Doctoral Dissertation
Doctoral Program Material Science and Technology (38th Cycle)

Design of advanced biobased photocurable formulations: from dynamic covalent network to composite scaffold for tissue engineering

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Abstract

The transition towards sustainable materials has become a global priority, driven by environmental concerns and the growing demand for high-performance systems with minimal environmental impact. Thermosets, widely used in industrial and biomedical fields due to their excellent stability and mechanical strength, face two key challenges: their permanent cross-linked structure prevents reprocessing, and their dependence on fossil-derived feedstocks hinders sustainability. This thesis was developed to address these issues through the design of bio-based, UV-curable, and reprocessable polymer networks intended for use in both industrial and biomedical applications.

The research is divided into two main thematic areas. The first focuses on the synthesis of bio-based dynamic networks derived from epoxidised castor oil. Using a transesterification catalyst, covalent adaptable networks (CANs) were developed. They retained the high-performance characteristics typical of thermosets while introducing dynamic bond exchange reactions, which enable reprocessing, reshaping, and recycling, key aspects of circular economy principles. These materials were further enhanced by incorporating multiwalled carbon nanotubes (MWCNTs), which conferred electrical conductivity and improved mechanical performance. The resulting multifunctional composites combined a high bio-based content with advanced properties suitable for applications in coatings.

The second research area focused on biomedical applications, integrating sustainability with bioactivity and functionality. The initial investigations involved the development of chitosan-based hydrogels reinforced with tellurium-doped and silanized bioactive glasses (BGs). These systems merged the intrinsic biocompatibility of chitosan with the osteoconductive, antioxidant, and antimicrobial properties of functionalized BGs, offering promising potential for soft tissue regeneration.

The research then moved towards the fabrication of highly porous composite scaffolds using a high internal phase emulsion (HIPE) templating approach combined with UV-induced thiol–ene photopolymerization of acrylated epoxidised soybean oil (AESO) and Trimethylolpropane tris(3-mercaptopropionate) (TMPTMP). The resulting polyHIPE scaffolds exhibited a highly interconnected, open-cell structure resembling cancellous bone, making them ideal for tissue engineering applications. The inclusion of bioactive glasses enhanced both the mechanical and biological performance, thereby increasing osteointegration. Additionally, additive manufacturing techniques were employed to control scaffold architecture, allowing for the production of customised 3D-printed constructs with tunable porosity and patient-specific geometries.

In subsequent works, AESO-based scaffolds were 3D-printed and reinforced with bioactive glasses of varying compositions. The BGs were doped with Cu and Te to impart angiogenic and antimicrobial properties, and then silanized to enhance interfacial adhesion with the polymer matrix. Each formulation was carefully analysed to understand the influence of composition and filler content on mechanical behaviour and bioactivity. The final stage of the thesis bridged the two research lines by introducing reprocessability into AESO-based scaffolds, combining biodegradability, bioactivity, and dynamic reprocessability within a single system. This demonstrated the feasibility of extending covalent adaptable network principles to biomedical materials, merging sustainability and functionality.

In conclusion, this thesis demonstrates that the combination of renewable monomers, dynamic covalent chemistry, functional fillers, and advanced manufacturing techniques can lead to thermosets and scaffolds that are not only sustainable and high-performing but also tailored for real-world applications. The reprocessable epoxy composites and bioactive scaffolds developed exemplify how biobased polymers can meet industrial and biomedical demands, contributing to the transition toward a more sustainable and circular materials economy.