# **Bio-based Epoxy Thermosets and Their Composites**

A thesis submitted in partial fulfilment of the requirements for the degree of

# **Doctor of Philosophy**

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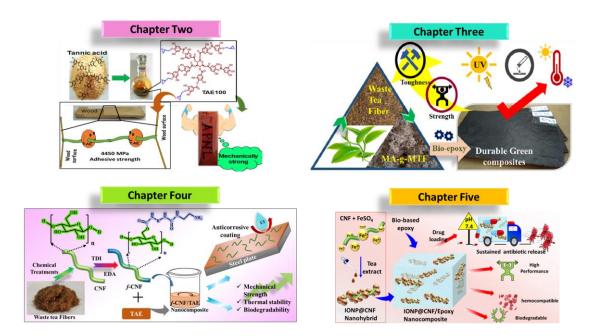
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### Summary, conclusions and future avenues

### Highlights

This chapter synopsizes all the investigations performed in this thesis and their crucial outcomes in a concise way. The chapter summaries all the preceding chapters and presents their accomplishments succinctly. Most importantly, it documents all the anticipated research opportunities stemming from the investigation presented, which may attract further attention for in-depth research in the field of bio-based epoxy thermosetting nanocomposites. The graphics shown below represents the highlights of the thesis, chapter-wise from **Chapter Two** to **Chapter Five**.



#### 6.1 Summary and conclusions

The present investigation focuses on the development of a benign epoxy and its composites from sustainably sourced raw materials. The study showcases the efficiency of different sustainably derived micro-/nano-reinforcing agents in improving the performance of bio-based epoxy. Some additional adventitious attributes like environmental durability, corrosion resistance, and drug-releasing ability were induced in the epoxy systems, endowing them with multidimensional applicability.

**Chapter One** provides an overview of the systematic evolution of bio-derived epoxies in the light of existing state-of-the-art literature reports. Different limitations stemming from the inherent brittleness of sustainable epoxies are addressed along with some cutting-edge amelioration tactics, including the incorporation of sustainable reinforcing agents. Additionally, the chapter briefly discusses the standard protocols used for the synthesis, extraction, fabrication, and characterization of these epoxies, bioreinforcing agents, and composite systems as well as evaluation of their properties. Considering the relevance of the investigation in the present scenario, the scopes, objectives, and plan of action strategized for the present investigation have been catalogued.

In order to synthesize a bisphenol-A (BPA)-free bio-derived epoxy, various methodologies have been adopted using different precursors and the approaches have been optimized. **Chapter Two** describes the technical aspects of the bio-based epoxy derived from tannic acid (TAE) by the conventional polycondensation approach. The structural complexities of the epoxy were analyzed using available spectroscopic and analytical methodologies. The thermal stability, mechanical and adhesive strength, antioxidant activity, transparency, and biodegradability of the epoxy were evaluated and compared with three contrast epoxy resins derived using BPA at different weight percentages with tannic acid (TA).

To improve the performance of this sustainably derived epoxy, different multifunctional reinforcing agents have been utilized, which are demonstrated in **Chapters Three**, **Four** and **Five**. Thus, **Chapter Three** explained the reinforcing effects of chemically modified natural fibers on TAE. Industrial waste tea fibers (WTFs) have been employed for obtaining the microfibers by treating them with alkali, which was later modified with maleic anhydride (MAH). The incorporation of these modified fibers into

the epoxy system resulted in biocomposites with enhanced tensile strength and thermal stability compared to unmodified ones. The biocomposites offered considerably good durability upon exposure to aggressive environments such as temperature, UV light, harsh chemical media, etc. The retention in their tensile strength values indicated substantial withstanding capacity of the biocomposites to external stimuli. Simultaneously, the biocomposites possessed biodegradability which was assessed by the soil burial test.

Sustainably derived nanostructures like cellulose nanofibers (CNFs), preferably derived from waste or biomass, possess great potential as reinforcing agents. Additional benefits are obtained when the surface characteristics of these nanostructures are altered by introducing suitable chemical linkages. **Chapter Four** deals with the extraction of CNFs from industrial WTFs using successive chemical and mechanical treatments. CNFs were further functionalized with pendant amine groups using toluene diisocyanate and ethylene diamine. These functionalized CNFs when imbedded into TAE improved the mechanical strength and glass transition temperature of the latter. The optimum interfacial interaction achieved between the TAE matrix and the nanofibers by the introduction of amine functionalities was responsible for such improvements. The corrosion resistance performance of the nanocomposites was evaluated against mild steel plates. This confirmed the prospective application of the nanocomposites as biodegradable anticorrosive coatings.

**Chapter Five** of the thesis discusses the generation of a nanohybrid by decorating biogenic iron oxide nanoparticles (IONPs) over the surface of CNFs. The in-situ nucleation and growth over the CNF surfaces were accomplished by using polyphenolic extracts derived from the WTFs. Herein, WTFs were completely utilized for extracting the polyphenolic extract and CNFs in a sequential process. The nanohybrid, at different weight percents, was incorporated into TAE for obtaining a biocompatible (confirmed from the hemolysis and cell viability study) and biodegradable (under an accelerated bacterial environment) high-performance epoxy nanocomposite. The nanocomposite was explored for prospective utility in the field of biomedical science, as a drug-carrying vehicle where pH responsive release behavior was unveiled.

This chapter, as discussed, is highlighting the summary and conclusions of the investigation. Delineated below are the key accomplishments of this work.

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- The study showcased the effective production of a bio-based epoxy, TAE through a straightforward and facile method involving the polycondensation reaction between TA and epichlorohydrin.
- On crosslinking with an amine-based sustainable curing agent, the highly transparent epoxy thermosets revealed better performance compared with the contrast thermosets containing BPA along with TA, marked by a high tensile strength, high thermal stability, good resistivity to harsh chemical environments and biodegradability.
- MAH-modified microfibers incorporated biocomposite of TAE possessed good resistance to harsh environmental conditions, marked by considerable strengthening under UV and elevated-temperature exposures.
- CNFs derived from WTFs were functionalized with pendant amine groups using toluene diisocyanate and ethylene diamine.
- These functionalized CNF-reinforced TAE nanocomposites demonstrated enhancements in thermal and mechanical properties. A maximum improvement of 61% (27.79 MPa compared to 17.3 MPa of TAE) was recorded in the tensile strength of the nanocomposite with a very low loading (0.2 wt%) of *f*-CNFs with considerably high elongation (50.4%).
- The biodegradable nanocomposite was successfully applied as an anticorrosive coating on mild steel plates and exhibited a maximum protection efficiency (99.95%) of the nanocomposite with 0.3 wt% *f*-CNFs loading even after 7 days immersion in 3.5% NaCl solution.
- An innocuous nanohybrid was derived by growing IONPs over the surface of CNFs and the nanohybrid was imbedded into TAE to generate a high-performance nanocomposite. Improvements of 41.6% in tensile strength and 150 J/m<sup>3</sup> in toughness were achieved for the nanocomposite.
- The biocompatible and biodegradable nanocomposite could be tuned for sustained release of an antibacterial drug that can be explored in biomedical delivery prospects. A maximum release of 76% was achieved for nanocomposite with 1 wt% nanohybrid content, in an environment of pH 7.4.

Thus, the study brings to light the promising advantages of sustainably derived epoxy and its composite systems for multitudinous applications. Nevertheless, achieving practical implementation in real-world scenarios demands further in-depth scrutiny within their specific domains.

### 6.2 Future avenues

The present thesis inquisitively documents the possibilities of eco-friendly and resourceefficient epoxy-derived materials in heterogeneous professions. The study further unveils fresh avenues for enthusiastic research endeavors, a few of which are listed below.

- The literature, although, exemplifies the utilization of various renewably sourced building blocks for the synthesis of bio-based epoxy resins, there still remains a wealth of efficient and abundant biomolecules waiting to be further explored in this domain. Besides, the amalgamation of precisely designed moieties possessing optimized content of rigid and flexible domains can be accentuated to attain advanced attributes in the bio-based epoxy.
- Incorporation of hybridized microfibers in bio-based epoxies for producing structural composites with add-in attributes namely, sound absorption ability, unprecedented crack or impact resistance, etc., can be directly explored in the aviation industry.
- Attempting pragmatic approaches for tailoring exotic nanostructures and monitoring their impression in bio-based epoxies along with exploring their feasibility in different realms of science and engineering.
- Concerning the future of the prepared nanocomposites in the field of anticorrosive coatings, more sophisticated and advanced tactics need to be explored to widen their practicability.
- For the prepared materials to be applied in the field of biomedicine, a deeper insight into their biocompatible nature is required. Several clinical trials need to be performed *in-vitro* as well as *in-vivo* to establish their innocuous nature and to prove their potency as drug releasing agents under authentic physiological environments.
- To complement the real possibilities and sustainable nature of the prepared materials, a comprehensive life cycle assessment needs to be performed, which will picturize the genuine impacts offered by these materials from cradle to grave.