



Sustainable functionalized bio-silica and carbon reinforced 2,2'-Diallylbisphenol-A epoxy-based composites: high-k dielectrics

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Abstract

Advancing epoxy composite technology requires innovative matrices and functional fillers. This study introduces a system formulated from a tetra functional epoxy resin (DABA-EE), synthesized exclusively from 2,2'-diallyl bisphenol-A (DABA) via a dual epoxidation approach targeting both phenolic and allyl groups, as confirmed by NMR and FTIR analyses. The resin was cross-linked with 1-(3-aminopropyl)imidazole (API), and a series of composites was prepared by reinforcing the matrix with sustainable, high-performance materials. These reinforcements/fillers were functionalized bio-silica from Bermuda grass (FBS), activated bio-carbon from Palmyra sprouts (FAC), and graphene (G). A comparative analysis of the resultant polymer composite materials (DABA-EE-API-FBS/FAC/G 5–25) versus the pristine epoxy polymer (DABA-EE-API) demonstrated substantial improvements. We investigated the effect of these fillers on thermal and dielectric properties of the composites. All of the composites had surfaces that repelled water, with contact angles against water greater than 100°. The graphene-based composite DABA-EE-API-G25 exhibits an extremely high dielectric constant of 100.02 at 1 MHz, due to a greater number of charges being built up at the graphene-epoxy interfaces.

Highlights

- Tetra-functional DABA-EE epoxy synthesized via dual epoxidation.
- Graphene, epoxy functionalized bio-silica and bio-carbon used as fillers.
- Developed composites show hydrophobic surfaces (contact angle > 100°).
- Graphene-epoxy interfaces enable strong charge accumulation.
- The fillers contribute in the enhancement of thermal and dielectric properties.

Keywords 2,2'-diallylbisphenol-A · Epoxy · Bio-silica · Bio-carbon · Graphene · Hydrophobicity · Dielectric constant

Introduction

Epoxy resin is a highly promising and versatile reactive polymer known for its excellent adhesive properties, mechanical strength, chemical resistance, and electrical insulation. [1, 2]. When epoxy resins cure, they cross-link with hardeners to form a thermoset material that is very strong and stable and is free of volatile by-products. [3]. They protect electronic parts from moisture and dust and provide electrical insulation. This makes them essential for the production of printed circuit boards and electrical devices. [4, 5]. Jungang et al. studied the synthesis of epoxy resin from bisphenol-A and bisphenol-S using epichlorohydrin, examining reaction kinetics via alkali consumption and epoxide values. Dehydrochlorination happened faster with bisphenol-A than with bisphenol-S, which showed that bisphenol-A was more reactive. The reactions of

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phenol with epichlorohydrin and with the terminal epoxide both showed second-order kinetics, with rate constants and activation energies calculated. [6]. Transforming allylic C=C bonds into epoxy groups is a crucial process in the creation of polymeric structures capable of enduring elevated temperatures. The change in structure makes crosslinking perform better, which provides the materials with more strength and thermal stability [7, 8]. Meta-chloroperoxybenzoic acid (m-CPBA) was commonly utilized as an oxidizing agent for the selective epoxidation of alkenes, including allyl groups. When m-CPBA reacts with the carbon–carbon double bond, it facilitates the formation of epoxide relatively at low temperatures. This process is very useful because it is very stereoselective and regioselective. This means that it is an effective way to make intermediates that contain epoxides and can be changed chemically later [9, 10].

Fillers/reinforcements play an essential role when added to epoxy resin, altering its properties and improving the composite's performance, such as thermal resistance, mechanical strength, and dimensional stability [11, 12]. In addition, the effectiveness of the materials depends on the dispersion of the fillers within the epoxy matrix and the strength of the interfacial bonding between the filler particles and the resin [13]. To date, many studies have highlighted that numerous efforts have been made to enhance the characteristics of epoxy composites by incorporating a range of fillers (such as silica, Al_2O_3 , TiO_2 , fly ash, and bio-carbon) to improve the performance and applicability of this versatile matrix [14–17]. The growing need for eco-friendly, sustainable, and high-performance materials has led to the development of composites that use sustainable fillers, offering advantages such as low cost, biodegradability, and reduced environmental impact compared to synthetic alternatives. Previous studies have demonstrated that biogenic fillers enhance the thermomechanical properties of the composites. Fiore et al. investigated the epoxy composites reinforced with economical *Arundo donax* filler, analyzed various particle sizes and weight percentages (5% 10%, and 15%). The findings indicate that increased filler content enhances stiffness, whereas strength diminishes due to inadequate filler-matrix bonding. This highlights the need for surface treatments to improve interfacial adhesion in natural filler composites [18]. Surface modification of fillers plays a vital role in enhancing the interaction between the filler and polymer matrix in composite materials. Silane coupling agents (APTES, APTMS, GPTMS, etc.) are commonly used to bond with inorganic fillers, such as silica, chemically, and to add functional groups that can react with the polymer matrix. [19–21]. These treatments facilitate improving the filler dispersion, reducing agglomeration, and strengthening the adhesion at the interface. Recent advances in epoxy composite design have emphasized understanding how network architecture and

interfacial interactions govern performance. *Abdollahi* et al., demonstrated that polyetheramine modification significantly alters crosslinking density and glass transition temperature in epoxy networks providing a framework for understanding how flexible amines segments influence dielectric behaviour [22]. For silica-reinforced systems, *Katyama* et al., showed that silica nanofillers suppress molecular motion and reduce dielectric loss compared to other inorganic fillers, attributed to strong interfacial between silanol groups and the epoxy matrix [23]. These insights suggest that filler surface chemistry and network structure jointly control response in epoxy composites.

Dielectric materials are electrical insulators that limit the flow of electricity when they come into contact with an electric field. When they become polarized, they store energy, which makes them great for things like capacitors and circuit boards. Recent advancements, such as high-k dielectrics, have garnered attention for enhancing these materials, resulting in smaller and more efficient electronic devices [24–26]. Amorphous carbon, graphite, and carbon nanotubes (CNTs) are traditional carbon-based additives that have been used for a long time to improve electrical, thermal, and mechanical properties. Graphene, on the other hand, possesses properties that make it a very effective nanofiller for polymer composites, including its large surface area, high aspect ratio, and excellent intrinsic properties that enhance its performance [27, 28]. Graphene is a revolutionary material, composed of a single layer of carbon atoms arranged in a hexagonal lattice. Its remarkable strength, outstanding electrical conductivity, and exceptional thermal characteristics have garnered substantial interest in academic research and a diverse range of industrial applications [29]. Meanwhile, using an activated carbon filler yields a dielectric constant of 8.43 at 25 wt% [30]. On the other hand, epoxy resin is an attractive host due to its thermal–mechanical stability and wide industrial applications. However, their inherent low dielectric constant limits their capacitive density. Controlled interphase formation with functionalized fillers is therefore essential to maximize the relative permittivity of the dielectric materials. *Min* et al. formulated a new epoxy composite by evenly dispersing graphite nanosheets (GNPs) throughout the material. This approach provides the material with a high dielectric constant (>230) and improved thermal conductivity ($0.54 \text{ W/m} \cdot \text{K}$) at just 1.892 vol%. These composites also have a dielectric constant above 100 even at high frequencies (10^5 – 10^6 Hz), with optimal performance, and they lose less energy at 0.949 vol. % near 10^4 Hz. The oxygen-functionalized surfaces of GNPs form a strong interface, enabling the composite to work better [31]. However, the mechanisms underlying polymer-filler interactions are not well understood. In this work, we explore interfacial strategies of different fillers/reinforcements that enable high-k performance without compromising reliability

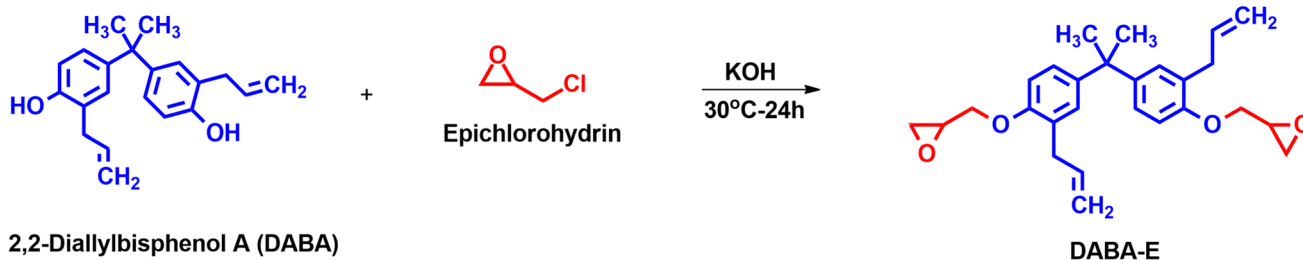
by systematically comparing filler surface chemistry, microstructure, and broadband dielectric response.

This study focuses on the synthesis of a tetrafunctional epoxy resin via a two-step process for application in high-performance, sustainable composites. The synthesis commenced with the reaction of 2,2'-diallyl bisphenol-A (DABA) and epichlorohydrin, yielding a bis-epoxy resin (DABA-E). Then, the bis-epoxy resin was further epoxidized using mCPBA as an oxidizing agent to transform its allyl groups into corresponding epoxy rings, making a tetra-functional epoxy monomer (DABA-EE). The resulting tetra epoxy monomer was cured with 1-(3-aminopropyl)imidazole (API) at room temperature and thermal treatment. To engineer multifunctional properties, this base matrix was integrated with three distinct fillers: functionalized bio-silica from Bermuda grass (FBS), functionalized activated bio-carbon (FAC) from Palmyra sprout, and high-performance graphene (G) to tailor the composite's inherent properties. The thermomechanical stability, hydrophobicity, and dielectric behavior of the resultant composites were investigated, and the results have been disclosed.

Materials and characterizations

Materials

Analytical grades of 2,2'-diallyl bisphenol-A (BLD Pharma), epichlorohydrin (Sigma-Aldrich), meta-chloroperoxybenzoic acid (Sigma-Aldrich), ethyl acetate (BLD Pharma), 3-glycidypropyl trimethoxysilane (GPTMS) (SRL Chemicals), dichloromethane (BLD Pharma), 1-(3-aminopropyl)imidazole (API) (SRL Chemicals) were purchased from their respective chemical agencies and were used as received without any further purification. The graphene (surface area of 600 m²/g) was purchased from Terracarb Pvt. Ltd., Coimbatore, India. For the preparation of bio-silica, Bermuda grass was collected from the local area of Coimbatore district, Tamil Nadu, India. For bio-carbon, Palmyra sprouts (*Borassus flabellifer*) were obtained from a farmhouse in the agricultural region of Salem district, Tamil Nadu, India.



Scheme 1 Synthesis of DABA-E

Synthesis of DABA-E (bi-functional epoxy of 2,2'-diallyl bisphenol-A)

In a 250 mL two-necked round-bottomed flask, 5 g (0.0718 mol) of 2,2'-diallylbisphenol-A, 16.49 mL (0.1783 mol) of epichlorohydrin, and 4 g (0.0713 mol) of KOH catalyst were taken. The reaction mixture was stirred at 30 °C for 24 h. After the completion of the reaction, the crude product was extracted using ethyl acetate and washed with water and brine. The organic portion was dried over sodium sulfate, and the solvent was removed under reduced pressure using a rotary evaporator, yielding a yellow resinous product (Yield: 91%). The schematic representation of the synthesis of DABA-E is shown in Scheme 1.

¹H NMR (400 MHz, CDCl₃) δ 7.03–6.95 (m, 4H), 6.71 (dd, J=7.9, 1.0 Hz, 2H), 5.95 (ddt, J=16.8, 10.1, 6.6 Hz, 2H), 5.04–4.95 (m, 4H), 4.18 (dd, J=11.1, 3.1 Hz, 2H), 3.94 (dd, J=11.1, 5.4 Hz, 2H), 3.35 (dt, J=6.6, 1.6 Hz, 4H), 3.32 (ddd, J=5.4, 2.7, 1.3 Hz, 2H), 2.85 (dd, J=5.0, 4.1 Hz, 2H), 2.74 (dd, J=5.0, 2.6 Hz, 2H), 1.61 (s, 6H).

Synthesis of DABA-EE (tetra-functional epoxy of 2,2'-diallyl bisphenol-A)

To synthesize DABA-EE, to a stirred ice-cold solution of 10 g (0.025 mol) of DABA-E in 100 mL of dichloromethane (DCM), 1.84 g (0.0107 mol) of m-chloroperoxybenzoic acid was added slowly over the course of 30 min, and the mixture was stirred at ice-cold temperature for 2 h to ensure optimal reaction. The crude product was then washed with distilled water, brine, and a 10% sodium bicarbonate solution to remove impurities. The organic layer was dried over sodium sulfate, collected, and processed in a rotary evaporator to remove the DCM solvent under reduced pressure, yielding a brownish, resinous product (Yield: 85%). The schematic illustration of the synthesis of DABA-EE is shown in Scheme 2.

¹H NMR (400 MHz, CDCl₃) δ 7.04 (d, J=6.7 Hz, 4H), 6.77–6.70 (m, 2H), 4.23 (dd, J=11.1, 3.0 Hz, 2H), 3.93 (ddd, J=11.1, 9.3, 5.6 Hz, 2H), 3.34 (dd, J=6.1, 3.2 Hz, 2H), 3.20–3.12 (m, 2H), 2.89 (q, J=4.4 Hz, 4H), 2.81–2.72 (m, 6H), 2.54 (td, J=5.3, 2.7 Hz, 2H), 1.63 (s, 6H).

In graphene-based composites, the range of frequency-independent DC conductivity extends to higher frequencies at lower filler concentrations. As the concentration increases, the AC conductivity rises to about 10^{-6} S/cm. As shown in Fig. 12c, within the measured frequency range and graphene loading (5–25%), the conductivity increases continuously, suggesting progressive growth of conduction pathways and no definite percolation threshold established. It is worth noting that space-charge polarization does not contribute much to the rise in conductivity. This graphene structure promotes electronic interactions between the graphene and the epoxy resin matrix, forming a stacked-layer interconnection network. This enhanced polarization leads to a higher dielectric permittivity but also limits the AC conductivity (σ_{ac}) at high frequencies. To quantitatively identify the possible percolation transition limit, the σ_{dc} is estimated from the low-frequency plateau region at different filler concentrations (Fig. S1) and used for percolation analysis. According to percolation theory, the scaling law describes this relationship. [75]

$$\sigma \propto (p - p_c)^t \quad (2)$$

where p_c is the percolation threshold; this model explains the sharp rise in conductivity that occurs when the filler concentration (p) exceeds p_c . The corresponding conductivity transition regime was analyzed using the percolation power-law expression,

$$\log \sigma = A + t * \log(p - p_c) \quad (3)$$

where (σ_{dc}) is the electrical conductivity, (p) is the graphene concentration, (p_c) is the percolation threshold, (A) is a proportionality constant, and (t) is the fitted critical exponent. The fitted log–log plot shown in Figure S2(a & b) reveals that the extracted p_c and t values are strongly dependent on the fitting range, and no unique percolation exponent was observed. The fitted A , p_c , and t values are given in Table S1. As seen, the t values gradually change by varying the p_c values within the selected fitting range. For the highest correlation, $R^2=0.9999$, the t value reached 1.2, which is lower than that of the universal 3D percolation network [76]. However, the nonuniversal t -value is also reported for graphene/polymer composites, with values between 1–1.3 being reported for 2D [77, 78]. These results indicate that the transport phenomena do not follow the unique percolation limit over the measured graphene range (5–25%). The obtained t values are effective exponent, should be influenced by the fitting range, partial network formation, tunneling, and limited data points [79, 80]. Therefore, the highest correlation coefficient alone is not sufficient to ensure a unique percolation threshold for graphene-based

polymer composites [80]. This result further concludes that, a unique percolation threshold may be established over the range of 25%, which start to exhibits the characteristic of interconnected conductive network.

Conclusion

A new tetra-functional epoxy resin (DABA-EE) was developed by a dual epoxidation process, and 1-(3-aminopropyl) imidazole was used as a curative agent (DABA-EE-API). This base epoxy resin matrix was reinforced with the functionalized bio-silica (FBS), functionalized activated bio-carbon (FAC), and graphene (G) reinforcements to develop a composite, and the dielectric properties were observed through the specific surface interaction. The BET surface area of the FBS, FAC, and G are 9.40, 28.37 and 600 m²/g respectively, indicating that the methods of activation, functionalization, and chemicals were used significantly to influence their surface area, which contributes to an increased value of dielectric constant by the space charge accumulation and enhancing polarization. The addition of increasing weight percentages of these distinct fillers into the epoxy resin matrix increases its dielectric, thermal, and hydrophobic behavior of the composites. The reinforced composite has contributed to enhancing the thermal stability up to 253 °C with improved char yield of the composites, also modified the surface of the composites into a hydrophobic behavior in nature. From the observed dielectric constant values, the addition of increasing weight percentages of these distinct fillers into the epoxy resin matrix increases its dielectric constant value. The composite DABA-EE-API-G25 achieved a remarkably high dielectric constant of ($k=100.02$) due to intense charge buildup at the interfaces between the graphene and the polymer matrix. In contrast, the composites with functionalized activated bio-carbon DABA-EE-API-FAC25 ($k=8.99$) and bio-silica DABA-EE-API-FBS25 ($k=5.09$) showed more modest and comparable values, respectively. Their GPTMS treatment ensured excellent dispersion that primarily strengthens the epoxy's inherent ability to store charge through dipole polarization rather than forming conductive pathways. Therefore, the activation and functionalization of bio-silica, bio-carbon methods modified its structure, functional groups (OH, CHO, C=O, COOH, etc.), porosity, and surface area, which influences the dielectric, thermal, and surface properties of the composites. Accordingly, the resulting materials may be suited to applications such as EMI shielding, microwave absorption or resistive switching devices where the energy dissipation is either tolerable or desirable, while their room temperature stability further supports their use in printed circuit boards and various niche applications.

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Data availability The data that support the findings of this study are available from the corresponding author upon reasonable request.

Declarations

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

Conflict of interest The authors declare no conflict of interest.

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