New Hydrogel obtained by Chitosan and Dextrin-VA Co-polymerization

Reinaldo Ramos, Vera Carvalho, Miguel Gama
IBB Institute for Biotechnology and Bioengineering, Universidade do Minho, Braga, Portugal

Introduction

Hydrogels are a class of three-dimensional, highly hydrated polymeric networks (1). They are suitable for biomedical applications because of their high tissue compatibility mainly caused by the high water content of the gels (2).

Chitosan (CS) (poly[β-(1-4)-2-amino-2-deoxy-D-glucopyranose]) is a non-toxic and biocompatible co-polymer of N-glucosamine and N-acetyl-glucosamine produced by partial deacetylation of chitin. Chitosan is susceptible to structural modifications due to the high number of hydroxyl and amine reactive groups.

Dextrin is a glucose-containing polysaccharide linked by α-(1→4) D-glucose units, with the same general formula as starch, although it is smaller and less complex. This polysaccharide is produced by partial hydrolysis of starch, which can be accomplished by the use of acids and/or enzymes.

In the present work, we describe the development of a novel hydrogel obtained by polymerization of chitosan with dextrin-VA.

Material and Methods

The enzyme-catalyzed modification of dextrin, with various degrees of substitution (DS) was as described elsewhere (3). Briefly, dextrin (4 g) and two volumes of VA (650 µl gave a DS of 20% and 2.6 ml a DS of 70%) were dissolved in anhydrous DMSO (60 ml).

Chitosan-dextrin-VA hydrogels were obtained after reticulation of chitosan with dextrin-VA. Chitosan was dissolved in acetic acid 0.1 M (1.5%, w/v). The pH was raised to 6 so that a higher number of reactive amines (NH_2) were available. The chitosan solution was mixed with various quantities of solid dextrin-VA and transferred to a casting mould. The polymerization occurred at room temperature for 24 h.
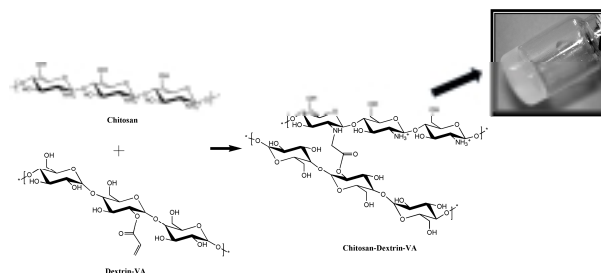
Rheological analyses of the hydrated hydrogels (25 mm diam. disk, 2 mm thick) were carried out using a Reologica StressTech HR reometer in parallel-plate geometry, with a variable gap.

Diffusion coefficients of glucose in the chitosan-dextrin-VA hydrogels were calculated using lag-time analysis (4).

Swelling ratios and mass loss profiles were calculated and the hydrogel structure was analysed by SEM.

Results and discussion

The chitosan-dextrin-VA hydrogels were obtained upon reticulation of chitosan with dextrin-VA. The amine group reacts with the acrylate group of dextrin-VA (figure 1), according to a conjugate 1, 4 addition mechanism. In this reaction, the nucleophile (amine group) reacts with the C=C double bond of the α,β–unsaturated acrylate group.

Figure 1 - Reaction between chitosan and dextrin-VA leading to hydrogel formation

Rheological analyses demonstrated that the hydrogel has a solid-like behavior with G' (storage modulus) >> G'' (loss modulus). The viscoelastic material functions G* and η are similar to values determined for brain tissue: a constant G* and η that increased with decreasing frequency (5).

Diffusion coefficients of $3.9\times10^{-6}\pm1.3\times10^{-6}$ cm^2/s and $2.9\times10^{-6}\pm0.5\times10^{-6}$ cm^2/s, for the hydrogels with a DS of 20 and 70% respectively, were obtained, using the lag-time analysis.

Mass loss profiles varied according to dextrin concentrations. After 30 days, the less concentrated hydrogels (60 mg dextrin/ml CS) lost 70% of mass against 50% for the more concentrated ones (90 and 180 mg dextrin/ml CS).

In the initial period of the incubation (day 5), an increase in the swelling ratio was detected for all hydrogels, due to the uptake of water into the hydrogel matrix. Then, the swelling ratio of all hydrogel samples started to decrease and stabilized until day 20. The swelling ratio increased again due to the high mass loss in the last days, leading to a loosened polymer network.

SEM observation revealed a porous structure, with pores ranging from 50 to 150 µm.

References

1. Peppas N.A., Bures P., Leobandung W., Ichikawa H. Hydrogels in pharmaceutical formulations. Eur J Pharm Biopharm, 50, 27– 46, 2000
2. Drury J.L., Mooney D.J. Hydrogels for tissue engineering: scaffold design variables and applications. Biomaterials, 24, 4337-51, 2003
3. Carvalho J., Gonçalves C., Gil A., Gama FM., Production and characterization of a new dextrin based hydrogel. Euro. Pol. J, 43, 3050-59, 2007
4. Teixeira J.A., Mota M., Venâncio A Model identification and diffusion coefficients determination of glucose and malic acid in calcium alginate membranes. Chem Eng J, 56, B9-B14, 1994
5. Woerly S., Pinet L., de Robertis L., Van Diep D., Bousmina M. Spinal cord repair with PHPMA hydrogel containing RGD peptides (NeuroGelTM). Biomaterials, 22, 1095-1111, 2001