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Eco-friendly and mussel-inspired hydrogels and glues from soy proteins and natural polyphenols with marked water-resistance and favourable antibacterial and biocompatibility profile for wound treatment applications

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PURPOSE OF THE ABSTRACT

Soy proteins isolates (SPI), the main industrial waste in soybean processing, are of particular interest for implementation of natural adhesives since they are cost-effective and easy to handle. Yet, they show low strength and poor water resistance. Many efficient adhesion systems are found in Nature, notably mussel byssus proteins whose extraordinary wet adhesion properties result from interaction of the catechol system of the abundant 3,4-dihydoxyphenylalanine residues with lysine amino groups giving rise to cross-linked networks. Inspired by this system, the present work was aimed at improving the adhesive properties of soy proteins by use of natural polyphenols, particularly catechols.

Initially, it was shown that SPI thermal denaturation is critical to get adhesiveness as evaluated by a simple test involving application of the adhesive on wood specimens that are then pressed together and soaked in water. Caffeic acid (CA) and its methyl ester, gallic acid and chlorogenic acid (CGA) were evaluated as additives. SPI was dissolved in water at 10% w/w and taken at 85°C for 1 h, then the polyphenol was added at 28 mM and the resulting mixture taken to 50°C and to pH 9. These conditions were expected to favor oxidation of o-diphenols to the corresponding quinones that can be entrapped by nucleophilic residues of the protein with formation of a network [1]. Development of a green chromophore typical of benzacridines from oxidative coupling of CGA with lysine provided evidence for an effective interaction for SPI/CGA [2]. (figure left panel)

Water resistance of wood specimens glued with the SPI/polyphenol proved higher (> 22 days for SPI/CGA) compared to that obtained using SPI alone (1 day). In addition, shear strength of SPI/polyphenol wood specimens was higher than 2.07±0.70 kgcm-2 obtained for SPI/urea previously described [3] with values up to 3.21±0.19 kgcm-2 in the case of SPI/CA. When chicken skin specimens were glued using SPI/CGA a water resistance up to 7 days was observed.

Determination of the amount of free SH and NH2 groups on the final material as lyophilized powder by Ellman's reagent and o-phthalaldehyde (OPA) reagent assay indicated the substantial involvement of these groups in the formation of cross-linking between polyphenols and SPI. On this basis it can be concluded that a covalent interaction between phenols, and polymers derived from their oxidation, with SPI occurs and this results in the formation of a lattice structure that confers the adhesive feature to the glue (figure, middle panel)

To further expand the potential of these materials for biomedical applications, an agarose/SPI hydrogel was prepared starting from water solution of agarose and adding thermally denatured SPI at 1:2 w/w ratio. Then the obtained solution was cooled in a Petri dish and immersed in a 10 mM water solution at pH 9 of the appropriate

polyphenol. The resulting hydrogel obtained from CGA with agarose/SPI showed a green color, due to the formation of benzacridine systems, indicating the reaction of CGA with lysine residues of proteins present inside the preformed gel. (figure, right panel). The cytocompatibility of these gels proved good both on keratinocytes and fibroblasts as was their hemocompatibility. The hydrogels exhibited a marked antimicrobial activity against gram-positive (S. aureus ATCC 12600, methicillin-resistant S. aureus (MRSA) WKZ-2, and S. epidermidis ATCC 35984), but much lower against gram negative bacteria in the contact and diffusion tests.

In conclusion, the good underwater adhesive properties of the materials prepared from SPI and natural catechols under sustainable conditions make them promising candidates for the development of surgical glues and wound dressing devices.

FIGURES

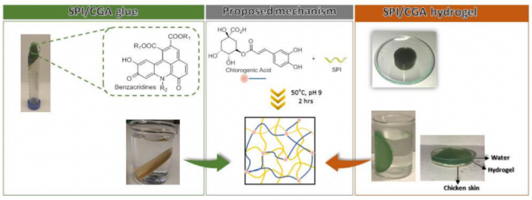

FIGURE 1

figure

Preparation and water resistance of glue and hydrogel from soy protein and chlorogenic acid

FIGURE 2

KEYWORDS

underwater adhesion | soybean processing waste | polyphenols | glues

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