

Rheological Properties of Thermo-responsive Poly(N-Vinylcaprolactam)-g-Collagen Hydrogel

Authors : Serap Durkut, A. Eser Elcin, Y. Murat Elcin

Abstract : Stimuli-sensitive polymeric hydrogels have received extensive attention in the biomedical field due to their sensitivity to physical and chemical stimuli (temperature, pH, ionic strength, light, etc.). This study describes the rheological properties of a novel thermo-responsive poly(N-vinylcaprolactam)-g-collagen hydrogel. In the study, we first synthesized a facile and novel synthetic carboxyl group-terminated thermo-responsive poly(N-vinylcaprolactam)-COOH (PNVCL-COOH) via free radical polymerization. Further, this compound was effectively grafted with native collagen, by utilizing the covalent bond between the carboxylic acid groups at the end of the chains and amine groups of the collagen using cross-linking agent (EDC/NHS), forming PNVCL-g-Col. Newly-formed hybrid hydrogel displayed novel properties, such as increased mechanical strength and thermo-responsive characteristics. PNVCL-g-Col showed low critical solution temperature (LCST) at 38°C, which is very close to the body temperature. Rheological studies determine structural-mechanical properties of the materials and serve as a valuable tool for characterizing. The rheological properties of hydrogels are described in terms of two dynamic mechanical properties: the elastic modulus G' (also known as dynamic rigidity) representing the reversible stored energy of the system, and the viscous modulus G'' , representing the irreversible energy loss. In order to characterize the PNVCL-g-Col, the rheological properties were measured in terms of the function of temperature and time during phase transition. Below the LCST, favorable interactions allowed the dissolution of the polymer in water via hydrogen bonding. At temperatures above the LCST, PNVCL molecules within PNVCL-g-Col aggregated due to dehydration, causing the hydrogel structure to become dense. When the temperature reached ~36°C, both the G' and G'' values crossed over. This indicates that PNVCL-g-Col underwent a sol-gel transition, forming an elastic network. Following temperature plateau at 38°C, near human body temperature the sample displayed stable elastic network characteristics. The G' and G'' values of the PNVCL-g-Col solutions sharply increased at 6-9 minute interval, due to rapid transformation into gel-like state and formation of elastic networks. Copolymerization with collagen leads to an increase in G' , as collagen structure contains a flexible polymer chain, which bestows its elastic properties. Elasticity of the proposed structure correlates with the number of intermolecular cross-links in the hydrogel network, increasing viscosity. However, at 8 minutes, G' and G'' values sharply decreased for pure collagen solutions due to the decomposition of the elastic and viscose network. Complex viscosity is related to the mechanical performance and resistance opposing deformation of the hydrogel. Complex viscosity of PNVCL-g-Col hydrogel was drastically changed with temperature and the mechanical performance of PNVCL-g-Col hydrogel network increased, exhibiting lesser deformation. Rheological assessment of the novel thermo-responsive PNVCL-g-Col hydrogel, exhibited that the network has stronger mechanical properties due to both permanent stable covalent bonds and physical interactions, such as hydrogen- and hydrophobic bonds depending on temperature.

Keywords : poly(N-vinylcaprolactam)-g-collagen, thermo-responsive polymer, rheology, elastic modulus, stimuli-sensitive

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