

Abstract

Gelation Kinetics of Chitosan-Based Hydrogels Using Different Crosslinking Agents [†]

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Introduction:

The crossover time (t_c) between the storage modulus (G') and loss modulus (G'') is a key rheological parameter used to characterize the sol–gel transition in hydrogel systems. This metric provides valuable insight into gelation kinetics and is directly associated with the processability of polymeric materials. In this study, bio-based hydrogels were prepared from chitosan (Ch), a natural polysaccharide, and crosslinked with two different agents: glutaraldehyde (GA), a synthetic dialdehyde, and genipin (GNP), a natural crosslinker derived from *Gardenia jasminoides*.

Methods:

Hydrogels were prepared by dissolving 4% w/v Ch in 2% v/v acetic acid, and then mixing in a 4:1 ratio with aqueous solutions containing GA or GNP at concentrations of 1, 5, and 10% relative to Ch weight. Rheological oscillatory measurements were performed during the crosslinking reaction.

Results:

Time sweep rheological tests were used to determine t_c , which marks the transition from a liquid-like to a solid-like state. At the lowest concentration (1% w/w), GA was insufficient to induce gelation, whereas GNP at the same concentration produced a t_c of 269 min. At 5% w/w , both crosslinkers successfully formed solid-like gels with t_c values of 18.6 min for GA and 91.2 min for GNP. Increasing the crosslinker concentration to 10% w/w further decreased t_c to 4.2 min for GA and 57.6 min for GNP. As expected, higher crosslinker concentrations led to faster gelation (lower t_c values). These results show that GA promotes faster crosslinking but requires higher concentrations, while GNP enables gel formation at lower dosages but with slower kinetics.

Conclusions:

Both the chemical nature and concentration of the crosslinker significantly affect the gelation time and mechanical evolution of bio-based chitosan hydrogels. Moreover, genipin represents a suitable substitute for glutaraldehyde in the development of aldehyde-free hydrogels, allowing longer processing times.



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