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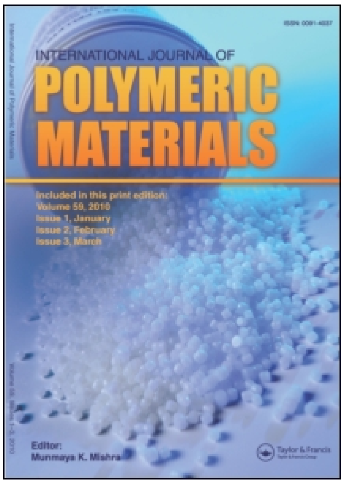

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Mechanical Properties of Bismaleimides Modified Polysulfone Epoxy Matrices

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Epoxy resin has been chemically modified using 4, 8, and 12% of bisphenol-A based polysulphone along with three types of bismaleimides, namely [N, N′-bismaleimido-4,4′-diphenylmethane (BMI-1), 1,3-bis (maleimido) benzene (BMI-2) and 1,1′-bis (4-maleimidophenyl) cyclohexane (BMI-3)]. The epoxy hybrid matrices developed, in the form of castings, were used to characterize their mechanical properties like tensile strength, tensile modulus, flexural strength, flexural modulus, impact strength, hardness, and dynamic mechanical analysis as per ASTM standards. Data obtained from mechanical studies indicate that the introduction of hydroxyl terminated polysulfone into epoxy resin enhanced the value of impact strength to the extent of 48% due to the formation of flexible graft structures. Similarly, the incorporation of bismaleimides into epoxy resin also improved both tensile and flexural behavior of epoxy resin. Further, the introduction of combination of both polysulfone and bismaleimides into epoxy resin improved the mechanical properties according to their percentage content. Among the bismaleimides-modified polysulfone epoxy matrices, the epoxy matrix modified with 8% polysulfone and 8% BMI-2 exhibited better mechanical properties than other modified epoxy matrices.

Keywords: bismaleimide, epoxy resin, fracture toughness, polysulfone, tan δ, tensile strength

INTRODUCTION

Epoxy resins are one of the most important classes of thermosetting polymers. The versatile characteristics of epoxy resins make them suitable for a number of industrial, engineering, and aerospace applications such as protective coatings, structural adhesives, electrical laminates, and matrices for advanced composite materials. Epoxy resins possess a

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number of useful properties like excellent chemical and solvent resistance, outstanding adhesion to various substrates, easy processability under various conditions, excellent dimensional stability and good mechanical properties [1–8]. However, the brittle behavior of cured epoxy resins, limited impact strength, inferior moisture, and weathering resistance restricts their utility for high-performance applications [9–24].

The chemical modification of epoxy resins using elastomer-based chemical modifiers results in improvement of thermal, mechanical, dielectric, and weather-resistant properties according to their nature and percentage incorporation. In the recent past, it was reported that thermoplastics have been used as chemical modifiers for thermosetting resins to improve their characteristic properties [25–38]. The prime objective of modification of thermosetting resins using high-performance thermoplastics with high *Tg* is to enhance the fracture toughness and impact strength in addition to an improvement in thermo mechanical properties.

An attempt has been made in the present investigation by incorporating varying percentages of hydroxyl terminated polysulfone and bismaleimides as thermoplastic and chemical modifiers, respectively, for epoxy resin to obtain hybrid epoxy matrices with improved characteristic properties, mainly toughness and thermomechanical properties to utilize them in various industrial and engineering applications for better performance and longevity.

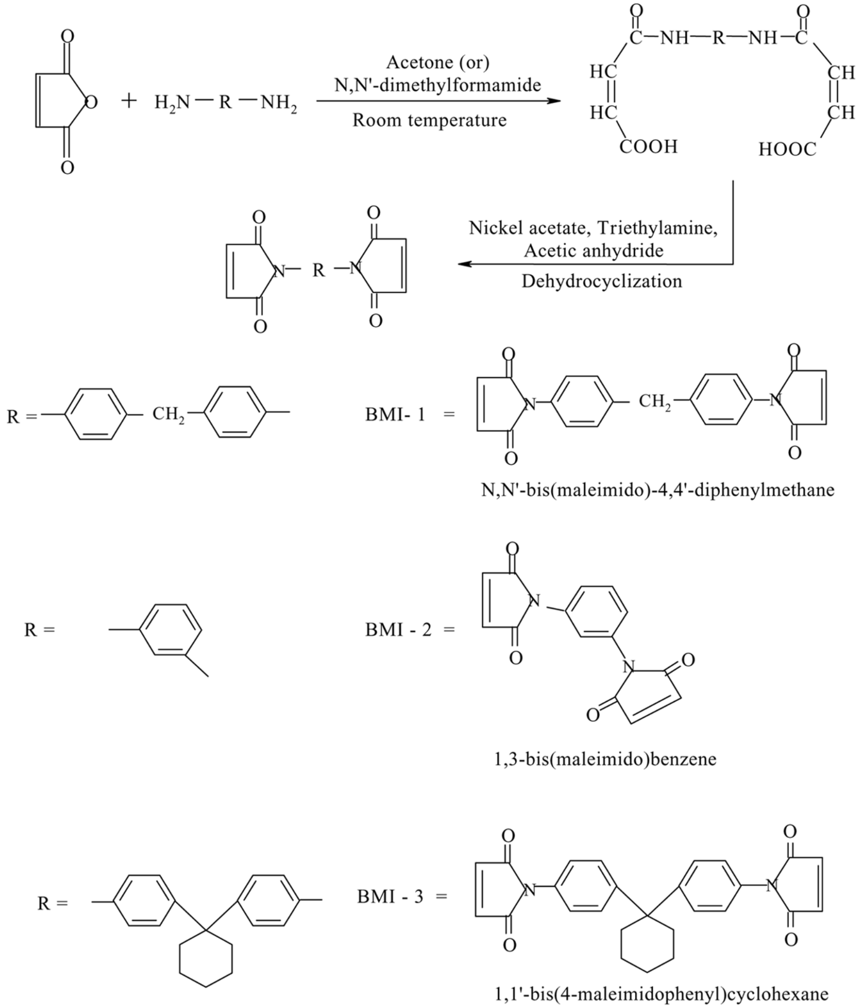
EXPERIMENTAL

Materials

Epoxy resin [diglycidyl ether of bisphenol A (DGEBA)] LY556 (epoxy equivalent about 180–190, viscosity about 10,000 cP) and 4,4-diaminodiphenylmethane (DDM) were obtained from Ciba-Geigy (India). Bismaleimides (Scheme 1), namely N, N′-bismaleimido-4,4′-diphenylmethane (BMI-1), 1,3-bis (maleimido) benzene (BMI-2) and 1,1′-bis (4-maleimidophenyl) cyclohexane (BMI-3), were synthesized by previously reported procedure [39]. Bisphenol A based Polysulfone ($M_w = 15000$, $Tg = 195°C$) obtained from Shauguang chemical factory, Shanghai, China was used as received.

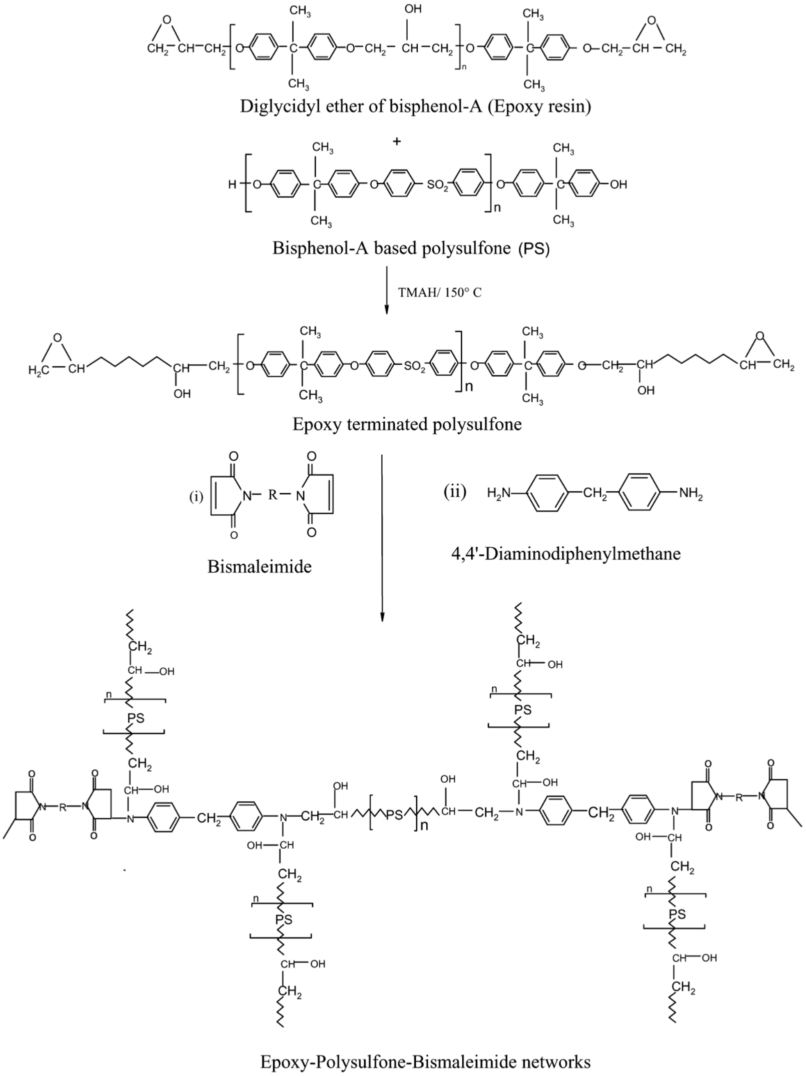
Preparation of Bismaleimide-Modified Polysulfone Epoxy Matrices

The hybrid polysulfone modified epoxy matrices were prepared by dissolving varying percentages (4, 8, 12 wt%) of bisphenol-A based



SCHEME 1 Synthesis of bismaleimides.

polysulfone in epoxy resin using tetramethylammonium hydroxide (TMAH) as catalyst with continuous stirring at 150°C for 2 h. The hybrid polysulfone modified epoxy matrix obtained was degassed under vacuum for half an hour and then cooled to 90°C. Further, the calculated amount of bismaleimide was dissolved in the hybrid polysulfone-epoxy resin under vigorous stirring, followed by the addition of stoichiometric amount of 4,4′-diaminodiphenylmethane curative (with respect to epoxy). The agitation was continued at 100°C until a homogeneous product was obtained. The product (Scheme 2) was subjected to vacuum to remove the trapped air and then cast and cured at 120°C for 3 h. The castings were then post cured at 180°C for 2 h and finally removed from the mold and characterized.



SCHEME 2 Formation of Bismaleimide modified Polysulfone-Epoxy—DDM graft polymer network structure.

TABLE 1 Mechanical Properties of Unmodified Epoxy, Polysulfone Modified Epoxy, Bismaleimides (BMI-1, BMI-2 and BMI-3) Modified Epoxy and BMIs Modified Polysulfone-Epoxy Systems

Sample code	Tensile strength (MPa)	Tensile modulus (MPa)	Flexural strength (MPa)	Flexural modulus (MPa)	Impact strength J/m
$E_{100}PS_0B_0$	61.2 ± 7	2713.7 ± 34	104.0 ± 8	1753.9 ± 38	98.3 ± 6
$E_{100}PS_4B_0$	61.8 ± 5	2723.2 ± 32	105.7 ± 7	1772.8 ± 35	122.0 ± 5
$E_{100}PS_8B_0$	62.3 ± 6	2736.9 ± 29	106.3 ± 6	1794.3 ± 31	132.9 ± 4
$E_{100}PS_{12}B_0$	63.1 ± 7	2752.6 ± 34	107.1 ± 4	1824.2 ± 28	146.1 ± 8
$E_{100}PS_8B1_4$	64.6 ± 3	2861.7 ± 34	112.4 ± 2	1896.8 ± 30	128.4 ± 6
$E_{100}PS_8B1_8$	66.5 ± 5	3079.4 ± 36	118.4 ± 4	1984.2 ± 32	123.7 ± 5
$E_{100}PS_8B1_{12}$	70.2 ± 4	3248.5 ± 31	123.8 ± 5	2103.8 ± 36	119.3 ± 3
$E_{100}PS_8B2_4$	65.1 ± 3	2902.7 ± 29	113.3 ± 4	2083.2 ± 27	125.5 ± 5
$E_{100}PS_8B2_8$	69.2 ± 5	3125.9 ± 31	119.5 ± 5	2242.3 ± 32	120.2 ± 4
$E_{100}PS_8B2_{12}$	71.8 ± 4	3274.3 ± 34	124.5 ± 3	2489.5 ± 35	116.4 ± 6
$E_{100}PS_8B3_4$	63.8 ± 4	2863.4 ± 32	110.5 ± 5	1872.7 ± 32	129.5 ± 5
$E_{100}PS_8B3_8$	65.9 ± 5	3012.7 ± 29	113.7 ± 6	1954.3 ± 28	125.3 ± 6
$E_{100}PS_8B3_{12}$	69.2 ± 6	3152.5 ± 27	117.5 ± 3	2082.5 ± 31	120.5 ± 4
$E_{100}PS_0B1_8$	74.0 ± 6	3233.3 ± 30	124.7 ± 5	2038.4 ± 30	90.2 ± 4
$E_{100}PS_0B2_8$	75.1 ± 3	3145.5 ± 31	129.2 ± 2	3098.2 ± 34	89.6 ± 2
$E_{100}PS_0B3_8$	70.8 ± 4	2912.7 ± 31	119.2 ± 4	2102.5 ± 31	93.5 ± 5

E—Epoxy; PS—Polysulfone; B—Bismaleimide.
B1-N,N′-bismaleimido-4,4′-diphenylmethane (BMI-1).
B2-1,3-bis (maleimido) benzene (BMI-2).
B3-1,1′-bis (4-maleimidophenyl) cyclohexane (BMI-3).

Test Methods

The IR spectra for unmodified epoxy, polysulfone modified epoxy and bismaleimides incorporated polysulfone modified epoxy matrices were recorded on a Perkin-Elmer (Model RX1) FT-IR Spectrometer.

The tensile strength was determined using dog–bone-shaped specimens according to ASTM-D3039, using an Instron testing machine (Model 6025; Instron, UK), at a crosshead speed of 10 mm/minute.

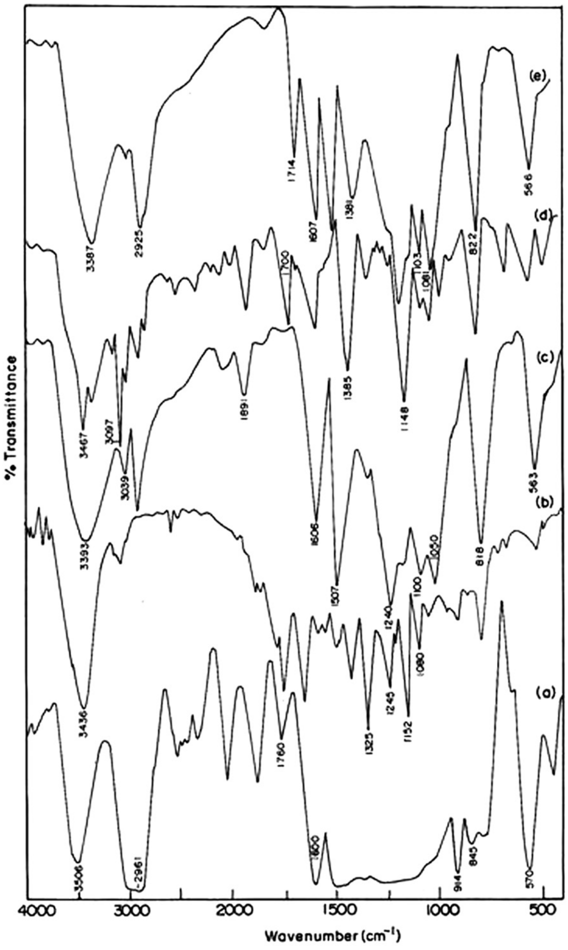


FIGURE 1 FTIR spectra of (a) unmodified epoxy, (b) polysulfone, (c) polysulfone modified epoxy matrix cured at 180°C, (d) bismaleimide (BMI-1), and (e) bismaleimide (BMI-1) modified polysulfone epoxy matrix.

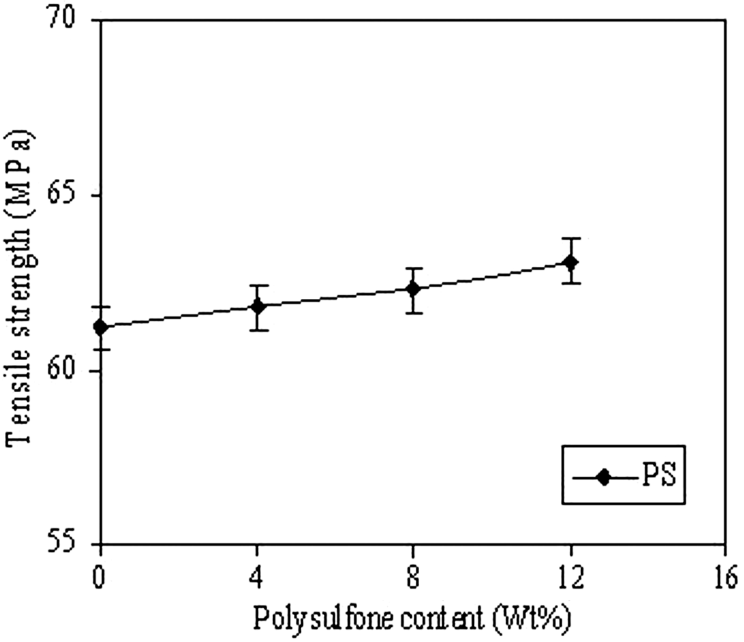


FIGURE 2 Effect of polysulfone content on tensile strength of epoxy systems.

The flexural properties were measured as per ASTM-D790, using an Instron testing machine (Model 6025 UK), at 10 mm/minute cross head speed. The Izod impact strength was evaluated as per ASTM D-256. Hardness of the specimens is measured using Durometer-Type D as per ASTM D2240. As many as five test pieces were used to generate the data points for the mechanical tests.

Dynamic mechanical analyses of the samples were measured by using NETZSCH DMA 242 dynamic mechanical analyzer. The experiments were conducted at dynamic strain of 0.3 and frequency of 1 Hz. The results are presented in Table 1 and Figures 1–12.

RESULTS AND DISCUSSION

IR Spectroscopy

The characteristic IR absorption peaks observed for polysulfone are shown in Figure 1. The IR absorption peaks for ether linkage, sulfonyl group, aryl group and –OH group present in polysulfone appear at 1325 cm^{-1}, 1245 cm^{-1}, 1080 cm^{-1} and 3436 cm^{-1}, respectively (Figure 1b). The epoxide ring of epoxy resin appears at 914 cm^{-1} (Figure 1a). The disappearance of

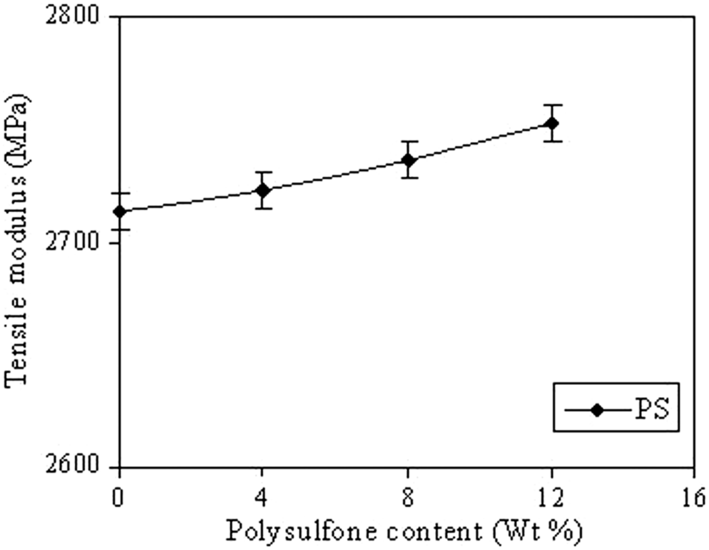


FIGURE 3 Effect of polysulfone content on tensile modulus of epoxy systems.

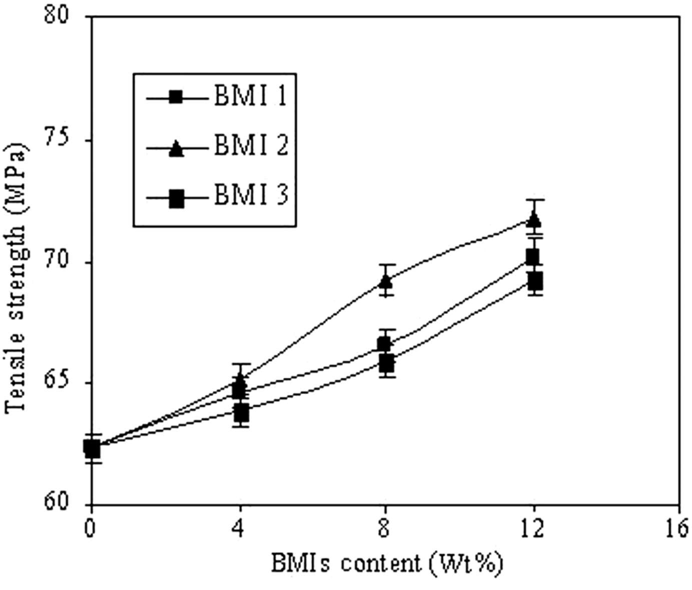


FIGURE 4 Effect of BMIs content on tensile strength of epoxy and polysulfone (8%) modified epoxy systems.

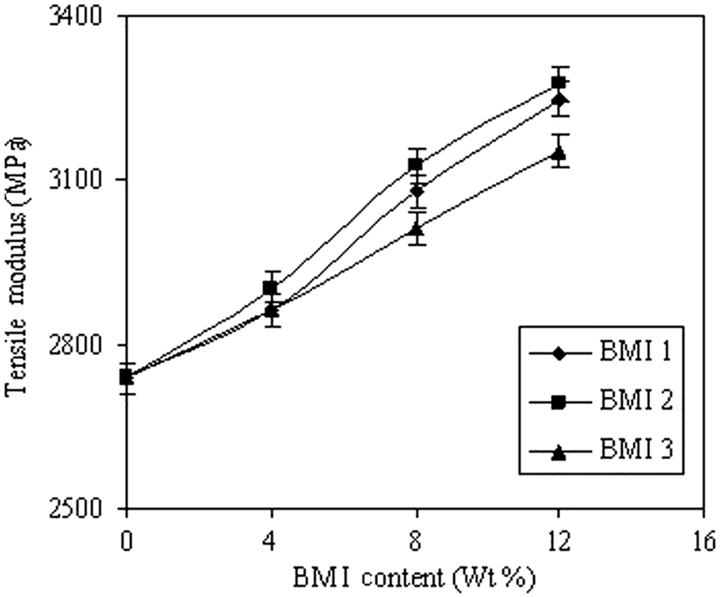


FIGURE 5 Effect of BMIs content on tensile modulus of epoxy and polysulfone (8%) modified epoxy systems.

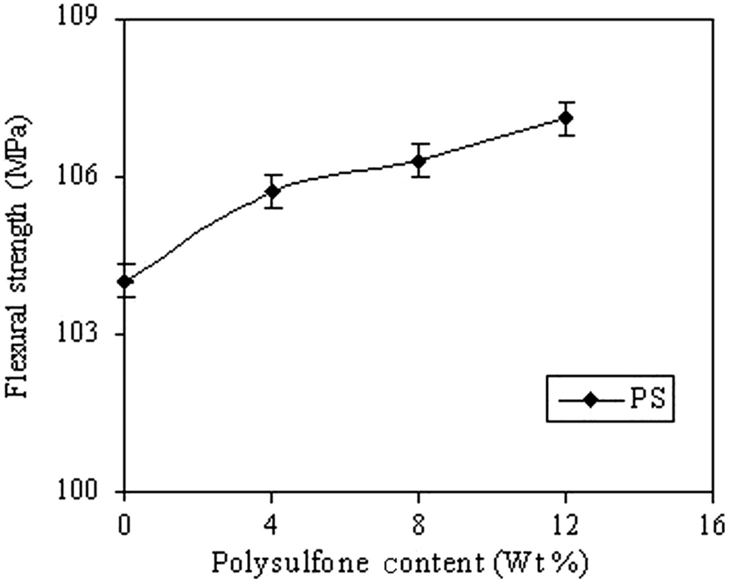


FIGURE 6 Effect of polysulfone on flexural strength of epoxy systems.

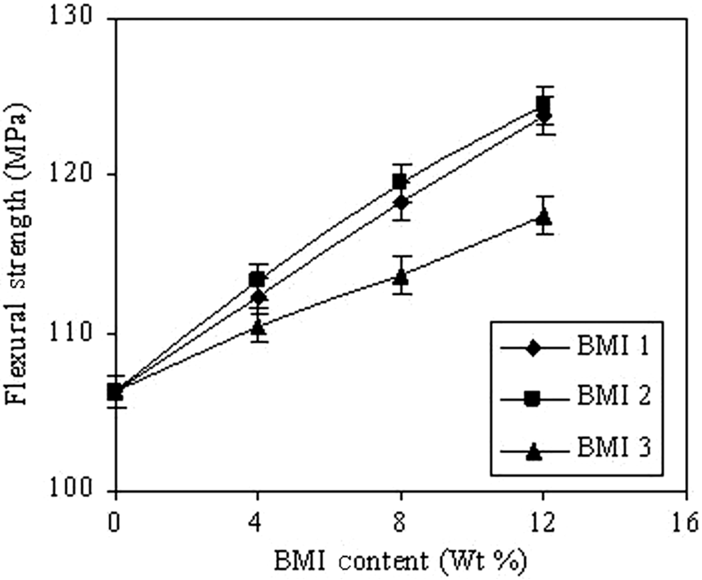


FIGURE 7 Effect of BMIs content on flexural strength of epoxy and polysulfone (8%) modified epoxy systems.

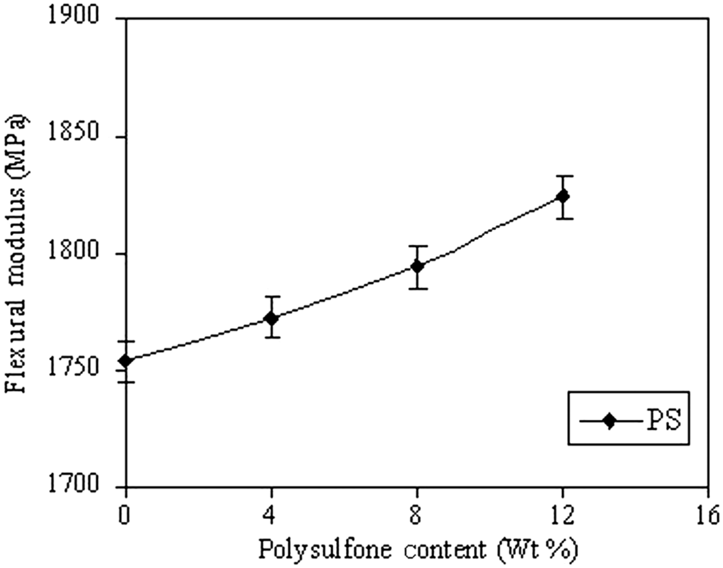


FIGURE 8 Effect of polysulfone content on flexural modulus of epoxy systems.

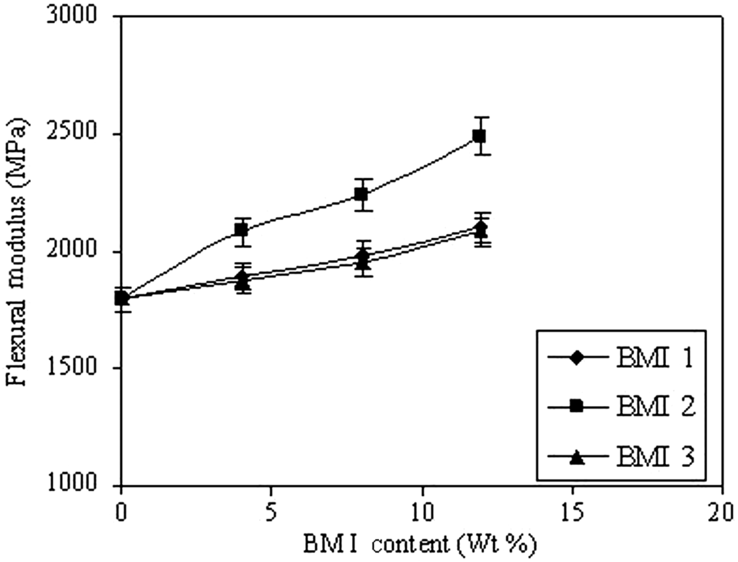


FIGURE 9 Effect of BMIs content on flexural modulus of epoxy and polysulfone (8%) modified epoxy systems.

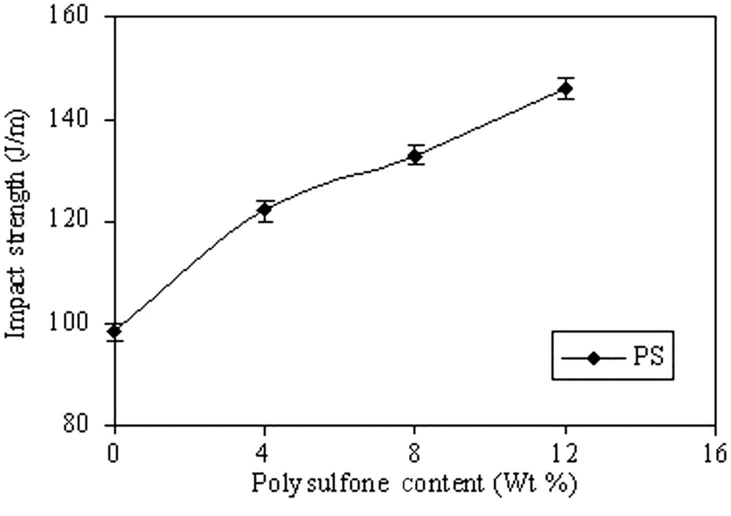


FIGURE 10 Effect of polysulfone content on impact strength of epoxy systems.

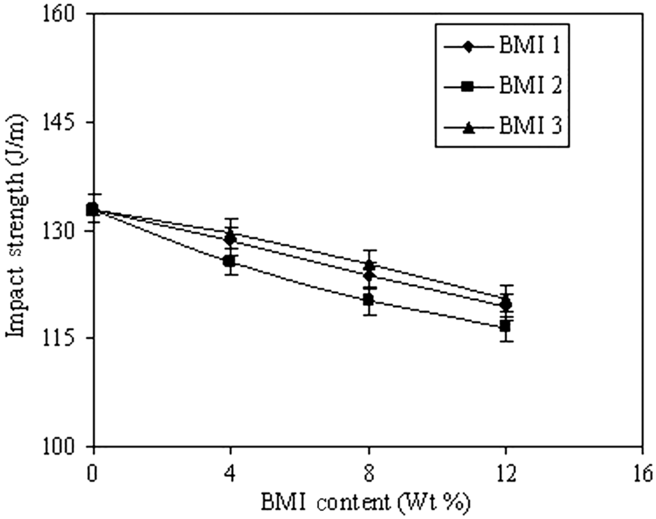


FIGURE 11 Effect of BMIs content on impact strength of epoxy and polysulfone (8%) modified epoxy systems.

IR absorption for oxirane ring of epoxy at 914 cm^{-1} (Figure 1c) was used to ascertain the occurrence of reaction between epoxy and polysulfone and the formation of graft polymer network structure (Scheme 2). The characteristic peaks at 3097 cm^{-1} and 1148 cm^{-1} indicate the presence of =C−H group and carbon-nitrogen-carbon linkage respectively in BMI-1 (Figure 1d). The disappearance of IR absorption peak at 3097 cm^{-1} was used to ascertain the reaction between epoxy and bismaleimide and the formation of network structure (Figure 1e).

Tensile Properties

Polysulfone Modified Epoxy Systems

The values of tensile strength of polysulfone modified epoxy matrices cured with DDM are presented in Table 1. It is noticed that the introduction of 4%, 8%, and 12% polysulfone (by wt%) into epoxy resin marginally enhances the values of tensile strength (Figure 2). The little improvement in the values of tensile strength is explained due to the inherent rigid aromatic molecular structure of polysulfone and the formation of graft network between epoxy and polysulfone. The values of tensile modulus obtained for the unmodified epoxy and polysulfone modified epoxy systems are also presented in Table 1 and they exhibit similar trend as observed in the case of values of tensile strength (Figure 3).

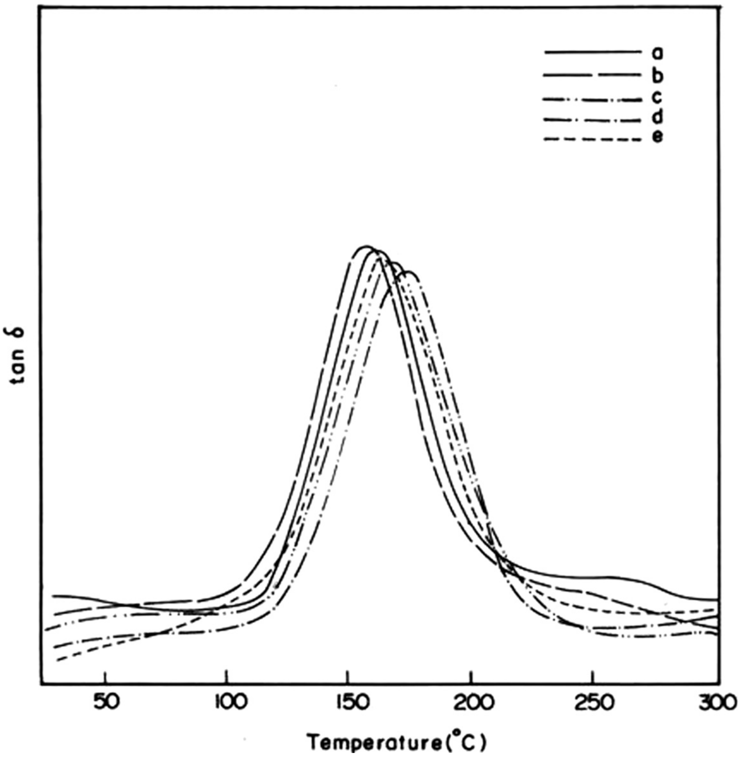


FIGURE 12 Variation of tan δ as a function of temperature (a) unmodified epoxy, (b) polysulfone (8%) modified epoxy and, (c) BMI-1 (8%) modified polysulfone (8%) epoxy system, (d) BMI-2 (8%) modified polysulfone (8%) epoxy system, (e) BMI-3 (8%) modified polysulfone (8%) epoxy system.

Bismaleimide-Modified Polysulfone–Epoxy Systems

The introduction of combination of both polysulfone (8%) and varying concentrations of bismaleimides into epoxy resin alters the values of tensile strength according to their percentage concentration (Figure 4). The values of tensile strength of bismaleimide modified polysulfone–epoxy systems increased with increasing percentage concentration of bismaleimides due to the formation of crosslinked network between the bismaleimides and epoxy systems. The improvement in tensile properties is maximum in the case of BMI-2-modified polysulfone–epoxy systems, when compared to those of BMI-1- and BMI-3-modified epoxy systems (Table 1). The values of tensile modulus obtained for bismaleimides modified polysulfone epoxy systems are also presented in

Table 1 and they exhibit a similar trend, as observed in the case of tensile strength (Figure 5).

Flexural Properties

Polysulfone-Modified Epoxy Systems

The values of flexural strength and flexural modulus of unmodified epoxy, polysulfone modified epoxy are presented in Table 1. The introduction of 4, 8, and 12% polysulfone (by wt%) into epoxy resin enhances the values of flexural strength to only an insignificant extent (Figure 6). The enhancement in the values of flexural strength is explained due to the resilient behavior imparted by molecular structure of polysulfone. Further, the formation of network structure between epoxy and polysulfone also may influence the flexural behavior.

Bismaleimides-Modified Polysulfone–Epoxy Systems

The values of flexural strength of bismaleimides modified polysulfone (8%)–epoxy systems are presented in Table 1. An improvement in the values of flexural strength is observed, when bismaleimides (BMI-1, BMI-2, and BMI-3) are incorporated into polysulfone–epoxy systems (Figure 7). The enhancement in the value of flexural strength is influenced by the homopolymerization of bismaleimides and the formation of crosslinked network structure between the epoxy and bismaleimides. Among the bismaleimides modified epoxy systems, BMI-2-modified epoxy systems exhibit higher values of flexural strength than those of BMI-1- and BMI-3-modified epoxy systems. For example, the flexural strength of 8% BMI-2-modified polysulfone (8%)–epoxy system increased by 12.4% when compared with polysulfone (8%)-modified epoxy system, whereas in 8% BMI-1- and BMI-3-modified polysulfone (8%)–epoxy systems, the flexural strength increased by 11.3% and 6.9%, respectively. The values of flexural modulus of modified epoxy systems follow a similar trend (Table 1 and Figures 8 and 9).

Izod Impact Strength

Polysulfone-Modified Epoxy Systems

Polysulfone incorporation into epoxy resin improves the values of impact strength relative to that of unmodified epoxy system (Figure 10). The incorporation of 4, 8, and 12% polysulfone into epoxy system enhances the values of impact strength by 24.1, 35.2, and 48.6%, respectively (Table 1). The improvement is explained due to the chain lengthening by the introduction of polysulfone and formation of flexible network structure within the system. Further, the presence of ether linkage in the polysulfone skeleton is also responsible for the enhancement in the values of impact strength due to its easy molecular rotation.

Bismaleimides-Modified Polysulfone–Epoxy Systems

The values of impact strength of bismaleimides modified polysulfone–epoxy systems are lowered when bismaleimides are incorporated (24). The decrease in the impact strength is based on the nature and concentration of bismaleimides. The restricted chain mobility due to the formation of network structure is the cause for lowering the impact strength. Among the bismaleimides-incorporated epoxy systems, the maximum reduction in the value of impact strength is observed for 12% BMI-2-modified epoxy system when compared with the same percentage modification using other bismaleimides (BMI 1 and BMI 3) due to the attainment of higher crosslink density by BMI-2-modified epoxy systems (Figure 11).

Hardness

The hardness of unmodified epoxy system is increased with increasing percentage incorporation of polysulfone. A hardness value of unmodified epoxy system is 84, whereas the values of polysulfone modified epoxy systems are increased to between 85 and 89 for increasing concentration of polysulfone up to 12% by wt. The incorporation of polysulfone into epoxy system improved the values of hardness according to the percentage concentration due to the formation of network structure between polysulfone and epoxy system. A further improvement was found in the values of hardness to between 87 and 91 upon increasing the concentration of bismaleimides (upto 12%) in the epoxy and polysulfone-modified epoxy systems.

Dynamic Mechanical Analysis

The dynamic mechanical analysis spectra of unmodified epoxy and epoxy modified with polysulfone and bismaleimides are presented in Figure 12. A single tan δ peak was observed for polysulfone modified epoxy systems. All the modified epoxy systems show similar dynamic mechanical behavior (tan δ). However, the polysulfone modified epoxy systems exhibit higher values of tan δ than that of unmodified epoxy, due to lower crosslink density. The incorporation of bismaleimides into polysulfone modified epoxy systems decreased the value of tan δ due to increased crosslink density. Among the bismaleimides modified polysulfone epoxy systems, BMI-2-modified system exhibits lower value of tan δ ascertaining the formation of higher crosslink density than other modified epoxy systems (Figure 12).

CONCLUSIONS

Data from mechanical studies indicate that the incorporation of polysulfone into epoxy resin improved tensile strength, flexural strength, and impact strength. Similarly, the incorporation of bismaleimides also increased the stress and strain properties with a marginal reduction in impact strength. Dynamic mechanical analysis was used to ascertain the formation of network structure between epoxy and polymeric modifiers (polysulfone and BMIs). Mechanical studies suggest that these bismaleimides-modified polysulfone–epoxy hybrid systems can be used in the form of coatings, adhesives, and matrices for the fabrication of advanced composites for different industrial and engineering applications with better performance and longevity than unmodified epoxy systems.

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