in food and various other applications [2].

CS is an unbranched polymer obtained from deacetylation of chitin, with inherent antimicrobial character, carries a positive charge and is soluble at pH below 6.5 [3–5]. Due to its cationic nature, it establish the electrostatic interactions with other compounds; hence, CS has been most widely used for fabrication of films with adequate barrier to water vapour. However, it seems economically feasible to combine it with other film forming biopolymers like GL/PVA, etc. considering the cost of its preparation [6]. Blending of two different biopolymers can intensely change the physico-chemical properties of film-forming solutions (FFS), thus affecting the functionality of the resulting films. Depending on their chemical structure, pH, molecular weight and hydration behaviour among others, all these changes occur due to the compatibility/incompatibility between both polymers [7].

GL is also one of biopolymers used in medical pharmaceutical applications with a good film forming ability used in production of biocompatible and biodegradable biomaterials, which can act as temporary replacements [8]. GL can readily absorb UV-light due to presence of aromatic amino acid in it. Several strategies have been employed to improve the physical performance of CS films; including chemical [9–11] or enzymatic treatment [12] and mixing with other polymers like polyethylene glycol (PEG) [13,14], polyvinyl alcohol (PVA) [15,16], starch [17], polyethylene glycol fumarate [8] and gelatin [18].

Furthermore, cross-linking has appeared as another important technique to improve the performance of CS/GL films. The cross-linkers like borate, tripolyphosphate [19], glycerol [20], sodium trimetaphosphate [21], and glutaraldehyde [22] can form cross-linked network structure with CS and GL chain by non-covalent or covalent interactions. However, an organic integrity of the merits of those developed cross-linking techniques remains necessarily not only for designing CS/GL blends with enhanced stability and strength, but also to expand their practical applications in different areas. In order to improve flexibility and processability of CS/GL films different types of plasticizers have been used into a polymeric material. The improvement in the flexibility of the polymer is there by increasing its intermolecular separation and lowering its glass transition temperature [23].

However, traditional methods of crosslinking suffer from several shortcomings such as lack of precise control over crosslinking, toxicity

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Fig. 1. Digital images of the synthesized transparent crosslinked films.

of the cross linkers, etc. This applies particularly to formaldehyde which in the presence of hydrochloric acid can form chloromethyl ether, the use of which is highly restricted as it possess a significant hazards [24]. Gluteraldehyde is considered as the most important crosslinker in the sense as it increases the strength of polymers, but studies reveals that it is carcinogenic in nature. Therefore, there is a necessity to use eco-friendly crosslinker which inhibits the lethal effect. In the present study, we tried to use a crosslinker which is biocompatible and safe to use. Here, crosslinking of CS and GL blend film with boric acid (BA) and the effect with its different concentration as crosslinker was investigated. BA is a non-volatile mineral and shows low-toxicity, fungicidal effect and antimicrobial properties. BA and its salts, have been used in medicine as a bactericide, a fungicide, and an antiseptic since the 1860s [25]. The objective of this study was to synthesized biodegradable packaging films with good transparency and flexibility with enhanced UV-light protection.

2. Materials and Methods

2.1. Materials

All the chemicals used were of analytical grade and were used as purchased without further purification. CS from shrimp shells (Degree of Deacetylation ≥75%) was purchased from HiMedia Laboratories, Mumbai. GL powder and BA (extra pure) were purchased from LobaChemie, Mumbai. PEG (mol. wt. 1400) was purchased from Thomas Baker Chemicals, Mumbai.

2.2. Preparation of Blend Films

Different ratios of solute and solvent were tried in order to obtain an ideal film of the two individual polymers separately as well as of blend. To prepare film forming solutions, GL was dissolved in distilled water whereas the CS was dissolved in 2% acetic acid (AA) solution. These solutions were kept on magnetic stirrer and stirred for 24 h at ambient conditions. The 10%(w/v) of GL and 2%(w/v) of CS solution was found to be suitable for synthesis of films and was optimized for fabrication of films. The resulting solutions were filtered by using cheese cloth. In order to form blend, both the solutions were mixed in different proportions, CS:GL (50:50, 60:40, 70:30, 80:20, 90:10 and 95:5), and was then allowed to settle down. The films were fabricated by pouring these FFS solutions with different ratio into petridishes further followed by evaporation of solvent at room temperature. The films were then peeled off from petridishes and kept in a desiccator for further use. The ideal films were then chosen for further investigations after examining their general morphology. The blend film having the concentration of CS:GL(90:10) was optimized to be used for further studies.

Table 1
Film thickness and apparent density of synthesized films.

Films	Film thickness(mm)	Apparent density(gm/cm^3)
CS	0.12	0.675
CSGL	0.15	1.744
CSGL2BA	0.13	1.459
CSGL3BA	0.12	1.554
CSGL4BA	0.13	1.582
CSGL5BA	0.14	1.56

2.3. Preparation of Crosslinked and Plasticized Blend Films

2.3.1. Optimization of Concentration of Crosslinker

Solutions of blend (2% CS (w/v) and 10% GL (w/v)) were mixed in ratio of 90:10 (CS:GL). On stirring solutions, BA was added slowly to blend solution. BA was used as crosslinker for the enhancement of physical and mechanical stabilities of the films. For this purpose a variation in the concentration of BA (2–5%) as cross-linker was done. These FFS were incubated at 30 °C for 24 h with gentle stirring using magnetic stirrer. These solutions were then filtered using cheese cloth, allowed to settle down and were then poured into petridishes for casting. The films so formed were peeled off from petridishes and kept in desiccator for further studies.

2.3.2. Optimization of Concentration of Plasticizer

The crosslinked bio-polymeric films achieves a good mechanical strength but lacks flexibility which is one of the important basic requirement of any polymer to be modified as films for their respective applications. Hence, PEG as plasticizer was subsequently added with varying concentration (5%, 10%, and 20%) to achieve desired flexibilities to the films. The solutions were then vigorously stirred for 24 h followed by the film formation, with same procedure as mentioned earlier.

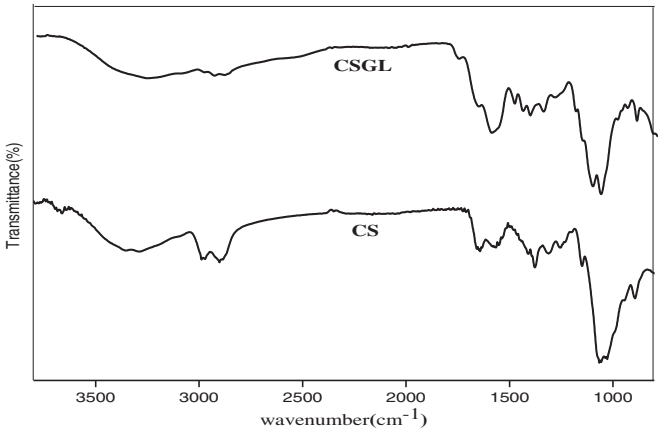

Fig. 2. FTIR spectra of CS and CSGL films.