

## *ABSTRACT*

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The emergence of biodegradable polymers and nanocomposites derived from renewable resources represents a crucial initiative in the development of products suited for both everyday and cutting-edge applications. Again, it is an arduous challenge to achieve high mechanical strength, good shape recoverability, and biocompatibility along with high self-healing efficiency in the same waterborne polyurethane (WPU). Congenitally, these properties are associated with conflicting features which makes it puzzling to optimize them. In this regard, the present thesis is dedicated to the preparation and precise chemical modification, characterization, performance assessment, and potential applications of smart WPUs their nanocomposites (WPUNCs) derived from renewable resources, with appropriate carbon-based nanomaterials. The work presented in this thesis can be categorized into the following parts: (a) preparation of anionic WPUs using a biobased internal emulsifier and other desired reactants, (b) structural modification of WPU through acrylate monomers and their characterization, (c) introduction of asymmetric architecture i.e. isophorone diisocyanate-isophorone diamine (IPDI-IPDA) moieties into the hard domains of the WPU matrix, (d) imparting dynamic S-S bond containing hard domain into the WPU for boosting self-healing ability and mechanical strength, simultaneously, (e) incorporation of S and Mo co-doped graphitic carbon nitride (Mo@S-CN) into the best self-healable WPU (SHWPU) for the preparation of photoluminescent SHWPU/Mo@S-CN nanocomposites, and (f) evaluation of various smart properties of them for multifaceted applications. The developed materials exhibited numerous eye-catching properties e.g., an optimal SHWPU film can be stretched up to 6300% from its original shape and the film exhibited the record highest toughness of 436.1 MJm<sup>-3</sup>. Outstandingly, another optimal SHWPU film exhibits remarkable toughness, to the extent that it can withstand the lifting of a 25 kg dumbbell, which is a staggering 53,648 times heavier than the weight of the film, without showing any signs of cracking. Moreover, SWPUA and SHWPU dispersions can serve as the primary components for creating 3D-printable bone scaffolds.

Therefore, the current study has unveiled a pathway for attaining various intelligent properties within a single polymeric system, for specific applications by utilizing engineered nanomaterials.

**Keywords:** Bio-based Waterborne Polyurethane, Self-Healing Material, Shape Memory Material, 3D-Printing, Anti-Counterfeiting, Biodegradable Polymer.