Journal of Polymers and the Environment
https://doi.org/10.1007/s10924-024-03248-y

ORIGINAL PAPER

Unlocking the Potential of Resveratrol-Derived Trifunctional Photosensitive Benzoxazines for Superhydrophobic, Low Dielectric and Photoluminescence Applications

Subasri Appasamy[1] · Balaji Krishnasamy[1] · Hariharan Arumugam[1] · Alagar Muthukaruppan[1]

Accepted: 24 February 2024

Abstract
In the present study, fully bio-based photocurable trifunctional benzoxazines derived from resveratrol were designed and characterized for multi-dimensional applications. The molecular structure was confirmed using ATR-FTIR and ^{1}H-NMR spectral techniques. The curing temperature and thermal stability were thoroughly studied using differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA) respectively. Notably, in DSC analysis RE-dda exhibited the lowest curing temperature of 191 °C among the synthesised benzoxazines. All the synthesised polybenzoxazines exhibited good thermal stability and start degrading after 263°C. Under UV irradiation, intriguing photoisomerization phenomena was observed in the case of RE-ffa and RE-lee based benzoxazines. The photoluminescence behavior of the UV irradiated benzoxazines was studied using fluorescence spectroscopy and RE-oda displayed a Stokes shift value of 318. Further benzoxazines were reinforced with cashew nut shell cake ash (CNSA) with a view to attain a low value of dielectric constant and to enhance the thermal stability of the composites. An incorporation of 10 wt% CNSA contributes to a reduction in the value of dielectric constant to 1.66, accompanied with a minimal value of dielectric loss of 0.0017. Furthermore, the hydrophobic behavior of the polybenzoxazines, composites and poly(RE-ole) coated cotton fabric was evaluated using water contact angle measurement. Poly(RE-ole) exhibited an impressive water contact angle value of 146°. Moreover, poly(RE-ole) coated cotton fabric displayed enhanced value of water contact angle close to superhydrophobic value of 151°. Data obtained from different studies infer the developed benzoxazines can be considered for water repellent and microelectronics insulation applications.

Keywords Photocurable benzoxazine · Resveratrol · Cashew nut shell cake ash · Low dielectric constant · Hydrophobic behavior

Introduction

An increasing demand in developing environmentally friendly and cutting-edge materials has been witnessed recently in the fields of materials science and chemistry [1–4]. Researchers are looking for alternative sources for chemical feedstocks as a result of growing concern over the sustainability of the environment and the depletion of fossil fuels [5, 6]. For the synthesis of sustainable materials, biophenols, which can be made from biomass and renewable resources like lignin, offer a promising route [7–11].

The class of thermosetting polymers known as benzoxazines is distinguished by its superior mechanical and thermal characteristics [12–14]. Due to their adaptability and simplicity in synthesis, they have attracted a lot of interest.

✉ Balaji Krishnasamy
balaji.psgtech@yahoo.co.in

✉ Hariharan Arumugam
hariharannalan@gmail.com

✉ Alagar Muthukaruppan
muthukaruppanalagar@gmail.com

[1] Polymer Engineering Laboratory, PSG Institute of Technology and Applied Research, Coimbatore 641 062, India

Published online: 21 March 2024

However, conventional benzoxazines are mostly synthesized from synthetic phenols obtained from petroleum sources, which are fast depleting non-renewable resources and significantly contributes to environmental pollution [15, 16]. This issue is addressed by facilitating the production of benzoxazines from bio-phenols by using renewable feedstocks [17, 18]. The problems of resource depletion and environmental impact can be significantly reduced by using bio-based precursor materials. One such field of study focuses on the production and application of photocurable benzoxazines produced from renewable bio-phenols [19, 20]. Particularly in the field of development of materials with low dielectric constant and photoluminescence behaviour, these materials expected to possess distinctive features and applications [21–23].

A sub-class of benzoxazines known as photocurable benzoxazines possesses a unique behaviour of undergoing photo reaction when exposed to ultraviolet (UV) or visible light. Functional groups can be introduced into photocurable benzoxazines to impart materials with photoluminescent characteristics. Such materials can be considered as useful in the field of optoelectronics, displays, and sensors [24, 25]. This feature has a number of benefits, such as precise control over curing procedures, limited energy usage, and possible material for 3D printing applications [26, 27].

By combining low dielectric constants with environmentally benign qualities, the development of renewable bio-phenol-based photocurable benzoxazines satisfy the requirement of micro-electronics insulation applications. Proper waste management ensures that bio-based products, derived from renewable resources, are utilized efficiently and sustainably, minimizing their impact on the environment [28–30]. This method paves an avenue to utilize bio-based precursors towards the development of cutting-edge materials from sustainable raw materials with protection of environment from adverse pollution problems.

Bio-wastes, preferably organic waste from agricultural residues and industrial sources, can be effectively reutilised and transformed into value added products, thereby reducing its impact on landfills and the environment. In accordance with transforming bio-wastes into the valuable products, bio-derived fillers are used in the form of silica, carbon, etc., to enhance certain properties of the materials according to their end use as insulators, semiconductors, high k and low k materials [31–33].

Low dielectric materials are essential in modern electronics and telecommunications to minimize signal loss, improve device performance, and reduce power consumption [34–36] Researchers continue to explore the synthesis routes, properties, and applications of these innovative materials, paving the way for a more sustainable and technologically advanced future. The recent advancements in the field of materials science and chemistry led to the development of bio-based benzoxazines for diverse applications. Some of the researchers focused on utilization of resveratrol, which is a naturally occurring polyphenol found in various plant sources, primarily in the skins of grapes, red wine, and some other fruits. It is often associated with potential health benefits due to its antioxidant properties. Only few literatures related to resveratrol-based benzoxazines have been reported [37–40].

In the current study, the photochemical and fluorescence properties of resveratrol derived bio-based benzoxazines have been reported for superhydrophobic and low dielectric applications. Five bio-based amines leelamine - derived from the bark of pine trees, octadecylamine (or stearlyamine) - fatty acid (stearic acid) derived from beef tallow, dodecylamine (or lauryl amine) – derived from lauric acid through coconut oil, oleylamine – derived from oleic acid through olive oil, furfurylamine – derived from corncob were used for the synthesis of bio-based benzoxazines [41–43]. To the best of our knowledge, the present work is considered as first of its kind to assess the photo responsive behaviour namely photo-isomerisation of bio-based trifunctional benzoxazines developed from resveratrol. These photocurable benzoxazines were characterised by spectral and thermal analyses. Further, the bio-ash (CNSA) derived from the oil extracted (waste) cashew nut shell (CNS) cake has been reinforced with benzoxazine matrix to obtain bio-composites. The CNSA reinforced composites possess an enhanced thermal stability and hydrophobicity with significantly low values of dielectric constant than those of neat benzoxazine matrix. Data obtained from different studies are discussed and reported.

Experimental

Materials

Resveratrol (CAS. No.501-36-0) and leelamine (CAS. No.1446-61-3) was purchased from BLD Pharmatech, India. Octadecylamine (CAS. No.24-30-1), dodecylamine (CAS. No.124-22-1) and oleylamine (CAS. No.112-90-3) was obtained from Alfa Aesar, India. Furfurylamine (CAS. No.617-89-0), 1,4-dioxane (CAS. No.123-91-1) and ethyl acetate (CAS. No.141-78-6) was procured from SRL, Mumbai. Sodium sulphate anhydrous (CAS. No.7757-82-6) was received from Sigma-Aldrich, India.