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Rheological Study of Chitosan/Montmorillonite Nanocomposites: The Effect of Chemical Crosslinking

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Abstract : The development of hybrid organic-inorganic nanocomposites has recently attracted great interest. Typically, polymer silicates represent an emerging class of polymeric nanocomposites that offer superior material properties compared to each compound alone. Among these materials, complexes based on silicate clay and polysaccharides are one of the most promising nanocomposites. The strong electrostatic interaction between chitosan and montmorillonite can induce what is called physical hydrogel, where the coordination bonds or physical crosslinks may associate and dissociate reversibly and in a short time. These mechanisms could be the main origin of the uniqueness of their rheological behavior. However, owing to their structure intrinsically heterogeneous and/or the lack of dissipated energy, they are usually brittle, possess a poor toughness and may not have sufficient mechanical strength. Consequently, the properties of these nanocomposites cannot respond to some requirements of many applications in several fields. To address the issue of weak mechanical properties, covalent chemical crosslink bonds can be introduced to the physical hydrogel. In this way, quite homogeneous dually crosslinked microstructures with high dissipated energy and enhanced mechanical strength can be engineered. In this work, we have prepared a series of chitosan-montmorillonite nanocomposites chemically crosslinked by addition of poly (ethylene glycol) diglycidyl ether. This study aims to provide a better understanding of the mechanical behavior of dually crosslinked chitosan-based nanocomposites by relating it to their microstructures. In these systems, the variety of microstructures is obtained by modifying the number of cross-links. Subsequently, a superior uniqueness of the rheological properties of chemically crosslinked chitosan-montmorillonite nanocomposites is achieved, especially at the highest percentage of clay. Their rheological behaviors depend on the clay/chitosan ratio and the crosslinking. All specimens exhibit a viscous rheological behavior over the frequency range investigated. The flow curves of the nanocomposites show a Newtonian plateau at very low shear rates accompanied by a quite complicated nonlinear decrease with increasing the shear rate. Crosslinking induces a shear thinning behavior revealing the formation of network-like structures. Fitting shear viscosity curves via Ostward-De Waele equation disclosed that crosslinking and clay addition strongly affect the pseudoplasticity of the nanocomposites for shear rates $\dot{\gamma}$ >20.

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