Abstract

Elastomers are an important class of materials used in various sectors, viz. automotive, sports, households, biomedical applications, etc. The elastomer products undergo fractures, deformation, and crack formation due to several external stimuli. These waste materials are either scrapped, incinerated, or dumped into the soil, which causes serious environmental issues and an extra budget for new products. The elastomers are traditionally crosslinked via sulfur, peroxides, metal oxides, or resins. Recycling and reprocessing of these crosslinked products have been challenging for the materials research community. From this point of view, a circular and sustainable approach will be great if the elastomers are crosslinked via dynamic crosslinking to enhance their service life and to induct reprocessing and recyclability characteristics in the elastomer. In this context, this thesis delineates the synthesis of multifunctional elastomers like polyurethanes (PUs), non-isocyanate-based PUs, and modification of commercially available elastomers like epoxidized natural rubber (ENR) and poly (ethylene-*co*-vinyl acetate-*co*-glycidyl methacrylate) (EVA-GMA) having self-healing recycling and reprocessing characteristics. For this purpose, different diene substituents (e.g., furfuryl alcohol, furfuryl amine, and anthracenyl acid) were grafted to the elastomer chain to prepare modified elastomers having reactive dienes. Next, these diene functionalized elastomers were further modified using suitable dienophiles like maleimide derivatives such as 1,1'(methylenedi-4,1-phenylene) bismaleimide (BM), polyhedral oligomeric silsesquioxane isobutyl maleimide (POSS-MI), and 1,2,4-triazoline-3,5-dione (TAD) derivatives like bifunctional TAD (bis-TAD) and phenyl TAD (Ph-TAD) via dynamic Diels-Alder (DA) and electrophilic substitution (ES) "Click" Chemistry. Interestingly, the "Click" reactions involving TAD-derived were much faster than the maleimide-derived elastomer. The multifunctional properties of these modified elastomers, like self-healing, mechanical, fluorescence switching, and hydrophobic properties, were analyzed by optical microscopy, tensile, AFM, nanoindentation (NINT), fluorescence spectroscope, and water contact angle (WCA) analyses. These DA/ES-Clicked elastomers showed significant self-healing, solution reprocessing, and recyclability characteristics, indicating this work's sustainability. These DA/ES-Clicked elastomers can have potential applications in specialty rubber products, chemosensors, functional paints, coatings, and adhesives.

Keywords: Dynamic crosslinking, PU, epoxidized natural rubber, EVA, Diels-Alder (DA), electrophilic-substitution (ES), self-healing, recycling, reprocessing, hydrophobic, adhesives, and fluorescence switching.