EPOXIDIZED NATURAL RUBBER BASED COMPOSITES FOR MULTIFACETED APPLICATIONS

Abstract

Epoxidized natural rubber (ENR), with its dual reactivity arising from both double bonds and epoxide functionalities, offers a versatile platform for developing advanced elastomeric materials. This thesis investigates various crosslinking strategies that exploit epoxide functionality in comparison to sulfur-based curing methods through multiple analytical characterization, optimization methods, and functional enhancements to improve mechanical performance, optical clarity, flexibility, and advanced functionalities like EMI shielding and ion transport properties.

A new curing mechanism was established using hydroquinone (HQ), which interacts with the epoxide groups of ENR to initiate crosslinking. This transparency, coupled with improved mechanical durability and thermal stability, positions HQ-cured ENR as a promising material for high-performance coatings, stretchable electronics, and flexible robotic components. Building on this approach, aminopropyl-terminated low molar mass polydimethylsiloxane (AP-PDMS) was introduced, enabling compliant hybrid networks through epoxy–amine curing catalyzed by HQ. These ENR–PDMS hybrids demonstrated exceptional softness, wide range flexibility (elongation up to ~ 800%), low glass transition temperatures, enhanced hydrophobicity, and high optical clarity. Their ability to homogeneously host silica nanoparticles further broadened their functional applicability, reinforcing their suitability for low-toxicity, transparent, and stretchable elastomer platforms.

To systematically maximize property performance, the Taguchi statistical method was applied to optimize the simultaneous loading of AP-PDMS, HQ, epoxidized soybean oil (ESO), and silica fillers. The optimized formulations yielded a dramatic improvement in tensile strength (~ 1130%) and tear strength (~ 295%), while ESO enhanced elongation at break (~ 36%), when compared to the least performing composites. This multi parameter optimization methodology enabled balanced property improvements with few trial runs. Further, in efforts to design multifunctional composites, 3 Aminopropyl triethoxysilane (APTES) functionalized silica (AS) was introduced as both a reinforcing filler and a chemical crosslinker with ENR, enabled by HQ catalysis. Incorporation of carbon fibers (CF) prepared from electrospun polyacrylonitrile into these ENR-AS networks imparted strong electromagnetic interference shielding efficiency (~ 22 dB at < 15 wt.%), marking these as lightweight flexible EMI shielding materials. Complementarily, the potential of ENR for solid state energy storage was demonstrated through the development of Li-salt loaded ENR/polyethylene oxide (PEO) blend polymer electrolytes incorporating high-entropy oxide (HEO) fillers. These composite membranes achieved ionic conductivities up to 0.9 × 10-3 S·cm-1 at 100 °C, facilitated by increased free-ion concentration, reduced relaxation times, and enhanced amorphous content. Dielectric and thermal analyses highlighted the synergistic role of HEOs in improving ionic transport properties, polymer relaxation behaviour, and overall thermal stability. These findings highlight the promise of ENR based electrolytes for high-temperature lithium battery applications.

Collectively, this work demonstrates that through strategic crosslinking, hybridization, formulation optimization, and multifunctional filler design, ENR-based composites can be transformed into a versatile family of materials with applications spanning transparent and stretchable electronics, hydrophobic coatings, EMI shielding, and high temperature polymer electrolytes for energy storage systems.

Keywords Epoxidized Natural Rubber, Crosslinking Mechanisms, Hydroquinone, Polydimethylsiloxane, Modified silica, EMI shielding efficiency, High Entropy Oxides, Polymer Electrolytes.