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ORIGINAL ARTICLE

Poly (vinyl alcohol)-alginate physically crosslinked hydrogel membranes for wound dressing applications: Characterization and bio-evaluation

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Abstract PVA-sodium alginate (SA) hydrogel membranes containing sodium ampicillin as a topical antibiotic were developed using the freeze–thawing method for wound dressing application. Aqueous solution of sodium alginate has been blended in a certain ratio with PVA, followed by the crosslinking method has been conducted by freeze–thawing method as physical crosslinking instead of the use of traditional chemical crosslinking to avoid riskiness of chemical reagents and crosslinkers. The physicochemical properties of PVA-SA membranes e.g. gel fraction and water uptake % have been performed. Increased SA content with PVA decreased gel fraction, elasticity, and elongation to break of PVA-SA membranes. However, it resulted in an increase in swelling degree, protein adsorption, and roughness of membrane surface. High SA content in PVA membranes had apparently an impact on surface morphology structure of hydrogel membranes. Pore size and pore area distribution have been observed with addition of high SA concentration. However, high SA content had an insignificant effect on the release of ampicillin. The hydrolytic degradation of PVA-SA membranes has prominently increased with increasing SA content. Furthermore, hemolysis (%) and *in vitro* inhibition (%) for both Gram positive and negative bacteria have been sharply affected by addition of SA into PVA, indicating the improved blood hemocompatibility.

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Thus, PVA-SA hydrogel membrane based wound dressing system containing ampicillin could be a good polymeric membrane candidate in wound care.

1. Introduction

The typical idea of wound dressing maintenance is based on the presence of a moist environment around the wound which absorbs the exudates from the wound surface. Therefore, complete and partial wounds showed an apparent increase in re-epithelialization rates when they were maintained in a moist local environment (Hinman and Maibach, 1963; Winter, 1962). Accordingly, hydrogels are three-dimensional crosslinked hydrophilic polymers with a very high intrinsic content of water, which can provide a moist or wetted environment to the wound area and absorb the exudates. From this principle, hydrogels have been chosen as good candidate for wound dressing materials. Many hydrogels are prepared by physical or chemical crosslinking methods, (Hassan and Stewart, 2000; Razzak et al., 2001; Yang et al., 2004) while the physical crosslinking method such as repeated freeze–thawing cycles is regarded the proper crosslinking method and has been explored for biomedical and pharmaceutical applications due to its non-toxicity, solvent free, and biocompatibility. Furthermore, the obtained gel from this method has feasible physical properties e.g. rubbery nature and high water uptake compared to that obtained by chemical crosslinking (Peppas and Stauffer, 1991). Polyvinyl alcohol hydrogels have been prepared with a freeze–thawing method for wound dressing application because of their derisible clinical features. Additionally, PVA possesses desirable properties such as non-toxicity, biocompatibility, high hydrophilicity, easier film forming ability, chemical resistance, and mechanical resistance (Kim et al., 2008). Alginate is derived from brown algae, is an anionic linear polysaccharide composed of 1,4-linked β-D-mannuronic acid residue and 1,4-linked α-L-guluronic acid residue varying properties (Ress and Welsh, 1997). Alginate polymer has high hydrophilicity, biocompatible and relatively economical use, it has been widely used in biomedical applications e.g. wound dressing, (Kim et al., 2008) scaffolds, (Zmora et al., 2002) and dental or surgical impression materials (Nandini et al., 2008). The importance of the polymer blending with PVA, has been increased due to the blended polymeric materials which have desired properties, improve physical properties, low cost of basic polymer materials, improved process ability of film formation, and biologically acceptable. Accordingly, some blended polymers have been utilized with PVA to improve the clinical properties of obtained polymeric membranes for wound dressing applications, such as alginate (Kim et al., 2008; Levic et al., 2011), dextran (Fathi et al., 2011; Hwang et al., 2010), and chitosan (Kanatt et al., 2012; Yang et al., 2008). The obtained hydrogel film can be used to cleanse a big area from secreting lesions depending on the idea of high absorption of strong hydrophilicity of PVA-SA hydrogels, which limit wound secretions and reduce bacterial contamination (Kim et al., 2008). Meanwhile, PVA-SA hydrogel blend combines advantages of both; PVA hydrogels alone possess high mechanical properties, while alginates advantage sufficient physical and biological properties. Thus, sodium alginate has been chosen and is suitable for this purpose due to its high hydrophilicity, high protein adsorption ability, and good biological properties. Additionally, alginate fiber film can catch in a wound and is an easily biodegradable material without unexpected behavior (Gilchrist and Martin, 1983).

In this study, crosslinked PVA-alginate (PVA-SA) hydrogel films, loaded with sodium ampicillin as an antibiotic model, were prepared by the freeze–thawing cycle method to avoid the common harms which are arising from the traditional chemical crosslinking. PVA-SA hydrogels formed a matrix of physically crosslinked polymeric chains containing uncrosslinked polymers, water, and sodium ampicillin. Properties of hydrogel films such as gel fraction, swelling behavior, mechanical, morphology, and roughness, in addition, release studies of ampicillin from the prepared films were investigated. Finally, bio-evaluation studies of essential wound dressing characters like protein adsorption, hemocompatibility, hydrolytic degradation and antibacterial activity behavior were investigated under *in vitro* conditions.

2. Experimental

2.1. Materials

PVA (typically average M_w = 72,000 g/mol; 98.9% hydrolyzed) was obtained from Biochemica, Germany. SA (medium viscosity, average M_w = 130,000 g/mol as determined by GPC), albumin from bovine serum (BSA, fraction V, minimum 96% electrophoresis, nitrogen content 16.2%), and ampicillin sodium salt were purchased from Sigma–Aldrich Chemie GmbH, Steinheim, Germany. Folin and Ciocalteu's phenol reagent (FC, 2 N with respect to acid), were purchased from Park Scientific Limited, Northampton, UK. Distilled water was used throughout this research. All other chemicals were used without any further purification.

2.2. Preparation of PVA-SA hydrogels

PVA-SA hydrogel membranes were prepared by freezing–thawing (F–T) cycle according to the reported procedure of (Peppas and Stauffer, 1991). Briefly, aqueous solution containing 10% (w/v) PVA and 1.5% (w/v) SA and 20 mg of sodium ampicillin sodium salt were carefully dissolved in de-ionized water. Different proportions of PVA and SA content (0%, 25%, 33%, 50%, 65%, and 75%) solutions were mixed, sonicating, and vortexing for three hours. Proper amounts of this mixture were poured in Petri dishes, followed by freezing at −20 °C for 18 h and thawing for 6 h at 25 °C for three continuous cycles, to provide mechanically acceptable hydrogels for further experiments. The obtained PVA-SA hydrogel membranes were frozen in liquid nitrogen for 10 min before being lyophilized fractures for SEM investigations. All samples were left in deionized water for 72 h to extract leachable sol fraction or unconnected HES from polymer matrix for further characterizations.