JOURNAL OF Applied Polymer SCIENCE

Determining the Mechanical Properties of Epoxy Resin (DGEBA) Composites by Ultrasonic Velocity Measurement

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ABSTRACT: In this study, the composites of diglycidyl ether of bisphenol A (DGEBA) epoxy resin that have been formed by mixing epoxy resin with allyl glycidyl ether (AGE) and 2,3-epoxypropyl methacrylate [glycidyl methacrylate (GMA)] were prepared in weight % ratios of 90 : 10, 80 : 20, and 70 : 30. A computer controlled analyzer with 35 MHz and a digital oscilloscope with 60 MHz were used for measuring the velocities of ultrasonic wave. The measurement of ultrasonic velocity carried out by pulse echo method at frequencies of 2.25 and 3.5 MHz at room temperature. The values of acoustic impedance (Z), Poisson ratio (μ), and coefficients of elasticity (L, G, K, E) of composites were calculated by values of densities and velocities that obtained. Thus, the effect of modificating epoxy resin (DGEBA) by AGE and GMA on mechanical properties of DGEBA was investigated using the ultrasonic method. Atomic force microscopy has been used for determining the microstructure of composites. By the results obtained from the investigation, it have been established that the longitudinal and shear ultrasonic wave velocities, and the values of all the elasticity constants of DGEBA were increased by modification with AGE and GMA. Also the most suitable combination ratio for the compound of DGEBA : AGE and DGEBA : GMA has been found as 80 : 20. © 2012 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 000: 000–000, 2012

KEYWORDS: epoxy resin; composite; velocity measurement; ultrasound; constants of elasticity

Received 15 July 2011; accepted 21 February 2012; published online 00 Month 2012
DOI: 10.1002/app.37534

INTRODUCTION

During the last few years, the industrial interest has been oriented toward the development of new materials to achieve high strength performance with low weight.[1] At the same time, there has been an increasing demand for quality caused by an increasing demand for safety, especially in the aerospace, aircraft, and automotive industry.[2]

Epoxy resin [diglycidyl ether of bisphenol A (DGEBA)] was widely used as the adhesives, coatings, and encapsulated materials, due to its good mechanical properties and attractive chemical and electronic properties.[3,4] As such, epoxy resins find a wide range of applications in products like paints, surface coatings, adhesives, and electrical accessories. However, epoxies seem to suffer from major draw backs in terms of poor resistance to crack initiation and growth[5] and low impact strength. The great majority of the studies[6–9] involve the chemical modification of epoxy resin with reactive liquid rubber. Mezzenga et al. modified DGEBA with an epoxy terminated hyperbranched polyester.[10] To improve of some properties, a second component such as polyurethane, silicone, and some other thermoplastics are added as modifiers to the epoxy resins.[11–13] Allyl glycidyl ether (AGE) and glycidyl methacrylate (GMA) were used by us for the first time as modifiers in DGEBA. Both modifiers have epoxide group in their structure and may be bound chemically to DGEBA. Therefore, there has been great interest in the chemical modification of polymers with the aim of enhancing their chemical properties and making them useful for special applications.[14–20]

The modulus of elasticity is one of the important parameter that indicates the quality of the materials. Modulus of elasticity determines stiffness—resistance of a body to elastic deformation caused by an applied force. The elasticity modulus of material can be measured using destructive methods like tensile and compressive tests and nondestructive approaches like ultrasonic methods. Ultrasonic methods are among the most common nondestructive techniques (NDTs) used in material science and industry.[21,22] Ultrasonic methods have advantages over destructive methods. Ultrasonic measurements can be made on actual components without destroying the samples. In addition, ultrasonic measurements can be performed for

Table I. Typical Properties of DER 321 Epoxy Resin Were Given by Dow Chemical Company

Property	Value
Epoxide equivalent weight (g/eq)	180–188
Epoxide percentage %	22.9–23.9
Epoxide group content (mmol/kg)	5320–5560
Color (Gardner)	3 Max.
Viscosity @ 25°C (mPa s)	500–700
Density @ 25°C (g/mL)	1.14
Flash point (°C)	121
Shelf life (months)	24

different orientations; this means that the number of elastic modulus measured for a single plane can be more than the number of elastic modulus measured using destructive techniques. Contrary to destructive methods, NDTs give information about material properties without deteriorating material microstructure and serviceability. The main advantages of NDT methods are: the possibility of on-site evaluation, repeatability at the same place during structural service, and quick test results.

Ultrasonic techniques are a versatile tool for investigating the changes in microstructure, deformation process, and mechanical properties.[23] The various parameters upon which the elastic modulus of polymers depend can be studied by measuring the ultrasonic wave velocities. When propagated in polymeric materials, acoustic waves are influenced by the polymer's structure and by molecular relaxation processes. It is possible to estimate the viscoelastic properties of polymeric materials from the velocity and attenuation of longitudinal or shear waves.[24] Ultrasound is finding an increasing number of applications in the modern world. Included amongst these are medical imaging, dentistry, particle sizing, food processing, welding, waste water treatment, and surgical processes.[25] Furthermore, ultrasonic methods have been successfully used to monitor polymer processing,[26] chemical reactions,[27,28] film formation from aqueous polymer dispersions,[29] glue processes, crystallization in polymers,[30,31] and also characterization of polymers.[32] Thus, recently, a lot of attempts have been made to study the sound velocity and attenuation of polymers.[33–42] The ultrasonic velocity method is a nondestructive, economical, and very precise method.[43,44] Moreover, the sound velocity through solids mainly depends on the intermolecular and intermolecular interaction potential.

Between the monomers of the linear polymers, which create the chains, there are very strong covalent bonds, and these covalent bonds provide the mechanical resistance. There are weak electrostatic gravitational forces (e.g., van der Waals forces, hydrogen bonds) between the molecular chains. These van der Waals forces and hydrogen bonds keep the chains of molecules close to each other.[32,45] The longitudinal modulus is related with bonds which creates the polymer chains between monomers. It defines the resistance to elongation. The shear modulus is related with bonds between molecular chains. It defines the resistance to transverse contraction.

AGE is a bifunctional molecule with a terminal epoxy and a terminal allyl groups. AGE is an interesting monomer because it forms unstable and energetically rich macroradicals. AGE has been largely used to modify cellulose[46–52] by the reaction of AGE epoxy group with the cellulose hydroxyl groups. 2,3-Epoxypropyl methacrylate (GMA), containing copolymers are of interest for their epoxy group[53] that can be used to crosslink domains, for example, in ordered block copolymers for nanoporous membranes.[54–56] GMA has also been used for homogeneous and heterogeneous polymer networks[57] and for coatings, matrix resins, and adhesives.[58–61] As it is seen, the related literature does not include any studies regarding DGEBA:AGE and DGEBA : GMA composites were made by ultrasonic methods. Therefore, in this study, it was aimed to determine the effect of AGE and GMA on mechanical properties of DGEBA and to investigate DGEBA composites' elastic properties by ultrasound velocity measurements.

EXPERIMENTAL

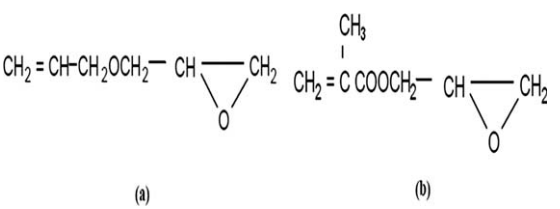
Materials

Materials used in the preparation of samples were procured from the following suppliers. Epoxy resin (DGEBA) type DER 321 was supplied from Dow Chemical Company, Midland, MI, USA in which epoxy equivalent weight 180–188 g/eq was used as matrix system. The more detailed properties of DER321 was given below in Table I. Cycloaliphatic polyamine (Polypox H 043) hardener from the same supplier were used. AGE (molecular weight = 114.15 g/mol, density 0.97 g/cm^3 (20°C), boiling point = 154°C [1013 hPa]), and 2,3-epoxy propyl methacrylate (GMA, molecular weight = 142.15 g/mol, density 1.08 g/cm^3 (20°C), Boiling point = 189°C [1013 hPa]) as modificators were supplied by Merck, Germany (Scheme 1).

Preparation of DGEBA Composites

As seen from Scheme 1, AGE and 2,3-epoxy propyl methacrylate (GMA) have epoxide group in their structure and they can bind to the DGEBA chemically by hardening process. We expect that both physically and chemically modification of DGEBA can increase the DGEBAs properties.

To obtain the composites of epoxy resin (DGEBA), AGE and 2,3-epoxy propyl methacrylate (GMA) were used as modificators of epoxy resin. An amount of AGE and GMA as 10%, 20%, and 30% of DGEBA used for each composite samples. The modificators (AGE and GMA) and DGEBA were mixed with continuous stirring till a homogenous creamy liquid was obtained. Then, an amount of 25% of mixtures was poured into all composite samples' mixtures. The modificators, DGEBA,



Scheme 1. Chemical structure of modificators used in this study: (a) allyl glycidyl ether (AGE) and (b) 2,3-epoxy propyl methacrylate (glycidyl methacrylate, GMA).

Table II. The Composition Rates and Amounts Were Used for DGEBA Composites

Components	Composition rate	DGEBA (g)	Modificator (g)	Hardener (g)
DGEBA : AGE (or GMA)	100 : 0	20.0	-	5.0
DGEBA : AGE (or GMA)	90 : 10	20.0	2.0	5.5
DGEBA : AGE (or GMA)	80 : 20	20.0	4.0	6.0
DGEBA : AGE (or GMA)	70 : 30	20.0	6.0	6.5

and hardener were mixed with continuous stirring till a homogenous creamy liquid was obtained again. The detailed amount of chemicals were used for obtaining composites of DGEBA were given below in Table II.

All the composite mixtures were poured into Teflon moulds (diameter: 28 mm, deepness: 5 mm). Primarily all mixtures were waited for 2 h at room temperature and then dried under vacuum at 60, 70, 80, 100, 120, and 130°C during 1 h for each temperature, respectively. To prevent the formation of gas bubbles in mixtures, this drying process was done. Thus, all the composite samples were obtained by this process.

Measurements

Density Measurements. The density of the composites was measured using Archimedes' principle with double distilled water as the liquid medium[62] by an analytical balance (Radwag AS220/C/2, capacity 220 g, readability 0.1 mg, Poland) and a kit of density (Radwag 220, Poland). Primarily, the temperature of the room inserted into the balance; then, the mass of the samples were measured in air and in water, and finally, the densities of the samples were measured by the balance automatically. All the densities were measured at same temperature as 25°C. The percentage error was 0.001% for the density measurements of the samples.

Ultrasonic Velocity Measurements. The ultrasonic wave velocities measurements were done by pulse echo method at room temperature. The ultrasonic pulses are provided by a 5800PR (35 MHz Panametrics Olympus, Waltham, MA, USA) generator. An electrical impulse with high amplitude and short duration excites the piezoelectrical transducer vibrating on the fundamental mode through the sample, and after reflections on the opposite face returns to the transducer. After propagation in the material, the output signal is displayed on the screen of a numerical oscilloscope (60 MHz GW Instek GDS—2062, Taiwan). 3.5 MHz (Diameter: 10 mm, V183-Panametrics Olympus, USA) longitudinal and 2.25 MHz shear (Diameter: 13 mm, V154-Panametrics Olympus, USA) contact transducers were used. As the coupling medium, glycerin (BQ—Panametrics Olympus, USA) was used for the longitudinal wave measurements, and shear wave couplant (SWC) (SWC-Panametrics Olympus, USA) for the shear wave measurements. Because that shear waves do not propagate in liquids, it is necessary to use a very viscous couplant as SWC when making measurements with these waves. The thicknesses of the specimens were measured 10 times using an analog micrometer and the accuracy of thickness measurement was found ±0.002 mm. The knowledge of the transit time through the thickness of the sample allows the determination of the wave velocities by eq. (1).

$$V = \frac{2d}{t} \tag{1}$$

where V, d, and t are the velocity of sound, the thickness of the sample, and the time-of-flight between subsequent back-wall signals on the oscilloscope, respectively. The measurements were repeated 10 times to check the reproducibility of the data. The accuracy of velocity measurements is about 0.04%.

Calculation of Elastic Constants. The elastic properties of composites were calculated according to the following equations which are valid for isotropic materials.[63–65]

$$L = \rho \times V_L^2 \tag{2}$$

$$G = \rho \times V_S^2 \tag{3}$$

$$K = L - \frac{4}{3}G \tag{4}$$

$$E = 2G\ (1 + \mu) \tag{5}$$

$$\mu = \frac{L - 2G}{2(L - G)} \tag{6}$$

$$Z = \rho \times V_L \tag{7}$$

Table III. Variation of Density (ρ), Longitudinal Wave Velocity (V_L), and Transversal Wave Velocity (V_S) of Composites of the DGEBA/AGE and DGEBA/GMA Samples for Different Compositions (90 : 10, 80 : 20, 70 : 30)

Composites and composition ratios		ρ (g/cm^3)	V_L (m/s)	V_S (m/s)
DGEBA : AGE	100 : 0	1.1640 ± 0.001	2160 ± 0.04	1048 ± 0.04
	90 : 10	1.1450 ± 0.002	2448 ± 0.03	1133 ± 0.04
	80 : 20	1.1290 ± 0.001	2861 ± 0.03	1210 ± 0.03
	70 : 30	1.1120 ± 0.002	2552 ± 0.04	1184 ± 0.05
DGEBA : GMA	100 : 0	1.1640 ± 0.001	2160 ± 0.04	1048 ± 0.04
	90 : 10	1.1390 ± 0.001	2523 ± 0.04	1224 ± 0.04
	80 : 20	1.2110 ± 0.001	2853 ± 0.03	1390 ± 0.04
	70 : 30	1.2100 ± 0.002	2801 ± 0.04	1365 ± 0.05

Table IV. Variation of the Elastic Parameters (L, G, K, E, μ, Z) of Composites of the DGEBA/AGE and DGBA/GMA Samples for Different Compositions (90 : 10, 80 : 20, 70 : 30)

Composites and composition ratios		μ	L (GPa)	G (GPa)	K (GPa)	E (GPa)	Z (10^6 kg/m^2 s)
DGEBA : AGE	100 : 0	0.3461 ± 0.01	5.430 ± 0.03	1.278 ± 0.02	3.726 ± 0.04	3.441 ± 0.05	2.514 ± 0.03
	90 : 10	0.3635 ± 0.02	6.867 ± 0.04	1.472 ± 0.03	4.904 ± 0.05	4.015 ± 0.04	2.804 ± 0.04
	80 : 20	0.3911 ± 0.02	9.249 ± 0.03	1.654 ± 0.04	7.043 ± 0.04	4.601 ± 0.06	3.232 ± 0.04
	70 : 30	0.3628 ± 0.01	7.250 ± 0.03	1.561 ± 0.03	5.168 ± 0.05	4.254 ± 0.05	2.840 ± 0.03
DGEBA : GMA	100 : 0	0.3461 ± 0.01	5.430 ± 0.03	1.278 ± 0.02	3.726 ± 0.04	3.441 ± 0.05	2.514 ± 0.03
	90 : 10	0.3460 ± 0.02	7.255 ± 0.04	1.708 ± 0.03	4.977 ± 0.05	4.598 ± 0.05	2.875 ± 0.04
	80 : 20	0.3445 ± 0.02	9.865 ± 0.04	2.340 ± 0.03	6.744 ± 0.05	6.293 ± 0.06	3.457 ± 0.03
	70 : 30	0.3441 ± 0.01	9.495 ± 0.03	2.257 ± 0.04	6.486 ± 0.04	6.068 ± 0.05	3.389 ± 0.04

where V_L, V_S, L, G, K, μ, E, Z, and ρ are longitudinal ultrasonic wave velocity, shear ultrasonic wave velocity, longitudinal modulus, shear modulus, bulk modulus, Poisson's ratio, Young's modulus of elasticity, acoustic impedance, and density of the samples, respectively. The estimated accuracy of elastic constants and acoustic impedance is about 0.04% and for Poisson's ratio measurements is about 0.02%, respectively.

Morphological Measurements. Atomic force microscopy (AFM) is a versatile technique, which can be used for the characterization of the polymer films, polymer–filler interactions, etc. AFM is one of the most important microscopic techniques used for the surface analysis of polymers on a nanometer scale. The added advantage of using AFM is that it can give distinguished surface topography and surface heterogeneity. The morphology of the neat DGEBA and DGEBA composites were examined with a Solver P47H atomic force microscope (NT-MTD) (Moscow, Russia) operating in tapping mode in air at room temperature. Diamond-like carbon (DLC) coated NSG01 DLC silicon cantilevers (from NT-MTD) with a 2-nm tip apex curvature were used at the resonance frequency of 150 kHz. The Nova 914 software package was used to control the SPM system and for the analysis of the AFM images.

RESULTS AND DISCUSSION

The experimental data of the ultrasonic velocity of longitudinal wave (V_L) and that of shear wave (V_S), density (ρ), and the elastic parameters (L, G, K, E, μ, Z) of composites of the DGEBA/AGE and DGEBA/GMA samples for different compositions are tabulated in Tables III and IV and Figures 1–9. Also some figures of AFM that show the microstructure of composites are given in Figure 10.

Density and Sound Velocity

As seen from Table III and Figure 1, the densities ranged between 1.1120 and 1.1450 g/cm^3 for composites of DGEBA/AGE and ranged 1.1390–1.2110 g/cm^3 for the composites of DGEBA/GMA, respectively. The highest density value was found at weight ratio of 80 : 20 for composites of DGEBA/GMA. It can be seen that the density of hardened DGEBA decreased with increase in weight percent of AGE from 10 to 30 (Table III). However, first, the density of hardened DGEBA decreased at

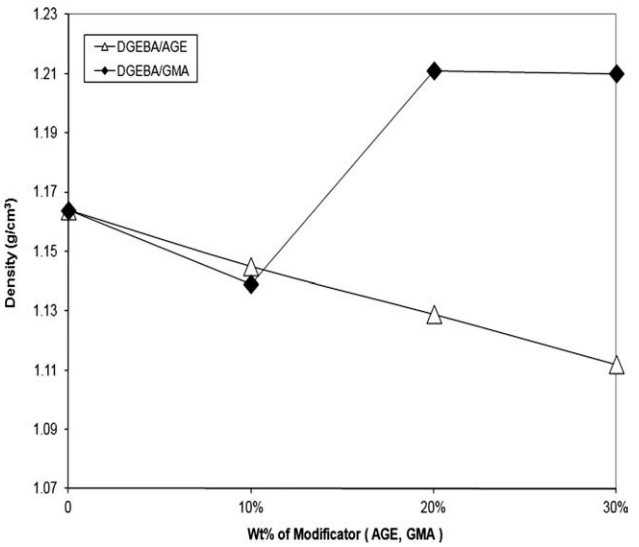


Figure 1. Variation of density with weight percent of modificators (AGE and GMA) in composites.

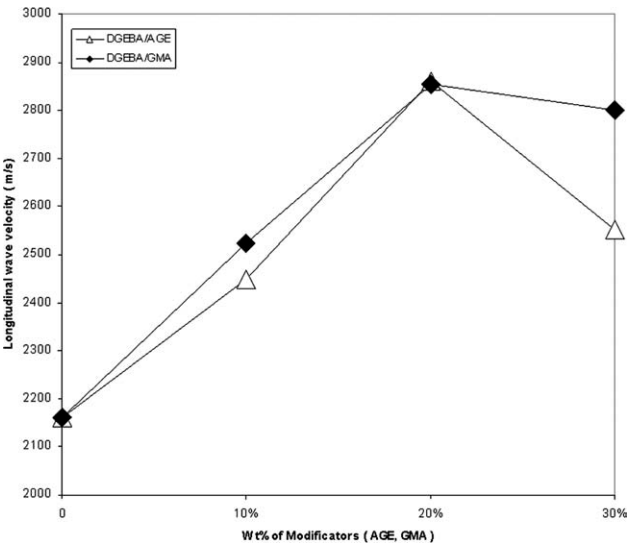


Figure 2. Variation of longitudinal wave velocity with weight percent of AGE and GMA in composites.

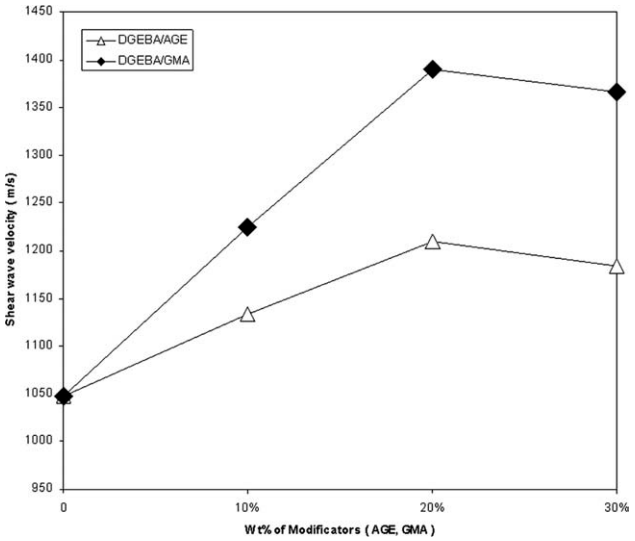


Figure 3. Variation of shear wave velocity with weight percent of AGE and GMA in composites.

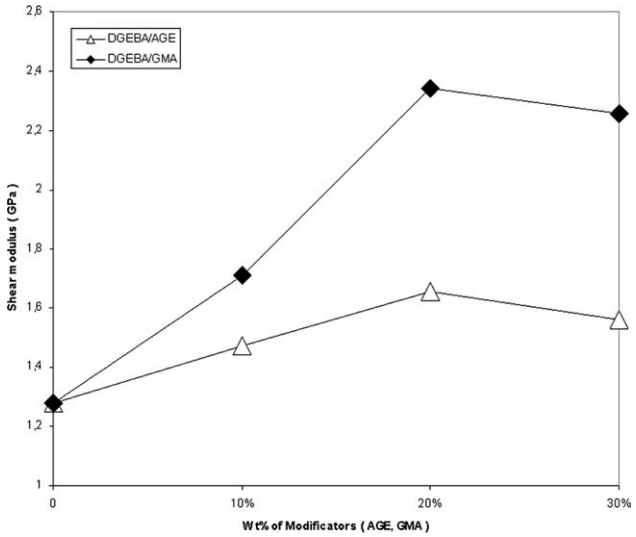


Figure 5. Variation of shear modulus with weight percent of AGE and GMA in composites.

10% addition of GMA. It was increased with increase in weight percent of GMA from 10 to 20.

The variation of velocity as a function of composite composition is presented in Table III and Figures 2 and 3. The variations of V_L and V_S with modificators (AGE and GMA) addition are illustrated in Table III and Figures 2 and 3 for DGEBA composites. Moreover, both longitudinal and shear ultrasonic velocities of DGEBA/AGE and DGEBA/GMA composites, as shown in Table III and Figures 2 and 3, are higher than hardened DGEBA. The V_L and V_S data for hardened DGEBA were obtained as 2160 m/s and 1048 m/s, respectively. The longitudinal ultrasonic velocities of DGEBA/AGE with the increase in weight percent of AGE from 10 to 30 are ranged from 2448 m/s to 2861 m/s and the shear ultrasonic velocities are ranged from 1133 m/s to 1210 m/s.

The longitudinal ultrasonic velocities of DGEBA/GMA with the increase in weight percent of GMA from 10 to 30 are ranged from 2523 m/s to 2853 m/s and the shear ultrasonic velocities are ranged from 1224 m/s to 1390 m/s.

As seen in Figures 2 and 3, although it was found a linearly proportional relationship between ultrasonic velocities and composition of AGE and GMA in the range 0–20 wt % of DGEBA/AGE and DGEBA/GMA composite system. Both longitudinal and shear ultrasonic wave velocity values decreased with increasing the amount of AGE and GMA from 20 to 30 wt % in DGEBA composite.

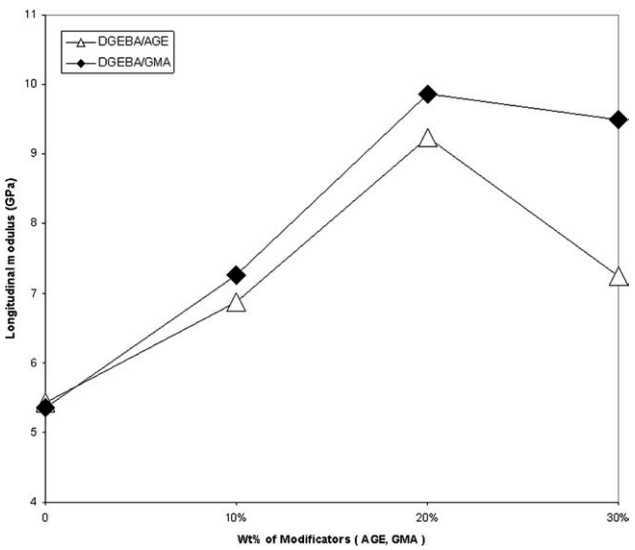


Figure 4. Variation of longitudinal modulus with weight percent of AGE and GMA in composites.

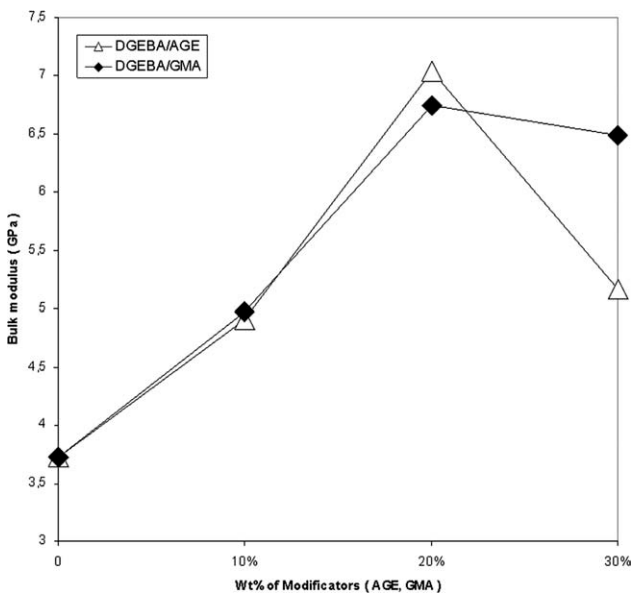


Figure 6. Variation of bulk modulus with weight percent of AGE and GMA in composites.

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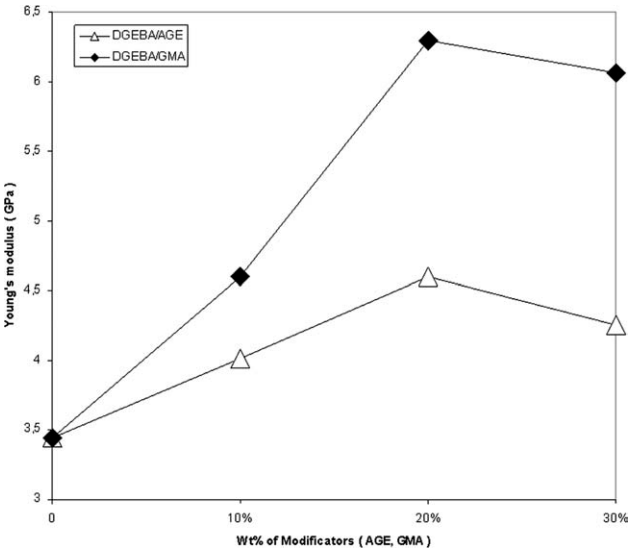

Figure 7. Variation of Young's modulus with weight percent of AGE and GMA in composites.

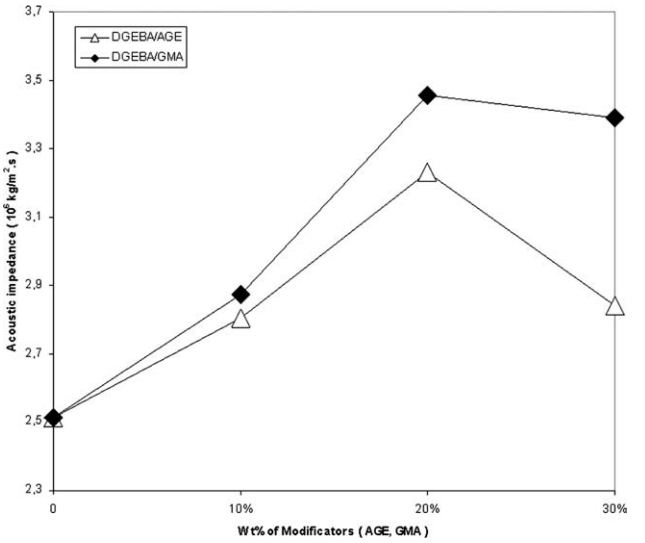

Figure 9. Variation of acoustic impedance with weight percent of AGE and GMA in composites.

Elastic Constants

The elastic constants (*L*, *G*, *K*, *E*) of hardened DGEBA resin and DGEBA composites were calculated using eqs. (2)–(5). The calculated values of elastic constants of hardened DGEBA resin and DGEBA composites as a function of weight percent of AGE and GMA are presented in Table IV and Figures 4–7.

As seen from Table IV, the longitudinal, shear, bulk, and Young's modulus increase with increasing in weight percent of AGE and GMA addition from 0 to 20, but value of all the elastic constants decreased by increasing the amount of AGE and GMA in matrix system from 20 to 30 wt %.

As seen from Table IV and Figures 4–7, the values of elastic constants of DGEBA composites are higher then hardened DGEBA resin. For example, longitudinal modulus of hardened DGEBA was measured 5.430 GPa. When AGE added into hardened DGEBA at 10 wt %, it was increased from 5.430 to 6.867 GPa. For samples of DGEBA/AGE composites, longitudinal modulus ranged from 5.430 to 9.249 GPa, shear modulus ranged from 1.472 to 1.654 GPa, bulk modulus from 4.904 to 7.043 GPa, and Young's modulus from 4.015 to 4.601 GPa, respectively. For samples of DGEBA/GMA composites, longitudinal modulus ranged from 7.255 to 9.865 GPa, shear modulus ranged from 1.708 to 2.340 GPa, bulk modulus from 4.977 to 6.744 GPa, and Young's modulus from 4.598 to 6.293 GPa, respectively.

From Table IV and Figures 4–7, it is seen that the samples of DGEBA/GMA composites' longitudinal modulus ranged from 7.255 to 9.865 GPa, shear modulus ranged from 1.708 to 2.340 GPa, bulk modulus from 4.977 to 6.744 GPa, and Young's modulus from 4.598 to 6.293 GPa, respectively.

When a comparison is made between the elastic properties of the DGEBA/AGE composites and its corresponding DGEBA/GMA composites, in general, DGEBA/GMA composites had higher longitudinal, shear, bulk, and Young's modulus than the DGEBA/AGE composites.

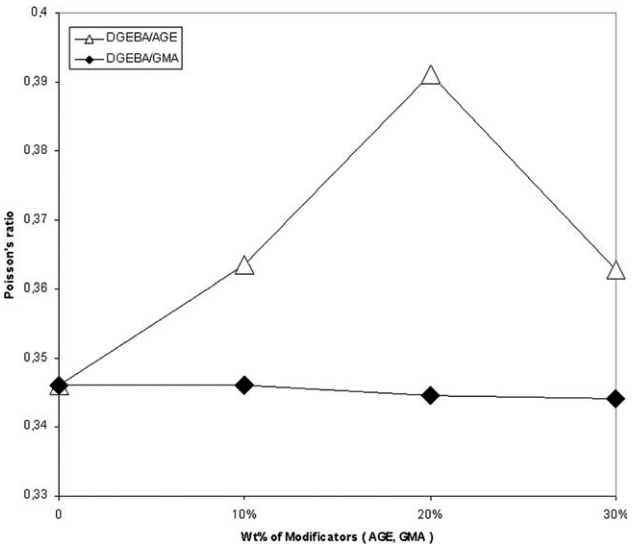

Figure 8. Variation of Poisson's ratio with weight percent of AGE and GMA in composites.

Poisson's Ratio

Poisson's ratio of hardened DGEBA resin and DGEBA composites were calculated using Eq. (6). Figure 8 illustrate the Poisson's ratio values as a function of the weight percent of AGE and GMA. It was found a linearly proportional relationship between addition of AGE in the range 0–20 wt % of hardened DGEBA. When AGE was added at 30 wt %, Poisson's ratio has decreased from 0.3911 to 0.3628.

As it is seen from Table IV and Figure 8, it can be seen that Poisson's ratio increase at 10 wt % of GMA addition. After this amount, it decrease inversely proportional in the range 10–30 wt % of GMA adition. From the results of Table IV and Figure 8, however, Poisson's ratio of hardened DGEBA has increased by addition AGE from 10 to 20 wt %. It is interesting to see

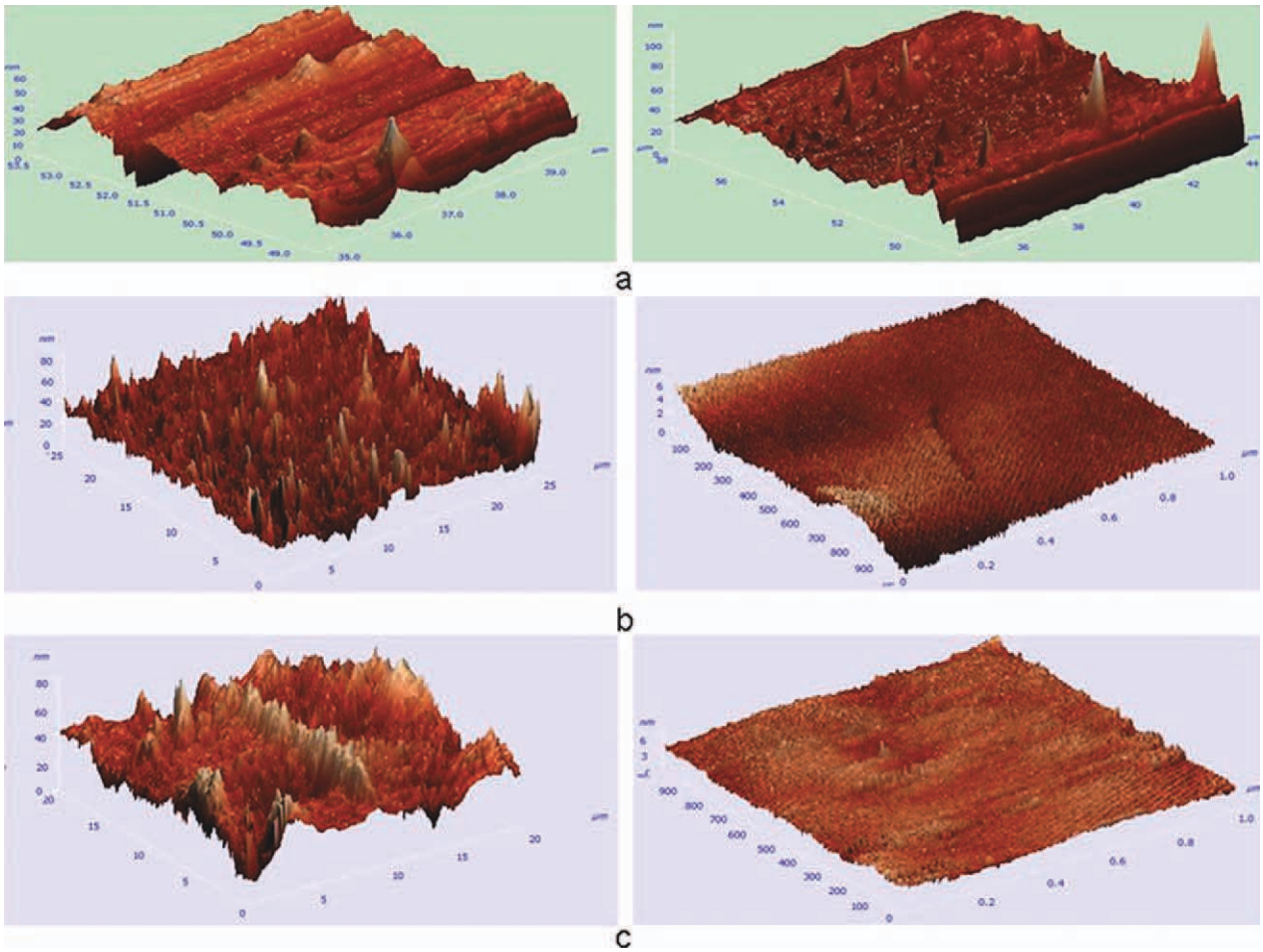

Figure 10. Three-dimensional AFM image of epoxy composite samples with: (a) hardened DGEBA resin, (b) DGEBA/AGE, and (c) DGEBA/GMA. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

that all the elastic constants of DGEBA/AGE possess plane parallelism to Poisson's ratio. Because usually there are an inversely relationship between Poisson's ratio and elastic constants. This is a very important result because that it shows that the assumption of inversely relationship between elastic constants and Poisson's ratio is not true every time. Therefore, the reason of this condition can be a new research subject for scientists.

In the other hand, we can obviously notice that the Poisson's ratio decreases as the percentage of GMA content in DGEBA/GMA increased from 10 to 30 wt %. The decrease of Poisson's ratio usually shows better quality and durability to impacts for most of materials. In accordance with the Poisson's ratio of AGE-modified composites are higher than GMA-modified epoxy composites. The Poisson's ratio of a stable, isotropic, linear elastic material cannot be less than −1.0 nor greater than 0.5 due to the requirement that Young's modulus, the shear modulus, and bulk modulus have positive values.[66] Most materials have Poisson's ratio values ranging between 0.0 and 0.5. Rubber has a Poisson ratio of nearly 0.5.

The highest Poisson's ratio value obtained for the DGEBA/AGE composites at 80 : 20 ratio was 0.3911 and the lowest Poisson's ratio value obtained for the DGEBA/GMA composites at 80 : 20 ratio was 0.3441 at 70.30 ratio.

Acoustic Impedance

The acoustic impedance of hardened DGEBA and DGEBA composites were calculated using Eq. (7). The acoustic impedance of hardened DGEBA was calculated as $2.514.10^6$ kg/m^2 s.

The acoustic impedance ranged from 2.804 to $3.232.10^6$ kg/m^2 s for DGEBA/AGE composites and ranged from 2.875 to $3.457.10^6$ kg/m^2 s for DGEBA/GMA composites (see Table IV and Figure 9), respectively. The acoustic impedance of a material is the opposition to displacement of its particles by sound. So, acoustic impedance indicates the resistance that materials shows when sound waves move in them. According to this defining, if materials have a big impedance they are more durable against to impacts.

Morphological Results

The AFM images (Figure 10) showed the representative 3D surface morphologies of the composites in various dimensions. As expected, the surface morphologies were quite different because of different chemical structure of modifiers.

As seen from AFM images, the heights on the surface of neat epoxy [Figure 10(a)] are not intense in comparison with the composite with AGE which has a relatively bumpy surface

[Figure 10(b)], the composite surface with GMA exhibited increased roughness [Figure 10(c)]. In our opinion, this is due to the carbonyl and methyl groups of GMA unlike AGE. As it is known, the ability of additives to migrate to the surface is defined by several factors such as size, mobility, end-group functionalities, relative composition, and molecular architecture. Generally, existence of modifiers with various chemical structures made the composite surface rougher. It can be clearly seen that AGE and GMA are well distributed throughout the hardened epoxy. In Figure 10(c), for composite containing GMA more rougher surface is observed; thus, a greater energy is needed to break this sample.

The sound velocities and elastic modulus are very sensitive to intermolecular interaction.[67] Therefore, it can be stated that all the elastic parameters are related with internal forces between atoms and molecules of materials. In conclusion, if we summarize the results of the study, it can be stated that DGEBA composites which were made by the DGEBA/GMA combination at ratio of 80 : 20 wt % have best mechanical properties. Consequently by comparison, the DGEBA/GMA composites exhibited greater enhancement in mechanical properties as compared with DGEBA/AGE composites. The degree of crosslinking and bigger molecular weight of GMA than AGE can be shown as the main reason of this condition. Because that molecular weight of GMA is bigger than AGE, the amount of crosslinks between DGEBA and GMA become bigger than amount of crosslinks between DGEBA and AGE. Therefore, the internal forces between atoms and molecules of DGEBA/GMA composites become bigger than DGEBA/AGE.

CONCLUSIONS

This study focused on the effects of chemical treatment of DGEBA resin using AGE and 2,3-epoxy propyl methacrylate (GMA) on the properties of DGEBA composites. The ultrasonic wave velocity as a function of AGE and GMA content was determined from prepared specimens. The results obtained by pulse-echo technique show differences of wave velocity for the specimens with different AGE and GMA content. The study has also assessed the ability of pulse-echo technique to carry out such a testing. The described method can be applied to the postproduction quality control of a finished composite product too. Also it was stated that the measurement of Young's modulus by ultrasonic methods is cheaper and easier than destructive methods.[67] Therefore, measurement of mechanical properties of materials by ultrasonic methods can be recommended to all researchers. Based on the above discussion, ultrasonic wave velocities, elastic constants, and AFM images, the following conclusions can be drawn:

1. According to the values of wave velocities and elastic constants, the most appropriate wt % ratios for DGEBA : AGE and DGEBA : GMA composites were determined as 80 : 20 ratio.
2. The best values of velocities, elastic constants, Poisson ratio, and acoustic impedance have seen at the composite of DGEBA/GMA have 80 : 20 ratio.
3. The pulse echo method has the ability to evaluate the mechanical properties of polymer composites.

Therefore, measurement of mechanical parameters of DGEBA/GMA composites for different ratios by ultrasonic methods can be recommended to all researchers.

ACKNOWLEDGMENTS

This study was supported by the Scientific Research Projects (BAP) Coordinating Office of Selcuk University by the project number of 08101027. The authors are grateful for the support that was provided by Selcuk University, Turkey.

REFERENCES

1. Dobrzanski, L. A. Engineering Materials and Material Design. Principles of Materials Science and Physical Metallurgy; WNT: Warsaw, **2006** (in Polish).
2. Wróbel, G.; Pawlak, S. *J. Achiev. Mater. Manuf. Eng.* **2007,** *22*, 51.
3. Zhengfang, W.; Weiqu, L.; Chaohui, H.; Songqi, M. *J. Appl. Polym. Sci.* **2011,** *121*, 2213.
4. Shell Development Company. EPON Resin Structural Reference Handbook; Shell Development Company: Houston, Texas, **1983.**
5. Pearson, R. A.; Yee, A. F. *J. Mater. Sci.* **1989,** *24*, 2571.
6. Shukla, S. K.; Srivastava, D. *J. Appl. Polym. Sci.* **2006,** *100*, 1802.
7. Kalfoglou, N. K.; Williams, H. L. *J. Appl. Polym. Sci.* **1973,** *17*, 1377.
8. Sanjana, Z. N.; Kupchella, L. *Polym. Eng. Sci.* **1985,** *25*, 1148.
9. Kunz, S. C.; Beaumont, P. W. R. *J. Mater. Sci.* **1991,** *26*, 3828.
10. Mezzenga, R.; Plummer, C. J. G.; Boogh, L.; Manson, J. A. E. *Polymer* **2001,** *42*, 305.
11. Sung, P. H.; Lin, C. Y. *Eur. Polym. J.* **1997,** *33*, 903.
12. Deligoz, H.; Yalcinyuva, T.; Ozgumus, S. *Eur. Polym. J.* **2005,** *41*, 771.
13. Lauter, U.; Kantor, S. W.; Schmidt-Rohr, K.; Mac Knight, W. J. *Macromolecules* **1999,** *32*, 3426.
14. Biswas, M.; Chatterjee, S. *Eur. Polym. J.* **1983,** *19*, 317.
15. Mirzaoglu, R.; Kurbanli, R.; Ersoz, M. In Handbook of Engineering Polymeric Materials; Nicholas, P., Ed.; Marcel Dekker: New York, **1997.**
16. Kurbanli, R.; Mirzaoglu, R.; Akovali, G.; Rzayev, Z.; Karatas, I.; Okudan, A. *J. Appl. Polym. Sci.* **1996,** *59*, 235.
17. Kurbanli, R.; Mirzaoglu, R.; Kurbanov, S.; Karatas, I.; Ozcan, E.; Okudan, A.; Guler, E. *J. Adhes. Sci. Technol.* **1997,** *11*, 105.
18. Kurbanli, R.; Okudan, A.; Mirzaoglu, R.; Kurbanov, S.; Karatas, I.; Ersoz, M.; Ozcan, E.; Ahmedova, G.; Pamuk, V. *J. Adhes. Sci. Technol.* **1998,** *12*, 947.
19. Ahmetli, G.; Yazicigil, Z.; Kocak, A.; Kurbanli, R. *J. Appl. Polym. Sci.* **2005,** *96*, 253.
20. Cıcala, G.; Recca, G. *J. Appl. Polym. Sci.* **2010,** *115*, 1395.

21. Brunarski, L.; Runkiewicz, L. Principles and Application Examples of Non-Destructive Method Testing of Concrete Structures; Institute of Building Technics: Warsaw, Procc., IBT, **1983.**
22. Carino, N. J. Nondestructive Test Methods, Concrete Construction Engineering Handbook; Nawy, E. G., Ed.; CRC Press: Boca Raton, FL, **1999.**
23. Rajendran, V.; Palanivelu, N.; Chaudhuri, B. K.; Goswami, K. *J. Non-Cryst. Solids* **2003,** *320*, 195.
24. Ferry, J. D. Viscoelastic Properties of Polymers; Wiley: New York, **1961.**
25. Mason, T. J. Chemistry with Ultrasound; Elsevier: New York, **1990.**
26. Gendron, R.; Dumoulin, M.; Piche, L. *Polym. Mater. Sci. Eng.* **1995,** *72*, 23.
27. Alig, I.; Lellinger, D.; Johari, G. *J. Polym. Sci. Part B: Polym. Phys.* **1992,** *30*, 791.
28. Parthun, M.; Johari, G. *J. Chem. Phys.* **1995,** *102*, 6301.
29. Alig, I.; Tadjbakhsch, S.; Zosel, A. *J. Polym. Sci. Part B: Polym. Phys.* **1998,** *36*, 1703.
30. Alig, I.; Tadjbakhsch, S. *J. Polym. Sci. Part B: Polym. Phys.* **1998,** *36*, 2949.
31. Alig, I.; Tadjbakhsch, S.; Floudas, G.; Tsitsilianis, C. *Macromolecules* **1998,** *31*, 6917.
32. Oral, I.; Guzel, H.; Ahmetli, G.; Gur, C. H. *J. Appl. Polym. Sci.* **2011,** *121*, 3425.
33. Marsh, G. *Reinf. Plast.* **2002,** 22.
34. Nesvijski, E. G. *Compos. Struct.* **2000,** *48*, 151.
35. Vaccaro, C.; Akers, D. Damage Assessment in a SMC Composite by Means of Ultrasonic Techniques, Review of Progress in Quantitative Non-destructive Evaluation; Plenum Press: New York, **1996;** Vol. *13.*
36. Potel, C.; Chotard, T.; Belleval, J. F.; Benzeggagh, M. Characterization of composite materials by ultrasonic methods: modelization and application to impact damage. *Compos. B. Eng.* **1998,** *29*, 159.
37. Rojek, M.; Stabik, J.; Wróbel, G. *J. Mater. Process. Technol.* **2005,** 162.
38. Wróbel, G.; Wierzbicki, L. *Mech. Manuf. Eng.* **2005,** 575.
39. Maslov, K.; Kim, Y. R.; Kinra, K. V.; Pagano, J. N. *Compos. Sci. Technol.* **2000,** *60*, 2185.
40. Mouritz, A. P.; Townsend, C.; Shah Khan, M. Z. *Compos. Sci. Technol.* **2000,** *60*, 23.
41. Scarponi, C.; Briotti, G. *Composites* **2000,** *31*, 237.
42. Imielinska, K.; Castaings, M.; Wojtyra, R.; Haras, J.; Le Clezio, E.; Hosten, B. *J. Mater. Process Technol.* **2004,** 157–158, 513.
43. Rajendran, V.; Bera, A. K.; Modak, D. K.; Chaudhuri, B. K. *Acoust. Lett.* **1997,** *20*, 168.
44. Nagarajan, A. *J. Appl. Phys.* **1971,** *42*, 3693.
45. Akkurt, S. Plastic Material Knowledge; Birsen Publishing House: Istanbul, Turkey, **1991**; p 138.
46. Sebe, G.; Brook, M. A. *Wood. Sci. Technol.* **2001,** *35*, 269.
47. Petri, D. F. S.; Choi, S. W.; Beyer, H.; Schimmel, T.; Bruns, M.; Wenz, G. *Polymer* **1999,** *40*, 1593.
48. Timar, M. C.; Pitman, M. C.; Mihai, M. D. *Int. Biodeterior. Biodegrad.* **1999,** *43*, 181.
49. Solpan, D.; Guven, O. *Angew. Makromol. Chem.* **1999,** *269*, 30.
50. Cetin, N. S.; Hill, C. A. S. *J Wood. Chem. Technol.* **1999,** *19*, 247.
51. Burton, S. C.; Harding, D. R. K. *J. Chromatogr. A.* **1997,** *775*, 39.
52. Casarano, R.; Matos, J. R.; Fantini, M. C. A.; Petri, D. F. S. *Polymer* **2005,** *46*, 10, 3289.
53. Chen, Z.; Bao, H.; Liu, J. *J. Polym. Sci. Part A: Polym. Chem.* **2001,** *39*, 3726.
54. Yang, S.; Ryu, I.; Kim, H.; Kim, J.; Jang, S.; Russell, T. P. *Adv. Mater.* **2006,** *18*, 709.
55. Rzayev, J.; Hillmyer, M. A. *Macromolecules* **2005,** *38*, 3.
56. Jackson, E. A.; Hillmyer, M. A. *ACS Nano.* **2010,** *4*, 3548.
57. Canamero, P. F.; de la Fuente, J. L.; Madruga, E. L.; Garcia, M. F. *Macromol. Chem. Phys.* **2004,** *205*, 2221.
58. Godwin, G. G.; Jone Selvamalar, C. S.; Penlidis, A.; Nanjundan, S. *React. Funct. Polym.* **2004,** *59*, 197.
59. Zhang, X.; Tanaka, H. *J. Appl. Polym. Sci.* **2001,** *80*, 334.
60. Verweij, P. D.; Sherrington, D. C. *J. Mater. Chem.* **1991,** *1*, 371.
61. Hild, G.; Lamps, J. P.; Rempp, P. *Polymer* **1993,** *34*, 2875.
62. Santhosh Kumar, S.; Seshu, Bai;, V.Rajasekharan, T. *J. Phys. D. Appl. Phys.* **2008,** *41*, 105403.
63. Perepechko, I. I. Acoustic Methods of Investigating Polymers (English Translation); Mir Publishers: Moscow, **1975.**
64. Gur, C. H. *Mater. Sci. Eng. A.* **2003,** *361*, 29.
65. Higazy, A. A.; Afifi, H.; Khafagy, A. H.; El-Shahawy, M. A.; Mansour, A. M. *Ultrasonics* **2006,** *44*, 1439.
66. Gercek, H. *Int. J. Rock. Mech. Min. Sci.* **2007,** *44*, 1.
67. Oral, I.; Guzel, H.; Ahmetli, G. *Polym. Bull.* **2011,** *67*, 1893.