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Cellulose-Reinforced Polyurethane Nanocomposites: Processing, Structure and Mechanical Properties

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Cellulose is the most abundant semi-crystalline bio-polymer on earth. Cellulose is produced by all plants, and by some living systems such as bacteria or tunicates. While the development of high-performance materials based on cellulose as a whole is limited due to cellulose structural defects, its transformation into nanocellulose could potentially broaden the range of cellulose-containing applications. Nanocellulose can be mechanically (cellulose nanofibrils) and chemically (cellulose nanocrystals) extracted and isolated from cellulosic materials. Nanocellulose shares common properties with cellulose in being bio-compatible, biodegradable, recyclable, and renewable. More importantly, nanocellulose combines a low density with a high stiffness, making it an excellent candidate for the reinforcement of polymer matrices. For these applications, cellulose nanocrystals are particularly interesting due to their high crystallinity and high stiffness. Cellulose nanocrystal/polymer nanocomposites exhibit superior mechanical properties compared to the neat polymer. Reinforcement mechanisms in isotropic nanocomposites have been widely studied and are known to follow the percolation theory. On the other hand, mechanical reinforcement can also be achieved via the alignment of the CNCs (Halpin-Tsai model), which has been only rarely studied due to the difficulty of aligning cellulose nanocrystals. The goal of this thesis was therefore to study the two reinforcement mechanisms within the same material, in order to determine which mechanism confers a stronger mechanical reinforcement. Different materials and techniques for aligning the CNCs within the polymeric matrix were explored. In Chapter2, neat polyurethane (PU) melt-spun fibers were produced at different spinning conditions to evaluate the alignment of the PU polymer chains and correlate it with the mechanical properties of the PU fibers. Upon gaining a basic understanding of PU melt-spinning, nanocellulose was added to the PU in order to produce nanocomposite materials. In Chapters 3, 4, 5, and 6 nanocellulose/polyurethane were fabricated using melt-spinning and solvent casting, creating anisotropic and isotropic materials, respectively. Nanocellulose with different aspect ratios were chosen, extracted from cotton and tunicates to evaluate their effects on the mechanical properties of the nanocomposites. Finally, in Chapter 7, electrospinning techniques for achieving the alignment of cellulose nanocrystals were similarly explored

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