SHORT ABSTRACT

In recent decades, increased environmental concerns to reduce carbon foot print and rising prices of petrochemicals have attracted much attention towards developing green and sustainable biodegradable polymers and nanomaterials, which will help in securing the planet's ecological balance. The studies based on utilization of renewable resources for development of value-added products ensuring environmental sustainability have become essential to confront the unprecedented implications of petroleum-based products related to global warming. Nature-based nanomaterials such as cellulose nanocrystals (CNCs), are derived from the abundantly available cellulosic biomass with unique inherent characteristics that have led to increased scientific and industrial attention. CNCs are the crystalline domain of cellulose with rod- like morphology and interesting properties such as high surface area, tunable aspect-ratio, chemical functionality, anisotropic mechanical properties, self-ordering behaviour to form optically active materials along with improved biocompatibility, biodegradability and non-toxic nature, provides it an upper hand in comparison to other nanoparticle derivatives. CNCs due to its sustainable nature, bio-origin and anisotropic mechanical behaviour have promising applications as reinforcing agents in polymeric nanocomposites, as targeted drug delivery vehicles, in biocatalysis, and in paper –based electronics as sensors. However, production of CNCs through industrially viable approach is still a challenging task due to high processing time, uncertainty in initial cellulosic precursor and requirement of stringent acid hydrolysis conditions. Moreover, literature survey suggests that processing of CNC-based polymeric nanocomposites through scalable extrusion techniques results in agglomeration due to their hydrophilic nature alongwith severe degradation in the molecular weight of polymer. Therefore, the present doctoral thesis aims to address both the techno-economic challenges associated with the processing of CNCs or its polymeric composites and develop advanced functional materials with CNCs as building block for high performance applications.

In the present work, CNCs have been fabricated from renewable feedstock such bamboo stems, rice husk, commercial pulp and newspaper for their potential application as commercially viable value added products, with a strategy to convert "wastes to wealth". The challenges associated with high production time was overcome with an indigenously developed CNC production unit coupled with continuous dialysis system which reduces the processing time to ~24-26 hours in comparison to ~7 days through traditional approach. Interestingly, CNCs with tunable physico-chemical, structural and morphological properties, are fabricated by simply varying the initial cellulose polymorphism or the type of acids used as hydrolyzing agents, explicitly during its processing step. Strategic modification of CNCs through this sustainable approach avoids the utilization of harmful and toxic chemicals which might affect the biocompatible and non-toxic properties of CNCs. Dispersion of surface-modified CNCs into polymeric matrix leads to the formation of percolation network which results in considerable improvement of mechanical and barrier properties of biodegradable polymers (such as polylactic acid (PLA) and polyhydroxybutyrate (PHB)), making them ideally suited for food packaging applications. An industrially viable approach for processing PLA/CNC films through reactive extrusion process have been developed which was successfully tested on a pilot-plant scale extruder in presence of wide range of compatibilizers. The reactively processed PLA/CNC cast films shows improved molecular weight characteristics, thermo-mechanical and barrier properties with dimensional stability and migration values within the standard limits, which is generally difficult to achieve through traditional extrusion process limiting their commercial viability. The developed films are promising for food packagings and result in improved self-life of ~5 months and ~2 weeks respectively for the storage of oil and dairy based products. In addition, these films have the potential to be produced at commercial scale through utilization of industrial scale extruders with improved dimensional stability, stable molecular weight and physico-chemical characteristics. Magneto-responsive CNCs, are one of the versatile nanomaterial which have been developed in this work, with potential applications as self-propagating nanobots with autonomous motion as targeted drug delivery vehicles, biocatalysts with high activity and as templates for fabrication of graphene nanoscrolls with tunable geometry and encapsulated metallic nanoparticles as hydrogen storage devices. Magnetic field directed alignment of such nano-block units introduces anisotropic thermo-mechanical, electrical and magnetic properties alongwith the hyperthermia characteristics finds potential applications in development of bio-inspired materials with microstructural features comparable to biological materials for high performance engineering applications. Therefore, this doctoral thesis focusses on functionalization of CNCs for development of polymeric bionanocomposites as well as their advanced functional derivatives and studying their efficacy in sustainable chemical processes, which have been summarized in nine different chapters and explained in details in subsequent sections.