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Research Article

Influence of Hydroxyl-Terminated Polybutadiene (HTPB) on the Mechanical and Electrical Properties of Epoxy Phenol Novolac Resin

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ABSTRACT

Epoxy phenol novolac resin is among the most widely used matrix materials in the fabrication of composite structures**.** Incorporating elastomeric droplets into the matrix is one approach to enhancing the properties of composite materials. The effects of two hydroxyl-terminated polybutadiene (HTPB) types with hydroxyl values of 0.75 and 0.45 on the impact strength, toughness, tensile properties, and dielectric characteristics of an epoxy phenol novolac resin were investigated. The results demonstrated that a 2.5 wt% addition of the first HTPB type (hydroxyl value = 0.75) led to a 15% increase in impact strength, whereas the second type (hydroxyl value = 0.45) exhibited a negligible effect. Incorporating 2.5 wt% of the first and second HTPB types improved toughness by 108% and 56%, respectively. Both HTPB variants increased elongation at break but reduced tensile strength and modulus. Additionally, the inclusion of 2.5 wt% HTPB resulted in a 70% reduction in the dielectric constant. The difference in the behavior of the two HTPB types was attributed to their varying compatibility with the epoxy phenol novolac resin. Higher hydroxyl content (0.75 vs. 0.45) improved compatibility, leading to better interfacial adhesion and more uniform dispersion within the matrix.

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1. Introduction

Recent developments in composite science and technology have highlighted the significance of optimized additive content and processing parameters in tailoring and improving material performance [1-3]. One approach to enhancing the impact resistance and toughness of composites involves the incorporation of elastomeric droplets within the matrix. Hydroxyl-terminated polybutadiene (HTPB) and its derivatives can form an effective elastomeric phase in composite matrices, significantly improving mechanical properties—particularly impact resistance—through their low glass transition temperature and pronounced molecular mobility. Thomas et al. [4] investigated the tensile, flexural, and impact properties of epoxy resin modified with hydroxyl-terminated polybutadiene (HTPB) droplets. Their results demonstrated that incorporating 10 wt% of these elastomeric droplets enhanced the impact strength of the epoxy resin by 47%. The inherent poor compatibility between hydroxyl-terminated polybutadiene (HTPB) and epoxy resin has prompted numerous studies to improve

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interfacial adhesion. Ozturk et al. [5] addressed this compatibility issue by employing silane-terminated polybutadiene as an innovative interfacial modifier. This approach yielded a 30% enhancement in impact strength and a 10% improvement in tensile strength compared to unmodified systems. Latha et al. [6] enhanced the compatibility between epoxy resin and hydroxyl-terminated polybutadiene (HTPB) by chemically modifying HTPB with epoxy groups, enabling covalent bonding at the interface. Their study revealed that incorporating 10 wt% of this modified HTPB resulted in substantial mechanical property improvements: shear strength increased by 90%, impact strength surged by 127%, and tensile strength rose by 29%, demonstrating the effectiveness of this chemical modification approach in creating stronger interfacial bonds within the composite system. Yujia et al. [7] developed an effective strategy to enhance epoxy resin's mechanical properties by modifying polybutadiene resin with fatty acid dimer diisocyanate (DDI) terminal groups. This modification yielded remarkable results at 15 wt% loading: impact strength increased by over 112% compared to unmodified epoxy, while tensile and flexural strength exhibited only negligible reductions. Building on these approaches, Abdollahi et al. [8] further enhanced the epoxy/HTPB compatibility by chemically modifying hydroxyl-terminated polybutadiene to create amine-terminated polybutadiene. The results demonstrated that the epoxy resin modified with amine-terminated polybutadiene exhibited a toughness improvement more than twofold greater than that achieved with hydroxyl-terminated polybutadiene. Tian et al. [9] also examined the influence of amino-terminated polybutadiene molecular mass on the curing behavior, viscoelastic properties, morphology, and impact strength of epoxy resin. Their findings revealed that reducing the molecular mass of amino-terminated polybutadiene led to enhanced toughening behavior in the epoxy system. Barcia et al. [10] studied the influence of epoxy-terminated polybutadiene (ETPB) on the mechanical properties of epoxy resin. Barcia et al. demonstrated that epoxy-terminated polybutadiene (ETPB) synthesized via maleic anhydride modification exhibited superior impact strength compared to ETPB prepared through toluene diisocyanate modification. In contrast, Rafal et al. [11] revealed that the positioning of epoxy groups along the polybutadiene chain significantly influences its toughening efficiency in epoxy composites. Hosseini et al. [12] synthesized a novel epoxy-functionalized polyurethane by epoxidizing hydroxyl-terminated polybutadiene (HTPB), which they subsequently employed as a toughening modifier for epoxy resin. While much research has focused on epoxy systems, similar toughening approaches have been successfully applied to phenolic resins. Nirmal et al. [13] demonstrated that hydroxyl-terminated polybutadiene (HTPB) could improve the toughness of phenolic resins by leveraging its elastomeric properties. In their study, a resole resin served as a compatibilizer between the novolac phenolic matrix and HTPB elastomeric domains. Notably, fracture toughness increased only in blends containing up to 10 wt% HTPB, with higher elastomer content leading to reduced fracture energy, highlighting the importance of optimal phase morphology in toughening efficiency. Jackson et al. [14] systematically evaluated the toughening effect of HTPB in both neat phenolic resin and phenolic resin/sisal fiber composites. Their results revealed a 130% enhancement in impact strength for the pure phenolic resin modified with 10 wt% HTPB. In contrast, the composite system containing sisal fibers exhibited optimal performance at only 2.5 wt% HTPB, achieving a more modest 11% improvement in impact strength. Notably, the study demonstrated HTPB's dual functionality as both a toughening agent and an adhesion promoter between the phenolic matrix and sisal fibers. Jafari et al. [15] investigated the influence of hydroxyl-terminated polybutadiene (HTPB) on the mechanical performance of phenolic resin/carbon fiber composites. Their results demonstrated that composites containing 38.41 wt% carbon fibers and 7.5 wt% HTPB exhibited a 50% greater impact resistance compared to composites with identical carbon fiber content but only 2.2 wt% HTPB. This significant enhancement highlights the critical role of HTPB concentration in optimizing the impact properties of high-performance composites. Hydroxyl-terminated polybutadiene (HTPB) also demonstrates significant potential as a dielectric enhancer for telecommunication cable insulation and electronic component coatings. Zhou et al. [16] systematically investigated the dielectric properties of HTPB-modified epoxy resins, revealing two key effects: (1) a reduction in both dielectric constant and dissipation factor across a broad frequency range, and (2) a marked increase in surface and volume resistivity. These improvements were attributed to the inherent nonpolar character of HTPB, which contrasts with the more polar nature of the base epoxy matrix.

While hydroxyl-terminated polybutadiene (HTPB) has been widely studied as a toughening agent for conventional epoxy and phenolic resins, its effects on epoxy phenol novolac resin, a unique resin combining phenolic backbone

rigidity with epoxy-group reactivity, remain unexplored. Novolac epoxy resin is composed of epoxy groups bonded to a phenolic backbone. The epoxy groups enable strong adhesion and ambient-temperature curing, while the phenolic structure enhances thermal stability. [17-21]. Crucially, the role of HTPB's hydroxyl content in governing compatibility, phase morphology, and interfacial adhesion within this distinct matrix has never been investigated, despite its implications for optimizing mechanical and dielectric performance. Furthermore, prior research has not addressed how the dual chemical nature of epoxy phenol novolac influences elastomer dispersion and energy dissipation mechanisms. This study fills these gaps by systematically evaluating HTPB's structure-property relationships in epoxy phenol novolac resin, offering new fundamental insights for designing advanced composites with tailored toughness and electrical properties. This study systematically investigates the influence of two distinct hydroxyl-terminated polybutadiene (HTPB) variants - differing in hydroxyl value - on the mechanical and dielectric properties of epoxy phenol novolac resin, including impact strength, fracture toughness, tensile performance, morphological, and dielectric characteristics.

2. Materials and Methods

2.1. Materials

Two hydroxyl-terminated polybutadiene (HTPB) resin variants, differing in hydroxyl content (0.75 and 0.45 mmol KOH/gr HTPB), were used in this study. The Epoxy phenol novolac resin (LR-630) was commercially obtained from Kavian Composite Company (Iran). This two-component system employs an amine-based curing agent. For molding, RTV-2 silicone resin (cure mix ratio 100:3, Kavian Composite Company, Iran) was employed.

2.2. Mold Making

The silicone molds were fabricated using the following procedure: First, wooden master samples were prepared in four distinct geometries with dimensions of (i) 63.5 × 7.12 × 4 mm^3, (ii) 50 × 8 × 4 mm^3, (iii) 110 × 110 × 2 mm^3, and (iv) 250 × 30 × 4 mm^3 (length × width × thickness). A containment pool was constructed using cardboard walls adhered to a glass substrate with adhesive. The master samples were then secured to the glass base within this enclosure. The silicone molding compound (RTV-2) was prepared by mixing the base resin with the hardener in a 100:3 mass ratio. After thorough mixing for 5 minutes to ensure complete homogenization, the silicone mixture was carefully poured into the containment pool to encapsulate the master samples. The assembly was left to cure at ambient temperature for 24 hours until complete polymerization was achieved. Finally, the cured silicone mold was demolded by removing the containment structure and extracting the master samples, resulting in negative replicas of the original geometries.

2.3. Preparation of Samples

At this stage, epoxy phenol novolac resin was weighed and transferred into a container. Hydroxyl-terminated polybutadiene (HTPB) resin was then added at varying weight percentages based on formulation. Samples containing 2.5, 5, and 7.5 wt% of either Type I (OH value = 0.75 mmol KOH/g HTPB) or Type II (OH value = 0.45 mmol KOH/g HTPB) hydroxyl-terminated polybutadiene (HTPB) were prepared. The mixture was homogenized for 4 h using a mechanical mixer. Subsequently, 50 g of hardener was incorporated into the mixture and blended for an additional 5 min. The resulting mixture was carefully poured into silicone molds and allowed to cure at ambient temperature for 24 h. After demolding, the samples were post-cured in an oven at 100 °C for 4 h. The batch size was sufficiently large to ensure that all tested properties were evaluated on samples from the same batch, maintaining identical curing times and mixing conditions.

2.4. Methods

The Izod impact strength was determined in accordance with ASTM D256 using a Zwick 5102 impact tester (ZwickRoell GmbH, Germany) equipped with a 0.5 J pendulum. The test specimens had dimensions of 63.5 × 12.7 × 4 mm^3 (width × length × thickness). The impact fracture surface morphology was examined at 125× magnification using a Hitachi SU3900 scanning electron microscope (SEM). Due to electron density contrast between the phenolic-epoxy matrix and hydroxyl-terminated polybutadiene (HTPB) phases, which rendered the samples electron-opaque, the fracture surfaces were subjected to toluene extraction for 24 hours. This treatment selectively dissolved the elastomeric domains, creating porous structures that enabled clear SEM observation. Prior to imaging, all samples were sputter-coated with a 20 nm gold layer to enhance conductivity. Tensile strength was measured using a Santam STM-50 universal testing machine (Santam Engineering Design Co., Tehran, Iran) following ASTM D638 standard test methods. The dumbbell-shaped specimens had dimensions of 250 × 30 × 4 mm^3 (length × width × thickness), and the tests were conducted at a

crosshead speed of 1 mm/min. Fracture toughness was evaluated in accordance with ASTM D5045 using single-edge notched bend (SENB) specimens (50 × 8 × 4 mm^3) on a Santam STM-150 testing machine (Santam Engineering Design Co., Tehran, Iran). Testing was performed at a constant crosshead speed of 10 mm/min. The dielectric constant was measured in accordance with ASTM D150 using square specimens measuring 110 × 110 × 2 mm^3 (length × width × thickness). The tests were conducted at a 50 Hz frequency.

3. Results and Discussions

Figure 1 demonstrates the influence of hydroxyl-terminated polybutadiene type I (HTPB1) concentration on the impact strength of epoxy phenol novolac resin. As illustrated in Fig. 1, incorporation of 2.5 wt% Type I hydroxyl-terminated polybutadiene (HTPB1) resulted in a measurable improvement in impact resistance, with the epoxy phenol novolac resin's impact strength increasing from 12.05 to 13.86 J/m. The elastomeric droplets effectively dissipated fracture energy, thereby inhibiting crack propagation and preventing catastrophic failure in the composite material.

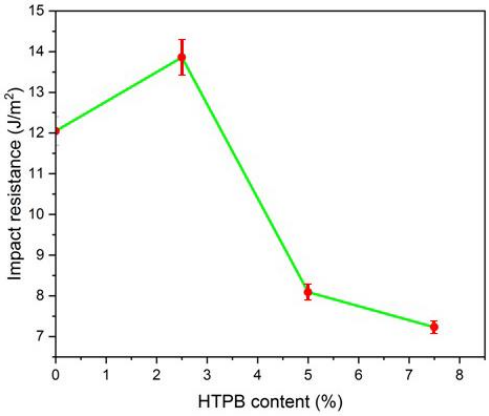


Fig. 1. Impact resistance of epoxy phenol novolac resin with varying concentrations of Type I Hydroxyl-Terminated Polybutadiene (HTPB1)

Mi et al. [22] systematically characterized the toughening mechanisms mediated by rubber particles, identifying four sequential energy dissipation processes:

(1) stretching and plastic deformation of the dispersed elastomeric phase,
(2) cavitation within the droplets,
(3) shear band formation in the surrounding matrix,
(4) development of multiple microcracks (crazes).

However, when liquid rubber is used as a modifier, the primary toughening mechanisms are cavitation of rubber droplets and shear yielding of the matrix [23]. Cavitation occurs when stress application forms voids in the HTPB droplets, concentrating stress in the surrounding epoxy phenol novolac matrix. This stress field initiates localized plastic deformation, creating shear bands near the droplets. Energy dissipates through void expansion, matrix yielding, and shear band propagation. Both mechanisms contribute simultaneously to toughness improvement [24-27]. However, when 5 wt% and 7.5 wt% of the first type of hydroxyl-terminated polybutadiene resin (HTPB1) were added, the impact strength of the epoxy phenol novolac resin decreased to 8.09 J/cm and 7.23 J/cm, respectively. This reduction indicates that adding HTPB1 beyond 2.5 wt% leads to diminished impact strength, likely due to droplet coagulation and non-uniform distribution within the epoxy phenol novolac matrix. The influence of varying amounts of hydroxyl-terminated polybutadiene (HTPB) resin on the impact strength of epoxy phenol novolac resin can be attributed to the role of droplet size in governing crack propagation. In mixtures containing low elastomer content (up to 2.5 wt%), crack growth energy is effectively dissipated through interactions with elastomeric droplets via the mechanisms previously described. However, at higher elastomer concentrations, droplet size increases while size distribution becomes less uniform. Consequently, crack growth energy bypasses the droplets without significant dissipation, leading to reduced impact resistance. Figure 2 presents the impact resistance of epoxy phenol novolac resin modified with varying amounts of hydroxyl-terminated polybutadiene type II (HTPB2).

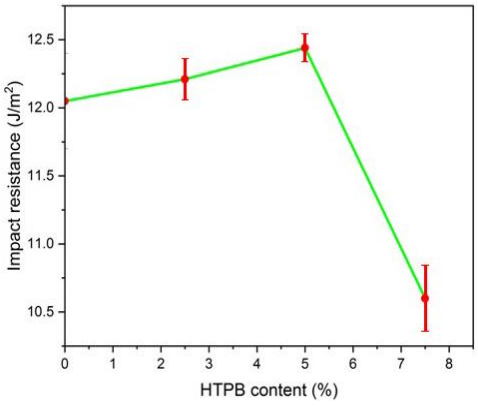


Fig. 2. Impact resistance of epoxy phenol novolac resin with varying concentrations of Type II Hydroxyl-Terminated Polybutadiene (HTPB2)

As shown, the addition of 2.5 wt% and 5 wt% HTPB2 resulted in minimal changes to impact resistance, with values of 12.21 J/m and 12.44 J/m, respectively. However, at 7.5 wt% HTPB2 loading, impact resistance decreased to 10.60 J/m due to droplet coagulation and non-uniform distribution within the epoxy phenol novolac

matrix. Thomas et al. reported a 47% enhancement in impact strength for an epoxy resin system when modified with an optimal 10 wt% hydroxyl-terminated polybutadiene (HTPB) [28]. In contrast, our study demonstrated a 15% improvement in impact strength for an epoxy phenol novolac resin system with only 2.5 wt% of type I HTPB (HTPB1). This significant difference in both the magnitude of improvement and optimal modifier concentration can be explained by fundamental differences in resin chemistry. Epoxy phenol novolac resins represent a distinct class of materials where phenolic resins are functionalized with epoxy groups through hydroxyl substitution on the phenolic ring. The incorporated epoxide groups enable ambient-temperature curing, forming a highly crosslinked network with superior mechanical properties compared to conventional epoxy resins. This structural difference accounts for both the lower optimal HTPB concentration required and the more modest impact strength improvement observed in our system.

Fracture surface morphology of impact-tested samples was analyzed using scanning electron microscopy (SEM). Figure 3 presents comparative micrographs (125× magnification) of: (a) unmodified epoxy phenol novolac resin, and (b-d) resin modified with 2.5, 5, and 7.5 wt% hydroxyl-terminated polybutadiene (HTPB1), respectively.

The pure epoxy phenol novolac resin (Fig. 3a) exhibited characteristic smooth, glassy fracture surfaces indicative of brittle failure. In contrast, HTPB1-modified samples (Figs. 3b-d) revealed a distinct two-phase morphology consisting of: (1) a continuous epoxy matrix and (2) dispersed elastomeric HTPB1 domains.

The 2.5 wt% HTPB1 composite (Fig. 3b) exhibited optimal morphology, with uniformly dispersed fine elastomeric particles.

At higher HTPB1 concentrations (5–7.5 wt%, Figs. 3c and 3d), the morphology exhibited distinct changes, including an increased population of larger droplets, a broader particle size distribution, and greater interparticle spacing. These morphological changes correlate with diminished toughening effectiveness, as evidenced by reduced performance in impact and toughness tests.

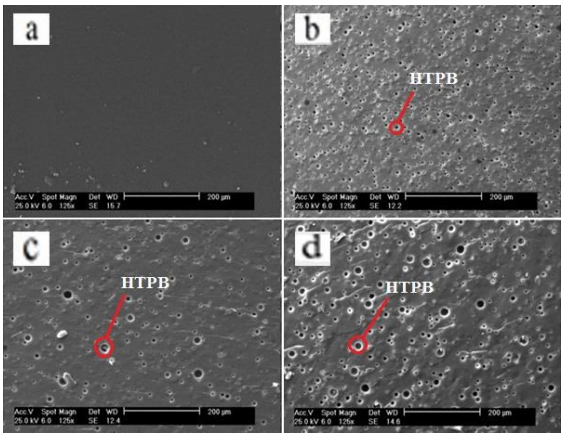


Fig. 3. Scanning electron microscopy (SEM) images of impact fracture surfaces for samples containing: (a) 0 wt%, (b) 2.5 wt%, (c) 5 wt%, and (d) 7.5 wt% hydroxyl-type I-terminated polybutadiene (HTPB1)

Quantitative image analysis was performed using ImageJ software to characterize the elastomeric droplet morphology (Figure 4). Using ImageJ software, the diameters of 100 droplets were measured, and the average was calculated for each sample with varying HTPB content. The results demonstrate a strong dependence of droplet size on HTPB1 concentration. When the HTPB1 content increased from 2.5 to 5 wt%, the average droplet area increased by a factor of two. A further increase in concentration to 7.5 wt% resulted in a greater than threefold expansion in average droplet area relative to the 2.5 wt% system. Scientific studies demonstrate that liquid elastomer droplet size critically controls toughening behavior: droplets below 100 nm resist cavitation, those between 100–500 nm undergo limited cavitation with partial shear yielding, while droplets in the 500 nm–5 μm range exhibit optimal performance through full cavitation and extensive shear yielding. However, droplets exceeding 5 μm reduce toughening effectiveness by transitioning from energy-dissipating phases to failure initiation points [23]. As the HTPB content increases, the average rubber particle diameter progressively exceeds the optimal range of 0.5-5 μm, resulting in diminished toughening effectiveness. Consequently, within the investigated composition range, the blend containing 2.5 wt% HTPB demonstrated the most favorable morphology and superior impact resistance properties.

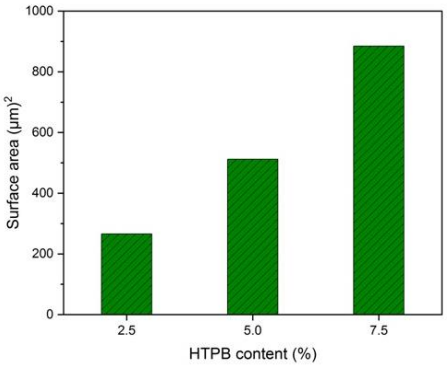


Fig. 4. Average elastomeric droplet area as a function of hydroxyl-terminated polybutadiene (HTPB) concentration

Figure 5 presents SEM micrographs (125× magnification) of fracture surfaces for both pure epoxy phenol novolac resin and its composites modified with 2.5, 5, and 7.5 wt% HTPB2. Microstructural analysis revealed two predominant trends: (1) progressive increases in HTPB2 content from 2.5 to 7.5 wt% resulted in substantially greater inter-droplet spacing between elastomeric domains, and (2) elevated HTPB2 concentrations facilitated pronounced droplet coalescence within the epoxy phenol novolac matrix. These morphological changes explain the limited improvement in impact resistance compared to the unmodified resin. The observed size growth of elastomeric domains at higher concentrations (7.5 wt%) directly correlates with reduced toughening efficiency, as larger droplets demonstrate diminished capacity for stress energy dissipation.

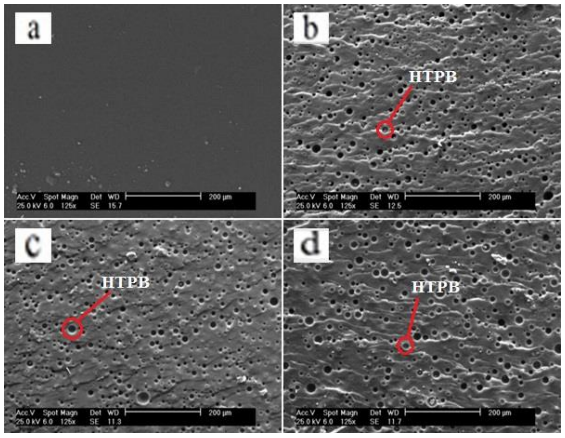


Fig. 5. Scanning electron microscopy (SEM) images of impact fracture surfaces for samples containing: (a) 0 wt%, (b) 2.5 wt%, (c) 5 wt%, and (d) 7.5 wt% type II hydroxyl-terminated polybutadiene (HTPB2)

Comparative SEM analysis of HTPB1 and HTPB2 modified resins demonstrates significant morphological variations at equivalent loading levels (2.5 and 5 wt%). The HTPB1-based systems consistently display (i) smaller average droplet diameters, (ii) narrower particle size distributions, and (iii) decreased inter-droplet spacing relative to their HTPB2 counterparts. These structural characteristics directly correlate with the observed mechanical performance, where HTPB1's finer dispersion morphology yields superior property enhancement compared to HTPB2. The coarser microstructure of HTPB2 composites, characterized by droplet coalescence and increased domain spacing, results in diminished toughening efficiency.

Fracture toughness tests were conducted on samples containing the first and second types of hydroxyl-terminated polybutadiene resin (HTPB1 and HTPB2), with a focus on specimens containing 2.5 wt% elastomer. As illustrated in Fig. 6, the fracture toughness of the pure epoxy phenol novolac resin was measured at 0.98 $MPa{\cdot}m^{1}/^{2}$. In contrast, the incorporation of 2.5 wt% HTPB1 increased the fracture toughness to 2.04 $MPa{\cdot}m^{1}/^{2}$, while the same concentration of HTPB2 resulted in a value of 1.53 $MPa{\cdot}m^{1}/^{2}$.

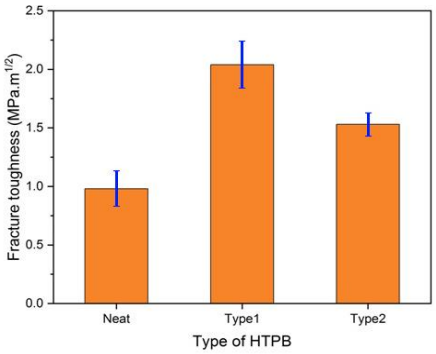
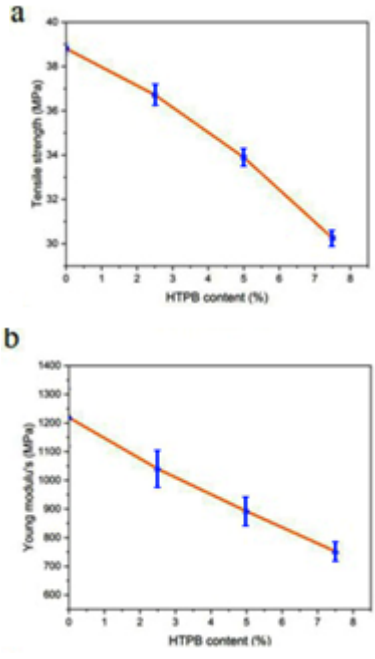


Fig. 6. Fracture toughness of epoxy phenol novolac resin modified with type I and type II hydroxyl-terminated polybutadiene (HTPB1 and HTPB2)

The modified blends containing hydroxyl-terminated polybutadiene resin type I (HTPB1) exhibited superior fracture toughness enhancement compared to those with type II (HTPB2). This improvement was primarily due to HTPB1's formation of smaller, more uniformly dispersed droplets and reduced inter-droplet spacing, which facilitated more effective energy dissipation and fracture resistance. Similarly, Abdollahi et al. observed a 111% improvement in fracture toughness upon incorporation of 5 wt% HTPB into the epoxy matrix [8]. This phenomenon was attributed to HTPB particles acting as stress concentrators, which promote shear localization within the matrix. Nirmal et al. reported a 57% improvement in fracture toughness for a Novolac phenolic base resin modified with 10 wt% hydroxyl-terminated polybutadiene (HTPB), using resole resin as a compatibilizer between the phenolic matrix and elastomeric HTPB droplets. In contrast, the present study demonstrated a more substantial 108% enhancement in fracture toughness for an epoxy phenol novolac resin system with only 2.5

wt% of type I HTPB (HTPB1). This significant improvement can be attributed to several key factors: First, the epoxy phenol novolac /HTPB1 system exhibited superior morphological characteristics, including smaller droplet size, more uniform distribution, and reduced interdroplet spacing compared to the Novolac/HTPB system reported by Nirmal et al. Second, the chemical structure of the epoxy phenol novolac resin provides inherent advantages - the epoxy groups actively participate in compatibilization by interacting with HTPB's hydroxyl groups, thereby promoting stronger interfacial adhesion than achievable in the Novolac/HTPB system.

The incorporation of hydroxyl-terminated polybutadiene resin (HTPB1) in the epoxy phenol novolac matrix resulted in a progressive reduction of tensile strength. As shown in Fig. 7a, the addition of 2.5, 5, and 7.5 wt% HTPB1 decreased the tensile strength from 38.81 MPa (neat resin) to 36.72 MPa, 33.90 MPa, and 30.25 MPa, respectively. This represents strength reductions of 5.4%, 12.6%, and 22.1% relative to the unmodified resin. The observed reduction in tensile strength in the presence of HTPB droplets is attributed to their role as stress concentrators. Since HTPB droplets possess lower tensile strength and modulus than the epoxy phenol novolac resin matrix, they create localized stress fields. Consequently, as the HTPB content increases, the tensile strength of the composite decreases [29, 30]. An additional factor, as proposed by Abdollahi et al., involves the reduction in crosslink density caused by hydroxyl-terminated polybutadiene (HTPB) droplets within the matrix [8].



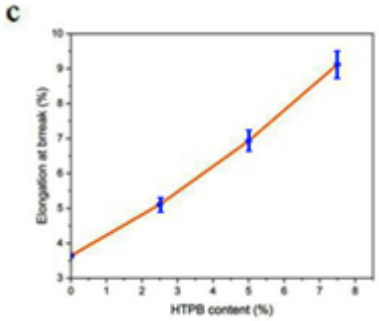


Fig. 7. Tensile strength, Young modulus, and elongation at break of resin modified with varying concentrations of hydroxyl-terminated polybutadiene (HTPB-Type 1)

The Young's modulus of the epoxy phenol novolac resin decreased progressively with the addition of hydroxyl-terminated polybutadiene type I (HTPB1), as illustrated in Fig. 7b. This reduction in stiffness reflects the increased ductility imparted by the elastomeric modifier. Such behavior is characteristic of thermoset resins modified with elastomers, where the soft, flexible nature of the additive lowers the overall rigidity of the system. Notably, at 7.5 wt% HTPB1 loading, Young's modulus decreased by 37% compared to the unmodified resin, demonstrating the significant influence of elastomer content on mechanical properties. The addition of type 1 hydroxyl-terminated polybutadiene resin (HTPB1) significantly improved the ductility of the epoxy phenol novolac resin, as evidenced by the elongation at break measurements (Fig. 7c). With HTPB1 loadings of 2.5, 5, and 7.5 wt%, the elongation at break increased from 3.64% (neat resin) to 11.5%, 6.93%, and 12.9%, respectively. This enhancement in strain capacity indicates increased material flexibility upon HTPB1 incorporation. The observed mechanical behavior can be attributed to HTPB1's role in reducing the crosslink density of the epoxy phenol novolac network. This structural modification explains both the improved ductility and the corresponding decreases in Young's modulus and tensile strength, as the less constrained polymer network allows for greater chain mobility while sacrificing some rigidity.

From the stress-strain curves of epoxy phenol novolac resin modified with varying amounts of hydroxyl-terminated polybutadiene type II (HTPB2), it is observed that adding 2.5 wt% HTPB2 slightly increases the tensile strength and Young's modulus while causing an imperceptible reduction in elongation at break (Fig. 8). However, at higher concentrations (5 wt% and 7.5 wt% HTPB2), the tensile strength decreases to 37.2 MPa and 35.12 MPa, respectively, and the Young's modulus drops to 1110 MPa and 964 MPa, respectively. Conversely, the elongation at break increases by 4.98% and 5.94%,

respectively. The results indicate that the reduction in tensile strength caused by adding hydroxyl-terminated polybutadiene elastomer type II (HTPB2) is less pronounced than that of type I (HTPB1). This difference arises because HTPB2 has lower hydroxyl content, leading to reduced compatibility with the epoxy phenol novolac resin.

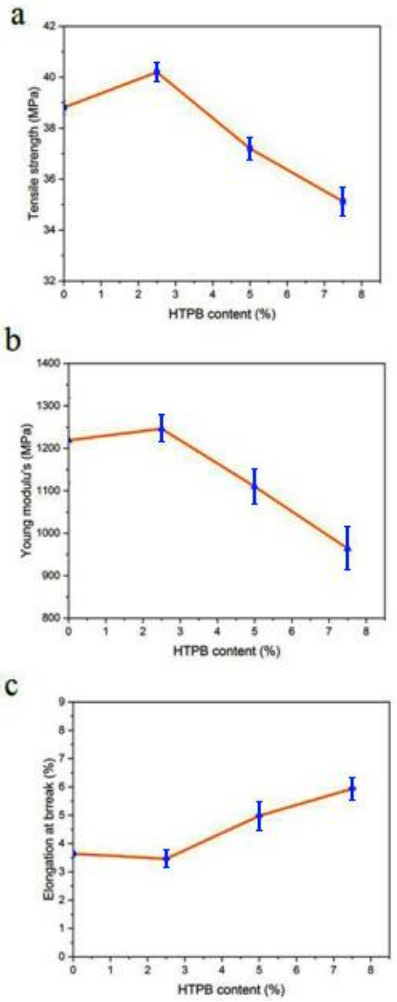


Fig. 8. Tensile strength, Young modulus, and elongation at break of resin modified with varying concentrations of hydroxyl-terminated polybutadiene (HTPB-Type 1)

We systematically investigated the dielectric properties of epoxy phenol novolac composites incorporating 2.5 wt% of two distinct hydroxyl-terminated polybutadiene (HTPB1 and HTPB2). Figure 9 summarizes the dielectric properties, revealing significant differences between the materials. The pure epoxy phenol novolac resin displayed a dielectric constant of 6.42 μF/cm, whereas the 2.5 wt% HTPB1 and HTPB2 composites exhibited substantially lower values of 1.88 and 2.06 μF/cm, respectively - corresponding to reductions of 70.7% and 67.9%. The dielectric constant quantifies a material's capacity to store electrical energy when subjected to an electric field. While epoxy phenol novolac resins inherently exhibit polar characteristics and consequently higher dielectric constants, the incorporation of hydroxyl-terminated polybutadiene (HTPB) - a nonpolar elastomer with low permittivity - induces multiple dielectric-reducing effects within the composite system. The observed reduction in dielectric constant arises first from a dilution effect, wherein the low-dielectric-constant HTPB component reduces the overall polarization of the high-dielectric-constant epoxy matrix. Additionally, the nonpolar nature of HTPB chains minimizes interfacial polarization at the HTPB-epoxy phenol novolac interface, resulting in suppressed Maxwell-Wagner-Sillars polarization effects. Moreover, the hydrocarbon backbone of HTPB restricts charge mobility and limits space charge accumulation within the composite material [31]. Notably, these results demonstrate superior dielectric reduction compared to Zhou et al.'s [16] reported a 19% decrease at 40 wt% HTPB loading in epoxy systems. Comparative studies demonstrate that HTPB modification more effectively reduces the dielectric constant of epoxy composites than nanoscale silica fillers [32]. A comparison of the dielectric constants obtained in this study with the datasheet values of commercial epoxy resins (~3.2 at 50 Hz and ambient temperature) reveals that incorporating hydroxyl-terminated polybutadiene as an additive significantly reduces the dielectric constant of the insulating resin. This finding highlights its potential utility in electrical insulation applications.

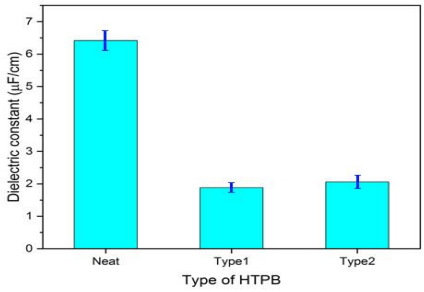


Fig. 9. Dielectric constant of epoxy phenol novolac resin and sample containing 2.5 wt% of hydroxyl-terminated polybutadiene (Type 1 or Type 2)

4. Conclusions

This study systematically investigated the influence of two hydroxyl-terminated polybutadiene (HTPB) elastomers—differing in hydroxyl values (0.75 vs. 0.45 mmol KOH/g)—on the mechanical and dielectric properties of epoxy phenol novolac resin. The findings revealed that the HTPB variant with the higher hydroxyl value (0.75) significantly enhanced the impact strength (15% increase) and fracture toughness (108% improvement) at an optimal loading of 2.5 wt%, while the variant with the lower hydroxyl value (0.45) exhibited only marginal improvements.

These results underscore the critical role of hydroxyl content in governing the compatibility, dispersion, and interfacial adhesion of HTPB within the epoxy phenol novolac matrix, as confirmed by scanning electron microscopy (SEM) analysis. Both HTPB types improved elongation at break but reduced tensile strength and Young's modulus, highlighting their effectiveness as flexibilizing agents. Notably, the addition of just 2.5 wt% HTPB led to a remarkable 70% reduction in the dielectric constant, a property highly advantageous for electrical insulations. This reduction was attributed to the non-polar nature of HTPB, which minimizes charge accumulation and interfacial polarization. The study not only advances the understanding of structure-property relationships in such systems but also highlights the potential of HTPB-modified resins for applications requiring balanced toughness, flexibility, and low dielectric constants, such as in advanced composites, electronic packaging, and insulation materials. Future studies should investigate the material's thermal behavior and environmental stability, including UV resistance and oxidative degradation characteristics, to enable comprehensive performance evaluation.

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Conflicts of Interest

The author declares that there is no conflict of interest regarding the publication of this article.

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