

## Bioinspired Composites with Adaptive Mechanical Properties

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The main objective of this thesis was to create new bioinspired mechanical adaptive materials, which can be switched between a rigid and a soft state on command. The materials mimic, more or less closely, the stimuli-responsive behavior of sea cucumber, in which their stiffness is controlled by regulating chemicals. This adaptive mechanical behavior is achieved through a composite architecture and by exploiting two different concepts.

Cellulose nanocrystals (CNCs) can be used as a reinforcing filler due to their high stiffness and strength. However, in many cases, the reinforcing effect of CNCs, especially of low-aspect ratio CNCs such as those derived from cotton (c-CNCs), does not match the theoretical predictions, arguably on account of CNC aggregation. With the goals to confirm this conclusion and to improve the dispersibility of CNCs, which affect to the mechanical properties of overall materials, a new strategy was explored. Using poly(ethylene oxide-co-epichlorohydrin) (EO-EPI)/CNC composites as a model system, the impact that a very small amount of poly(vinyl alcohol) (PVA) has on both the homogeneity, i.e. dispersibility of CNCs in the polymer matrix, and the mechanical properties of EO-EPI/CNC composites were investigated. Evidence of the compatibilization effect of the hydrogen bonding additive through CNCs morphology using laser scanning microscopy represents the first visual confirmation of the generally accepted reinforcing mechanism by PVA. This strategy is versatile and can be applied to a wide range of matrices. In order to develop the use of c-CNCs composites into mechanical adaptive application, the study of stiffness-changing of artificial composites based on c-CNCs was investigated. The prepared composites were investigated in the dry state and in the soft state by immersing the samples into water. The results indicate that the composites made from c-CNCs together with PVA not only show an improved reinforcement effect in the dry state, but they reveal that these materials can switch to a low modulus after a short period of exposure to water. This data show that c-CNCs filler can be equally used for making mechanical adaptive materials instead of tunicate-derived CNCs, which are from a source that is not viable for technological exploitation.

A completely new concept for the design of self-toughening polymers is presented. The approach that was investigated in this study involves the incorporation of plasticizer-filled microcapsules in an intrinsically rigid and brittle matrix polymer. The stimulating adaptability is demonstrated with composites composed of a low impact resistance and brittleness poly(lactic acid) matrix and poly(urea-formaldehyde) microcapsules that contained hexyl acetate as plasticizer. This design permits creating composite materials that exhibit mechanically induced mechanical morphing that manifests itself in the form of a rapid and pronounced self-toughening effect.

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