NEWLY CROSSLINKED CHITOSAN-BASED HYDROGEL BIOMATERIALS: PHYSICAL, CHEMICAL AND MECHANICAL INVESTIGATION

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Introduction

Hydrogel materials are well known for their ability to absorb large amounts of water while maintaining the structure due to the crosslinking of polymer chains. They have attracted a great attention for their potential application in a lot of biomedical areas such as tissue engineering, dressing materials and controlled drug delivery systems. Hydrogels have a number of advantageous properties - the ability to mimic biomechanical characteristics of native extracellular matrix (ECM), ensuring an appropriate microenvironment for cells and promoting the transport of nutrients. Furthermore, their porosity, high swelling ability, and hydrophilic nature make hydrogels excellent candidates as carriers of hydrophilic biologically active compounds. Generally, all of these properties of hydrogels are highly associated with the degree of crosslinking.

Materials and Methods

In this study, porous hydrogel materials were produced based on two biopolymers - chitosan (CS) and pectin (PC), crosslinked with phenolic monoaldehyde - 2,3,4-trihydroxybenzaldehyde (THBA). Additionally, a polyphenolic compound - rosmarinic acid (RA) and a calcium-rich sol-gel-derived bioactive glass (BG) were used as functional components. The aim of the research was to assess the impact of the presence of individual substrates on the biopolymer cross-linking process as well as the final properties of the obtained hydrogels, including the microstructure (SEM/EDX, μ-CT) mechanical and thermal analysis (TG). Moreover, hydrogels were analyzed using ATR-FTIR spectroscopy.

Results and Discussion

μCT analysis of crosslinked hydrogels proved nearly 100% interconnectivity of the pores and high porosity (94.9% – 96.5%), regardless of the composition of the hydrogels. The crosslinking with THBA resulted in improved mechanical properties of materials (FIG. 1). Formation of the Schiff base in the chitosan matrix was confirmed by development of a distinct yellow colour. The FTIR spectra of crosslinked hydrogels showed an absorption band at 1628 cm^{-1}, corresponding to the stretching vibration of imine bonding. Furthermore, an absorption band of the phenolic hydroxyl groups of THBA shifted from 1279 to 1268 cm^{-1}, which may be due to the H-bonding between CS and THBA. When analysing the TG curves, crosslinked materials showed lower water content as well as enhanced thermal stability compared to uncrosslinked hydrogels, confirming the presence of covalent Schiff base bonding. Moreover, in the case of uncrosslinked materials, temperature of thermal decomposition of CS-PC hydrogels tended to be higher compared to CS materials, which may indicate ionic interactions between both polymers.

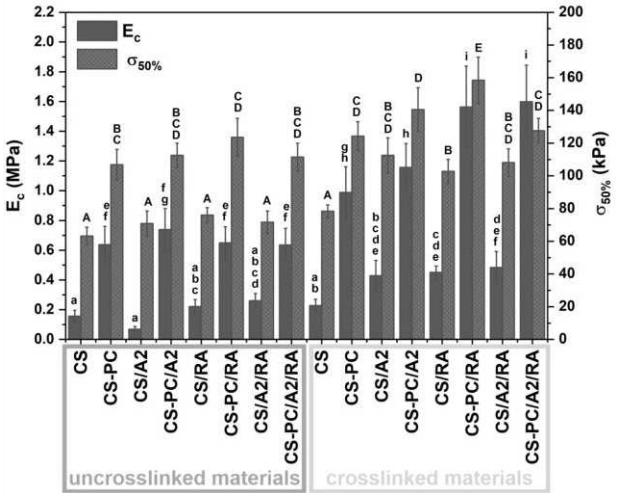

FIG. 1. Compression test results: Young's modulus and stresses corresponding to compression of a sample by 50% of the uncrosslinked and crosslinked hydrogels.

Conclusions

The presence of pectin, bioactive glass and rosmarinic acid, separately and in combination, affected the crosslinking process, while simultaneously modulating properties or imparting completely new ones. The obtained hydrogels represent promising multifunctional biomaterials with a wide range of physicochemical and mechanical properties with great potential for use in tissue engineering.

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References

[1] Hu, W., Wang, Z., Xiao, Y., Zhang, S., & Wang, J. (2019). Advances in crosslinking strategies of biomedical hydrogels. Biomaterials Science, 7(3), 843–855.
[2] Mallick, S. P., Suman, D. K., Singh, B. N., Srivastava, P., Siddiqui, N., Yella, V. R., Madhual, A., & Vemuri, P. K. (2020). Strategies toward development of biodegradable hydrogels for biomedical applications.