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European Polymer Journal
Volume 197, 10 October 2023, 112355

Valorization of agricultural waste to polybenzoxazine-carbon composites: Studies on microstructure, thermal and dielectric properties

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https://doi.org/10.1016/j.eurpolymj.2023.112355
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Highlights

- Novel card-bisphenol has been synthesized from agricultural bio-waste.
- Bio-based benzoxazine has been synthesized and reinforced with sustainable different bio-carbon resources.
- Increase in bio-carbon content increases the hydrophobic nature of the composites.
- Bio-carbon derived from *Tamarindus indica* exhibit higher dielectric constant.

Abstract

Agricultural waste valorization has become a necessity to reduce global warming. In this context, absolute bio-waste derived cardanol, furfurylamine (ffa), and benzaldehyde were utilized for the synthesis of a novel card-bisphenol (BBC) and its benzoxazine (BBC-ffa) resin. Further, the reinforcement of carbon building blocks derived from three different sustainable resources, viz.

Anacardium occidentale, *Borassus flabellifer*, and *Tamarindus indica* and named cashew apple carbon (CC), palmyra palm inflorescence (spadix) carbon (PC), and tamarind seed carbon (TC), respectively. The derived carbons were reinforced (1–20 wt%) separately with BBC-ffa polymer matrix [poly(BBC-ffa)] for the engineering of carbon integrated polybenzoxazine composites [poly(BBC-ffa)–CC/PC/TC] to achieve high-k dielectrics. Because the carbon with different functionalities intrinsically exhibit higher polarization and lower specific surface area. Dielectric constant (k) of the polybenzoxazine matrix (k = 4.62) has been increased to 8.58, 9.43, 10.60 for the reinforcement of 20 wt% of each CC, PC, and TC polybenzoxazine composites, respectively and the value of dielectric loss was also significantly decreased for the same. The dissemination of reinforced carbon particles in the polybenzoxazine matrix was ascertained through the surface analysis of the composites by FE-SEM. Based on the results of TGA, the carbon reinforced composites showed slightly lower thermal stability and about two-fold higher char yield than that of neat polybenzoxazine. Hydrophobicity of the composites enhanced with increasing the carbon content. The resulting sustainable polybenzoxazine-carbon composites could be effectively utilized as a gate dielectric in microelectronics.

Graphical abstract

Introduction

Benzoxazine (Bz) resin, a class of thermosetting polymers is in the limelight for the past few decades for its uniqueness such as excellent chemical resistance, near-zero volumetric shrinkage upon curing, minimal moisture adsorption, flexibility in molecular design, high glass transition temperature (T_g), and easy thermal curing without any addition of hardeners or catalysts [1], [2], [3], [4], [5], [6]. Another intriguing characteristic of benzoxazines is their easy and handy synthesis through Mannich-condensation of a phenol, primary amine, and formaldehyde [7]. For commercialization, benzoxazine resins face some difficulties in availability of raw materials which are derived from petroleum products. The massive utilization of petroleum-based polymeric products is one of the causes of potential toxic wastes which lead to severe environmental pollution [8]. Recently, researchers have been focusing on replacing petroleum-based raw