Doctoral Dissertation
Doctoral Program in Materials Science and Technology (38th Cycle)

Composite materials: new technology applications

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Summary

This PhD dissertation investigates the applications of Tannic Acid (TA), a naturally occurring polyphenolic macromolecule, as a multifunctional modifier in advanced composite materials and thermoplastic polymer systems. The research addresses three principal challenges in contemporary materials science: the inadequate interfacial adhesion between ultra-high molecular weight polyethylene (UHMWPE) fibres and polar matrix systems, the substantial environmental burden associated with petroleum-derived thermosetting resins, and the inherent susceptibility of polyolefin materials to thermal-oxidative degradation.

The experimental investigation encompasses three interconnected research domains. The initial phase establishes a sustainable surface modification methodology for UHMWPE fabric through aqueous TA-Na^+ coordination chemistry conducted at room temperature, thereby circumventing the requirement for volatile organic solvents and energy-intensive processing conditions. Systematic parametric analysis of immersion duration (1-6 hours) and TA concentration (1:1 to 40:1 TA:Na^+ weight ratio) identified optimal surface functionalisation conditions at 3-6 hours exposure and 10:1 concentration, yielding substantial improvements in wettability characteristics: water contact angle decreased from 107° to 71°, whilst epoxy resin contact angle improved from 82° to 66°. Notably, this investigation reports for the first time a significant enhancement in thermal decomposition resistance of UHMWPE fibres following TA surface treatment, with $T_{5\%}$ increasing from 336°C to 454°C in nitrogen atmosphere, whilst preserving the crystalline microstructure (>82% crystallinity retention).

The second research domain demonstrates the successful implementation of TA as bio-derived crosslinking agent for diglycidyl ether of bisphenol A (DGEBA) epoxy formulations, enabling curing temperatures compatible with thermoplastic fibre reinforcements (125°C) and conventional composite consolidation protocols (170°C). At 30 wt.% TA loading, the formulation achieved 63-84% epoxide conversion depending upon thermal treatment conditions. The TA-hardened systems exhibited superior flame retardancy characteristics, demonstrating 44% reduction in Peak Heat Release Rate and transition to self-extinguishing behaviour according to TL1010 automotive flammability specifications. Comprehensive mechanical characterisation across multiple reinforcement architectures (carbon, glass, aramid, and flax fibres) validated the universal applicability of TA-crosslinked matrices, with carbon fibre composites achieving 453 MPa flexural strength and glass fibre systems exhibiting 70% enhancement in Charpy impact resistance relative to conventionally cured reference materials.

The third section examines TA incorporation into linear low-density polyethylene (LLDPE) matrices via maleic anhydride-grafted polyethylene (PEGMA) reactive compatibilization. The optimised ternary LLDPE/PEGMA/TA (BLEND) formulation demonstrated remarkable oxidative stability enhancement, with Oxidation Induction Time increasing from 1.1 minutes to 82.8 minutes – representing a 75-fold improvement at 10 wt.% TA loading. Thermal decomposition onset temperatures increased by 7-16°C depending upon matrix composition, whilst maintaining adequate melt processability for industrial film extrusion applications.

This dissertation establishes TA as a technically viable and environmentally sustainable alternative to conventional synthetic additives, providing concurrent improvements in thermal stability, flame retardancy, and mechanical performance whilst reducing life-cycle environmental impact. Industrial validation through automotive component fabrication demonstrates scalable manufacturing feasibility.