

Swelling and Static–Dynamic Mechanical Behavior of Mica-Reinforced Linear and Star-Branched Polybutadiene Composites

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Received 2 January 1997; accepted 23 April 1997

ABSTRACT: Physical properties of mica-reinforced linear (LPB) and star-branched polybutadiene (SPB) composites have been studied with special reference to the effect of silane coupling agent, degree of crosslinking, and degree of mica loading, as well as the molecular architecture of polybutadienes. Tensile properties increase and swelling decreases with addition of mica for linear polybutadiene. Star-branched polybutadiene shows a reverse behavior, especially beyond 5% of mica. The improvements in mechanical properties are more pronounced in the case of silane-treated mica compositions of both type of polybutadienes. Dynamic mechanical spectra were obtained for linear and star-branched polybutadienes. Effects of mica loading and silane treatment on dynamic moduli are discussed. Dynamic mechanical moduli (E' , E'') of composites increase with increasing mica content for linear polybutadiene but decrease for star-branched polybutadiene beyond certain mica loadings. Effective damping regions were determined in terms of frequency and temperature. The glass transition temperature (T_g) increased slightly, and the damping peak ($\tan \delta$) broadened due to the rubber–filler interaction, especially after silane treatment for both polymers. © 1997 John Wiley & Sons, Inc. *J Appl Polym Sci* **66**: 1943–1952, 1997

Key words: linear and star-branched polybutadiene; mica reinforcement; swelling behavior; dynamic and static mechanical properties; composite

INTRODUCTION

The reinforcing effect of mineral fillers for polymers has been recognized since 1930s. Addition of filler materials to improve the mechanical, electrical, thermal, optical, and processing properties of polymers while reducing their cost has become a popular field of research.

At the present time, more than hundred differ-

ent types of reinforcing materials, both organic and inorganic, have been reported in the literature. However, among these, only a few have been commercialized and used extensively. Depending on the type of reinforcement provided, fillers are often divided into the following three classes: one-dimensional (fibers, whiskers, etc.); two-dimensional (flakes, platelets, etc.) and three-dimensional (beads, spheres, etc.).¹ When optimum reinforcement is desired, the well-known area-to-volume ratio (A/V) should be maximized. Fibers and platelets are two main classes of reinforcing particles that allow for this maximization. Use

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Journal of Applied Polymer Science, Vol. 66, 1943–1952 (1997)
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of these particles maximizes the particle–matrix interaction through the interface. The success of mineral platelet reinforcement is due to their desirable combination of cost and properties such as (1) price per unit mass, which is typically less than one-fifth of the common plastics; (2) stiffness and strength, both of which are greater than those of plastics; and (3) anisotropic alignment providing reinforcement in all directions, in contrast to uniaxially aligned fibers that enhance properties in one direction only.

Mica refers to a group of minerals whose chemical composition is $KAl_2(OH)_2(AlSi_3O_{10})$, and crystals exhibit a high degree of basal cleavage, which allows them to be split into very thin sheets that are strong, flexible, chemically inert, and transparent.² Mica has been used as a filler for plastics due to its low cost; easy availability; and outstanding electrical, thermal, and chemical resistance.

Much work has been carried out to investigate the mica-reinforced thermoplastics. Various kinds of thermoplastics have been used, and their performances were evaluated.³ The mechanical and flow properties of mica-reinforced polyolefins, such as polypropylene and polyethylene, were investigated by several authors.^{4–6} In order to improve the adhesion between the polymer binder and inorganic substrate, which then causes improvement in the strength of the resulting product, many methods have been proposed, including plasma,^{7–9} titania coupling agent,¹⁰ chlorinated paraffin,¹¹ silane coupling agent treatments,^{12–14} and ultrasonic delamination.¹⁵ In the area of rubber reinforcement with mica, there are some studies conducted on siloxane rubber compositions with good weatherability and quiet operation for wiper blades,¹⁶ good heat insulating properties and compressive strength for inorganic cellular thermal insulator,¹⁷ good solvent resistance and antisoiling property for cooking utensils,¹⁸ and good shock absorbing and vibration damping properties.¹⁹ Most of these are described in patents.

Among other rubbers, polyurethane^{20,21} and polyurethane–epoxy composites²² may be listed among other mica reinforced rubbers, which are especially important for their good damping properties.

Szentivanyi et al.²³ published a study on rubber hoses based on nitril rubber–mica composites for fuel line systems in which swelling, permeability,

and low temperature properties were optimized by selection of the mica filler.

A study on 1,2-polybutadiene and natural rubber composites as odorless and vibration-damping sheets having mica group minerals as fillers has been patented.²⁴

Debnath et al.²⁵ have reported the effect of silane coupling agent on vulcanization conditions, network structure, polymer filler interaction, physical properties, and failure mode of mica-filled styrene butadiene rubber.

This article reports a comparative study on the dynamic and static mechanical properties of linear (LPB) and star–branched polybutadiene (SPB) mica composites. The effect of degree of crosslinking, silane treatment of mica, and the degree of mica loading and molecular architecture of polybutadienes on both the mechanical and swelling behavior of resultant composites are discussed.

EXPERIMENTAL

Materials Used

Two types of polybutadienes manufactured by Monsanto, Italy, are used. Some characteristics of the samples used are given in Table I.

Muscovite mica (40 Å) used for this study was supplied by Sabuncular–Mica Trading Corporation of Turkey, Çine. Dicumyl peroxide (DCP) was obtained in commercial grade. The silane coupling agent, A-174, γ -methacryloxy propyl trimethoxy silane was supplied by Union Carbide (UK). Tetrahydrofuran (THF) was the product of Merck (Germany) and used as received.

Sample Preparation

Application of the coupling agent was done as follows. The silane coupling agent was added to distilled water previously adjusted to PH 5.5 with acetic acid to give a 2% solution. Solid mica was mixed with the coupling agent solution for 2 h and then filtered on a Buchner Filter. The amount of silane (A-174) in the silane-treated mica was determined by carbon hydrogen analysis as 0.4%.

All composites were prepared using silanized and unsilanized mica to give 0, 5, 20, 40 wt % of mica and were crosslinked to two different extents (0.5 and 2.0 phr DCP). The following nomenclature is used. Compositions of linear and star poly-

Table I Characteristics of Linear (LPB) and Star-Branched Polybutadienes (SBP) Used in the Present Study

Sample	MWD ^a	$M_w^a \times 10^5$	η_0^b (MPa)	% Cis 1 : 4 Isomer ^c	% Trans 1 : 4 Isomer ^c	% 1,2-Isomer ^c
LPB	1.81	3.30	0.60	80.80	9.71	9.50
SPB	2.20	5.20	32.81	72.41	8.12	19.50

^a Obtained from gel permeation chromatography (Waters; 25°C).

^b Zero shear viscosity determined from loss modulus (E'') values taken of 25°C and 1 Hz.

^c Determined from FTIR spectra of the samples by comparing 1239, 912, and 724 cm^{-1} peaks corresponding to trans 1 : 4 and 1 : 2, and cis 1 : 4 isomers, respectively (see Haslan and Willis²⁶).

butadienes without mica and having 2.0 phr DCP are indicated as LPB2 and SPB2, respectively. Linear polybutadienes with unsilanized and silanized mica are designated as LUM and LSM, respectively. Two numbers appended to the three letter words denote the amount of mica in percent and the amount of DCP used in phr. LUM 20/2 represents, for example, networks prepared with linear polybutadiene with 20% unsilanized mica and 2.0 phr DCP. The same notation is used for both types of polybutadienes. Mixing was achieved on an open two-roll mill with a nip gap of 0.25 mm. Optimum cure time at 150°C was determined using rheograms taken with Monsanto Rheometer R-100. Vulcanization was done in an electrically heated press at 150°C in 2-mm-thick steel molds. The samples were conditioned for 24 h at 18°C before testing. All properties were measured along the machine direction.

Physical Testing of the Samples

Stress-strain data were determined using a Zwick Universal Testing Machine (model 1446) at room temperature with strips having approximately 6 mm width and 50 cm/min drawing rate.

The swelling ratio q was defined as the ratio of the volume of the swollen to unswollen composite. The sample dimensions of $3.0 \times 1.6 \times 0.2$ cm were weighed and then placed in THF at room temperature, carefully blot-dried with absorbent tissue, and reweighed. The swelling ratio was obtained by means of the following equation²⁷:

$$q = V/V_0 = 1 + (W - W_0)D_0/W_0D_s$$

where V_0 and V are the volumes of unswollen and swollen composite, W_0 and W are the weights of the unswollen and swollen composite, and D_0 and D_s are the densities of the composite and solvent.

The dynamic mechanical thermal analysis (DMTA) of the samples was performed using a Polymer Laboratories dynamic mechanical thermal analyzer. Sample dimensions were ca. $16.0 \times 8.0 \times 2.0$ mm. The temperature and frequency ranges varied between -150 to 200°C and 0.1 to 5 Hz, respectively.

RESULTS AND DISCUSSION

Curing Characteristics of Composites

Rheometric data related with 5% silanized and unsilanized mica-loaded LPB2 and SPB2 are given in Table II.

As can be seen from the table, for LPB2, optimum cure time increases with the addition of unsilanized mica; whereas silane treatment doesn't alter the optimum cure time of the composite having unsilanized mica significantly. In other words, silane treatment increases the rate of vulcanization, as compared with that of the untreated one, which is probably due to the increased unsaturation resulted from methacryl group of the silane coupling agent.

Table II Curing Characteristics of the Mixes at 150°C

Sample	Minimum Torque (dN m)	Maximum Torque (dN m)	Optimum Cure Time (min)
LPB2	50.30	98.40	31.52
LUM 5/2	53.20	139.10	44.44
LSM 5/2	42.80	130.00	43.10
SPB2	13.20	112.30	19.37
SUM 5/2	16.30	146.70	17.07
SSM 5/2	19.70	144.20	17.40

Table III Physical Properties of Linear Polybutadiene Composites

Measurement	LPB2	LUM 5/2	LUM 20/2	LUM 40/2	LSM 5/2	LSM 20/2	LSM 40/2	LUM 20/0.5	LSM 20/0.5
Swelling ratio	3.61	3.51	2.94	2.87	3.33	2.81	2.22	5.13	3.72
Tensile strength (MPa)	0.80	0.90	1.38	1.78	1.15	2.23	3.55	1.72	2.38
Elongation at break (%)	38.49	39.51	42.86	42.14	50.10	44.73	50.76	235.00	288.00
Loss factor ^a ($\tan \delta$) $\times 10^{-2}$	1.625	1.37	1.05	4.15	1.27	1.30	2.48	5.86	5.75
Elastic modulus (MPa)	7.90	8.20	11.40	10.80	10.40	15.50	13.30	2.00	2.64

^a Data taken at 1 Hz and 25°C.

Addition of unsilanized mica to SPB2 decreases the optimum cure time and the maximum torque values, but the use of silanized mica decreases them in a small extent.

Vulcanization rate decreases in the case of both silanized and unsilanized mica in LPB2; but if the matrix is replaced with star type, much more care has to be taken to prevent scorching due to higher vulcanization rate.

All other physical properties are summarized in Tables III and IV.

Swelling of Composites

The swelling behaviors of linear and star-branched polybutadiene composites as a function of mica loading are presented in Figure 1(a,b), respectively.

It is evident from Figure 1(a) that mica causes a decrease in swelling for linear type polybutadiene. For samples with silanized mica, swelling

decreases rapidly up to 20% filler and remains unchanged beyond this value. For SPB2 in Figure 1(b), there is little decrease in swelling at small amounts of filler, followed by a sudden increase. Similar to the behavior observed for the linear polybutadienes, silane treatment decreases the swelling of the rubber, indicating enhanced polymer filler interaction.

As expected, both linear and star-branched polybutadienes having unsilanized mica and low degrees of crosslinking (0.5 phr DCP) (LUM 20/0.5 and SUM 20/0.5) show higher swelling ratios than the same composites with higher degree of crosslinking (2.0 phr DCP) (Table III and IV). Similar behavior was observed by Chen and Ma²¹ in fiber-reinforced blocked polyurethane composites. In addition, here, polybutadiene composites having low degrees of crosslinking also exhibit a decrease in swelling when silanized mica is used as reinforcer. This is a major benefit in terms of practical applications.

Table IV Physical Properties of Star-Branched Polybutadiene Composites

Measurement	SPB2	SUM 5/2	SUM 20/2	SUM 40/2	SSM 5/2	SSM 20/2	SSM 40/2	SUM 20/0.5	SSM 20/0.5
Swelling ratio	1.36	1.32	1.91	1.90	1.34	1.75	1.61	2.54	2.40
Tensile strength (MPa)	10.90	12.25	2.63	2.50	13.00	3.98	4.76	2.01	2.20
Elongation at break (%)	14.11	3.90	19.65	13.50	11.70	20.87	19.46	36.00	27.00
Loss factor ^a ($\tan \delta$) $\times 10^{-2}$	10.20	11.10	2.60	3.73	10.00	4.20	5.30	0.85	0.75
Elastic modulus (MPa)	115.00	278.00	23.60	35.20	158.00	19.20	32.20	15.21	16.44

^a Data taken at 1 Hz and 25°C.

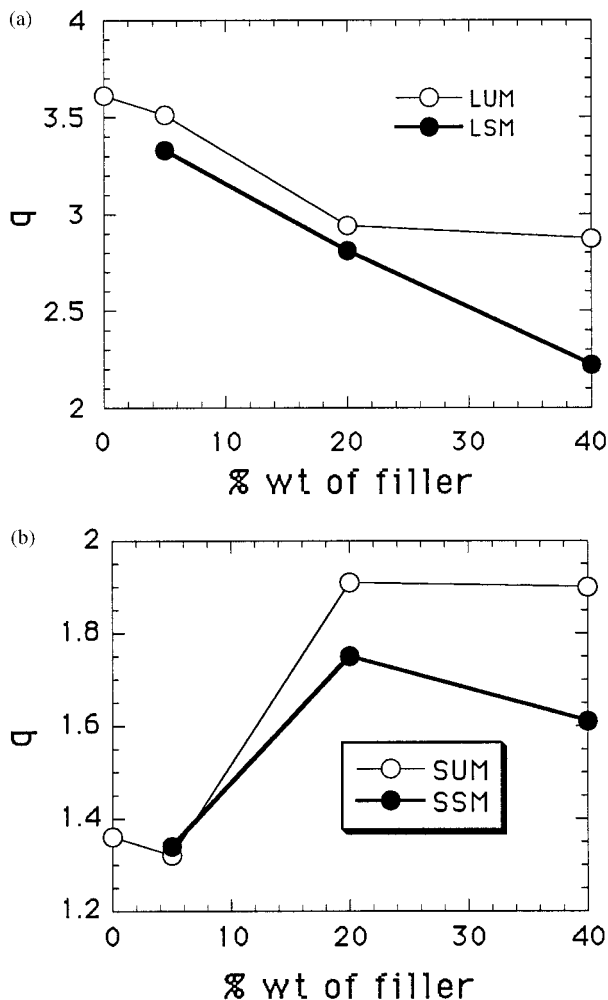


Figure 1 Plots of swelling ratios (q) of (a) linear polybutadiene and (b) star-branched polybutadiene composites having 2.0 phr DCP as a function of unsilanized and silanized mica loading.

Static Mechanical Properties of Composites

Figure 2(a,b) shows the variation of tensile strength with mica loading of the linear and star-branched polybutadiene composites, respectively.

SPB2 shows higher tensile strength and modulus than LPB2 as expected due to the beneficial star-branched structure of the former. It is also seen from Figure 2(a) that mica addition causes an improvement in the tensile strength of LPB, from 0.8 to 1.8 MPa. In the case of composites having 5, 20, and 40% mica, the rise of stress at break indicates both the orientation of mica along the direction of applied force, which is also evident from the higher elongation values as already reported by Malik and Prud'homme,²⁸ and the ad-

sorption of polymer segments at several sites of a filler particle, which effectively introduces giant multifunctional crosslinks into the system and increases the network chain density.²⁹

Effect of bridging via silanization on mica reinforcement is observed by an increase in tensile strength for both types of polybutadienes. It is well known that if there is adhesion between polymer and filler, tensile strength of the composite increases. If there is no or weak adhesion, tensile strength decreases.³⁰

As with swelling data, in Figure 2(b), tensile strength increases initially by addition of 5% mica to SPB2 but then suddenly drops beyond 20% loading. This may be attributed to a possible decrease in the melt viscosity of the system, especially at higher concentrations of mica, or to the higher molecular weight of SPB, as in the case of thermoplastics.³¹ The decrease in melt viscosity

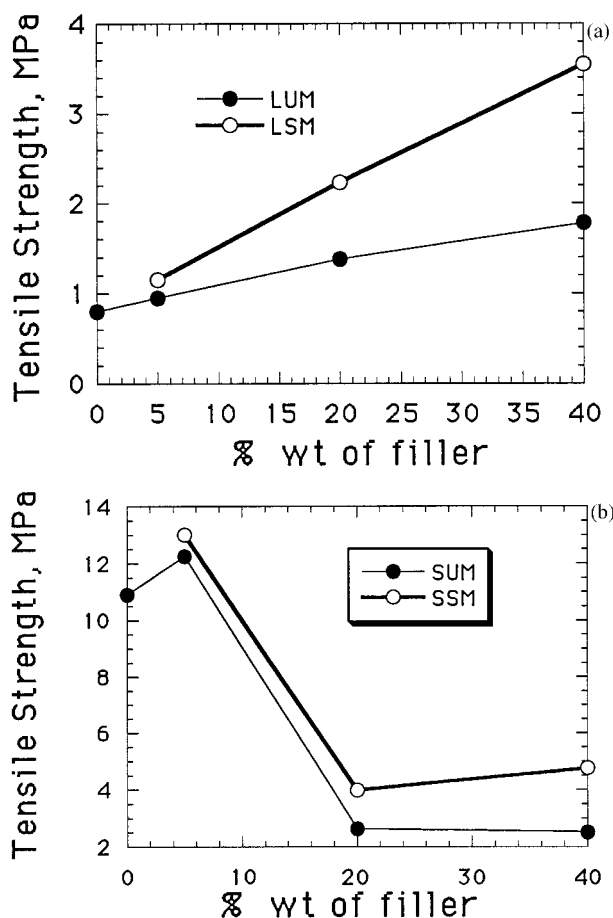


Figure 2 Variation of ultimate tensile strength with concentration of unsilanized and silanized mica for (a) linear polybutadiene and (b) star-branched polybutadiene composites having 2.0 phr DCP.

may result from the poor mixing of mica and high zero shear viscosity (η_0) of SPB2 (see Table I). One can speculate that this effect would yield in a greater reduction in ultimate strength at the composition, which also shows a maximum swelling, which is exactly what is observed.

The experimentally determined elastic moduli of the different linear and star-branched composites are also listed in Tables III and IV. The moduli of the composites increase with increasing amount of mica both for unsilanized and silanized samples. This increase is expected since the flexing of the matrix is prevented by relatively high modulus filler particles.³² Also, interparticle distance between two nearest neighboring particles will decrease with increasing amount of filler, and the number of percolation pathways formed by the shear stress regions will increase, which in turn will increase the elastic modulus as reported by He and Jiang.³³ On the other hand, 5% loading causes a more significant increase (more than twice) in the elastic modulus of SPB composites than that of LPB composites.

Silane treatment enhanced the stiffness of all composites of LPB, whereas the reverse response was found for SPB, except for 5% mica content, paralleling the tensile strength data.

Figure 3(a,b) presents elongation at break values of linear and star-branched polybutadiene composites, respectively.

Addition of mica does not make the linear polybutadiene composite brittle, as is evident from the higher percentage of elongation at break at high mica loading [Fig. 3(a)]. In the same manner, elongation increases with surface treatment of mica, which allows a decrease at surface energy of the filler, reduction in agglomeration, and improvement in flake alignment.²⁵

For SPB given in Figure 3(b), on the other hand, the concentrations at which maximum swelling occurs shows higher elongation for unsilanized samples; whereas silanization resulting in enhancement in tensile strength gives the composite an additional elongation, as in the case of linear polybutadiene.

Dynamic Mechanical Properties of Composites

Dynamic moduli of both linear and star polybutadiene composites are listed in Tables V and VI, respectively.

In Table V, one can observe that both E' (storage modulus) and E'' (loss modulus) increase with

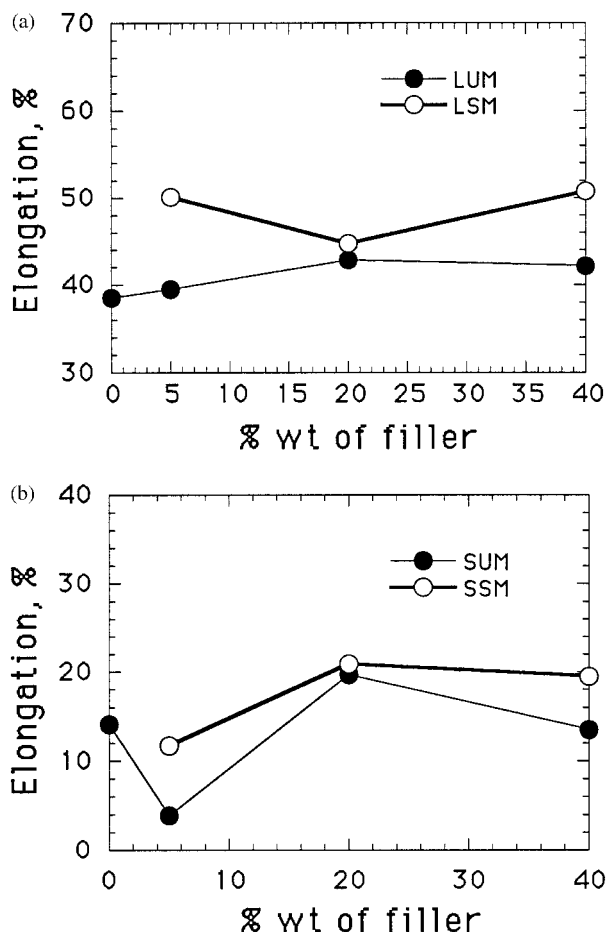


Figure 3 Variation of elongation at break with concentration of unsilanized and silanized mica for (a) linear polybutadiene and (b) star-branched polybutadiene composites having 2.0 phr DCP.

increasing filler content at each selected frequency. Silanization also causes an increase in E' , whereas E'' shows a decrease.

For SPB2 (Table VI), unsilanized mica introduces a decrease in both moduli, E' and E'' , just as in the static modulus and tensile strength. Silanization causes a further decrease.

For dynamic viscoelastic behavior, the characteristic zone of frequency scale can be easily determined. Figure 4(a,b) represents the variation of E' and E'' , respectively, with frequency (ν) for LUM 20/2, LSM 20/2, SUM 20/2, and SSM 20/2 samples.

In the low frequency range of 0.3–5 Hz, E' for both polymers is nearly constant or shows very small increment, whereas the change in E'' is relatively higher. This zone may be characterized as the plateau zone where E' changes little with fre-

Table V Dynamic Moduli of Linear Polybutadiene Composites at Different Frequencies

V (Hz)	E' (MPa)				E'' (MPa)			
	0.3	1.0	3.0	5.0	0.3	1.0	3.0	5.0
LPB2	3.87	3.90	3.91	3.93	0.03	0.06	0.08	0.09
LUM 5/2	4.70	4.72	4.73	4.74	0.03	0.06	0.08	0.10
LUM 20/2	8.93	8.96	8.98	8.99	0.06	0.09	0.12	0.14
LUM 40/2	13.52	13.64	13.76	13.82	0.512	0.54	0.57	0.61
LSM 5/2	4.76	4.78	4.79	4.80	0.03	0.06	0.08	0.09
LSM 20/2	6.39	6.40	6.42	6.43	0.06	0.08	0.11	0.13
LSM 40/2	18.29	18.39	18.50	18.55	0.40	0.44	0.52	0.56
LUM 20/0.5	5.22	5.40	5.58	5.68	0.25	0.32	0.38	0.42
LSM 20/0.5	5.22	5.41	5.59	5.67	0.24	0.31	0.37	0.40

quency and E'' values increase. This behavior is usually interpreted by a somewhat vaguely defined concept, entanglement coupling. The tangled contorted macromolecules form a temporary network in which the molecules act in some respects as though they were coupled at widely separated points; within the periods of oscillation in this frequency range, there is plenty of time for the network strands between coupling points to arrange their configuration and store elastic energy through an entropy change. There is not enough time for the much more complicated molecular rearrangements that would enable widely separated portions of a molecules to change their relative position, as these processes would involve complicated snaking motions through the entanglements.³⁴

The loss factor ($\tan \delta$) is one of the damping parameters of interest since it is a measure of

the ability of the polymer to convert mechanical energy into heat at a temperature or frequency of interest. $\tan \delta$ values measured at 25°C and 1 Hz for the composites are given also in Tables V and VI in relation with crosslinking density. These values increase with the decrease in the crosslinking density. This may be resulted from that; when the material was applied by external force in a less crosslinked state, more and more macromolecules were joined to transform the configuration. Then the internal friction among macromolecule segments increases to convert the mechanical energy into heat. Meanwhile, the interaction between macromolecules and fillers become violent. The increment of the internal friction and interaction increases the mechanical dissipation and enhances the vibration damping. Same observations were reported also by Weibo and Fengchang.²² Since silanization acts as additional crosslink

Table VI Dynamic Moduli of Star-Branched Polybutadiene Composites at Different Frequencies

V (Hz)	E' (MPa)				E'' (MPa)			
	0.3	1.0	3.0	5.0	0.3	1.0	3.0	5.0
SPB2	195.10	208.01	222.02	229.02	18.55	20.56	22.65	23.90
SUM 5/2	226.60	241.41	256.83	264.72	21.57	23.73	25.96	27.29
SUM 20/2	17.44	17.54	17.66	17.73	0.38	0.43	0.50	0.54
SUM 40/2	53.10	53.71	54.42	54.82	1.83	2.00	2.26	2.45
SSM 5/2	175.41	188.62	202.00	209.40	17.46	19.32	21.55	22.89
LSM 20/2	17.72	17.92	18.17	18.31	0.64	0.72	0.83	0.90
SSM 40/2	31.12	31.52	31.98	32.23	1.62	1.69	1.84	1.95
SUM 20/0.5	15.84	15.87	15.90	15.92	0.08	0.12	0.16	0.82
SSM 20/0.5	18.61	18.65	18.67	18.70	0.08	0.13	0.18	0.21

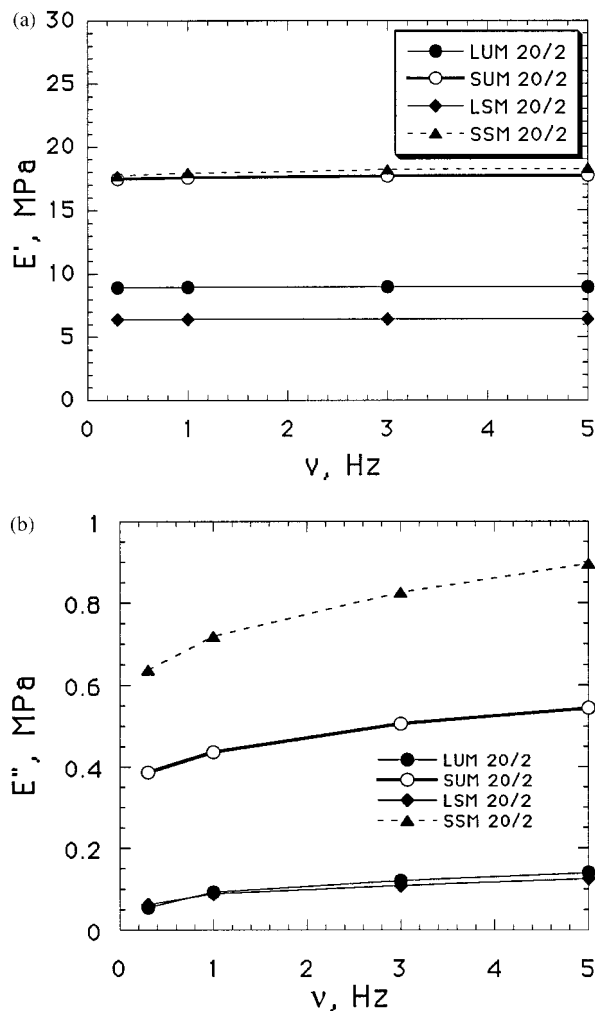


Figure 4 Variation of (a) storage modulus (E') and (b) loss modulus (E'') of unsilanized and silanized mica loaded linear and star-branched polybutadiene composites as a function of frequency.

points, the decrease in loss factor was observed for the both polymers when silanized mica was used (see LUM 20/0.5, LSM 20/0.5, SUM 20/0.5, and SSM 20/0.5 in Tables III and IV).

Figures 5 and 6 show the dynamic mechanical spectra of the linear and star-branched polybutadiene composites, respectively.

Although the interpretation of the rheological properties of linear and star polyisoprenes in terms of their large scale structure relies on the assumption that the local friction coefficient, hence, T_g , is independent of the structure, this is not necessarily true for the stars of very high arm molecular weight and high functionality due to crowding near the branch site. Thus, having two

distinct glass transition values for linear and star-branched polybutadienes used here are in well agreement with the above statement since the star-branched polybutadiene used in this study has a high arm molecular weight.

In Figure 5, one can observe that the glass transition temperature (T_g) of LPB2 is -88.2°C . In the presence of about 40% mica, the T_g of the composite is at the temperature (-86.8°C), and the damping peak becomes broad. Although the mica is inert at this temperature range, the dynamic mechanical properties of composite is different from the unfilled polymer due to the effect of mica–matrix interphase.

In this study, moreover, silane treatment also causes an additional broadness of the $\tan \delta$ peak. We believe that a slight upshift of the glass transition temperature, the broader damping peak, and the increased dynamic modulus are due to the bonding between matrix and mica.

In accordance with previous studies conducted on linear and star-branched polyisoprenes,³⁵ tangent loss peaks for star-branched polybutadiene are significantly broader than for its linear counterpart, which is also evident in Figure 6. The addition of 5% mica results in a shift of T_g of SPB2 from 38.9 to 65.8°C with a second transition. In the same manner, Wang and Xu³⁶ have reported that in rubber–mica composites, there exists an interphase of considerable fraction with a different T_g value from bulk of the matrix. The hindered segmental motion in this interphase appears in the dynamic mechanical measurements with a high and broad $\tan \delta$ and high loss modulus E'' . Here, silanization causes a further shift to 74.8°C , together with the additional broadening.

As illustrated in the figures, by adding the platelet filler and increasing the interaction between filler and polymer, glass transition range can be adjusted so that the composite becomes an effective damper at that temperature or frequency range of interest.

CONCLUSIONS

For LPB, addition of unsilanized mica marginally decreases the swelling of the composites in THF and increases the tensile strength, elongation at break, and modulus values. Silanization of mica surface by using γ -methacryloxy propyl trimethoxy silane, A-174 substantially improves the ultimate tensile strength, indicating enhanced poly-

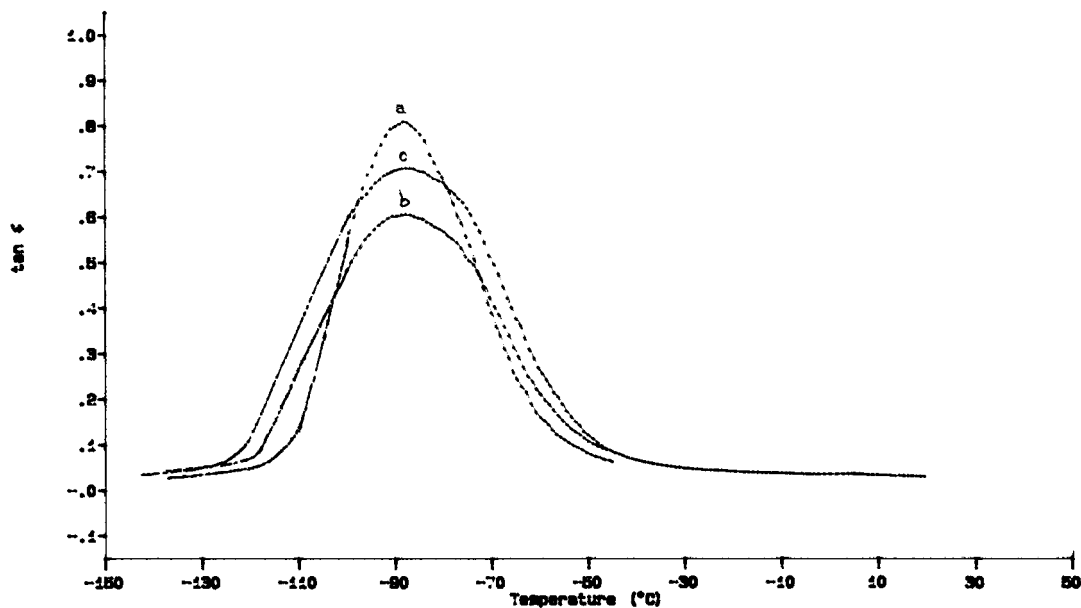


Figure 5 Dynamic $\tan \delta$ (damping) versus temperature plot (a) of LPB2, (b) LUM 40/2, and (c) LSM 40/2 samples.

mer-filler interaction, and increases the elongation to a small extent and causes a decrease in swelling.

In SPB, ultimate tensile strength, elongation, and elastic modulus show a drastic drop; whereas swelling shows an increase as compared to unfilled polymer, especially beyond 5% loading.

However, a similar beneficial effect is observed via silanization by improving tensile and swelling behaviors, as in the case of its linear counterpart. The detailed rheological studies of especially star-branched polybutadiene seems to be necessary.

Dynamic mechanical properties show that the moduli E' and E'' increase with increasing mica

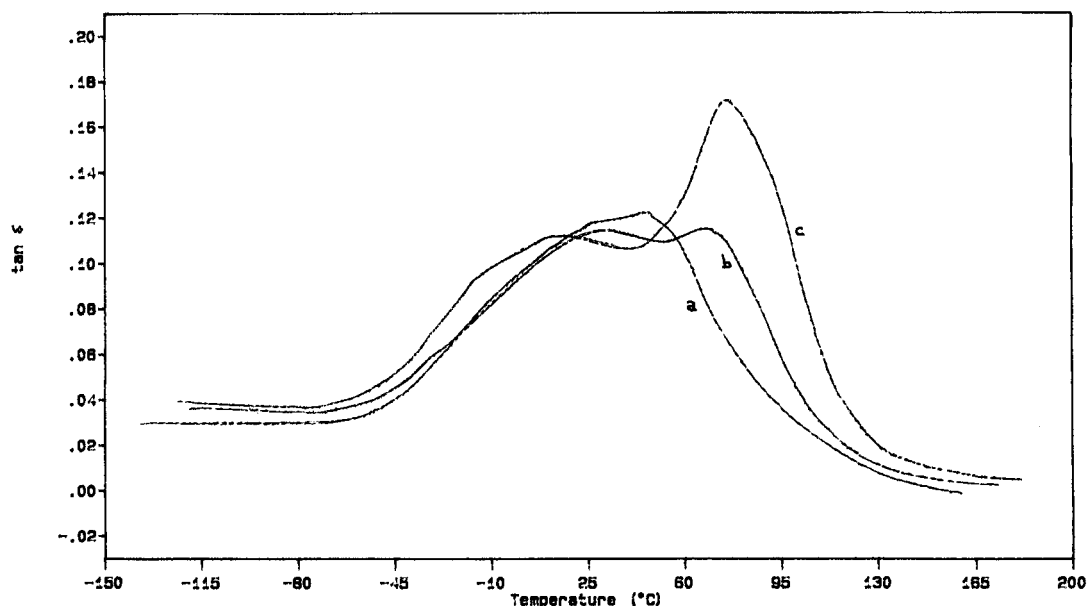


Figure 6 Dynamic $\tan \delta$ (damping) versus temperature plot of (a) SPB2, (b) SUM 40/2, and (c) SSM 40/2 samples.

content, and the damping peak becomes broad when silanized mica is used with LPB. The reverse is observed for SPB, especially beyond 5% loading. Furthermore, a slightly higher glass transition temperature (T_g) and broader damping peak ($\tan \delta$) for both polybutadiene composites are observed upon addition of unsilanized and silanized mica. This indicates a good vibration damping property of the material at wider temperature and frequency ranges.

Partial support given by Bogazici University Research Foundations, Project No. 96P003, is gratefully acknowledged. The authors are also thankful to the companies of Sabuncular and Union Carbide, Turkey, for supplying the materials.

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