Effective Thermal Conductivity of Three-Phase Styrene Butadiene Composites

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ABSTRACT: Transient plane source technique was used for the simultaneous measurement of thermal conductivity and thermal diffusivity of three-phase styrene butadiene rubber composites. Two series of styrene butadiene rubber composites were studied, having natural rubber as a variable filler in both the composites along with 10 phr of silica and clay, respectively. The measurements were done at room temperature and normal pressure. The experimental results show that there is a small variation in the thermal conductivity of both the composites with the filler (NR) fraction. It is interesting to note that the thermal conductivity shows a sharp decrease at 10 phr filler loading and then increases. The comparative study of these composites shows that the conductivity as well as the diffusivity of the silica reinforced composites is larger than that of the clay composites. The thermal conductivity of the filler NR has been evaluated using the Agari model. It has also been found that the composite with 40 phr of NR has the maximum thermal conductivity. © 2000 John Wiley & Sons, Inc. J Appl Polym Sci 76: 1799–1803, 2000

Key words: thermal conductivity; thermal diffusivity; filler fraction; transient plane source (TPS) technique; natural rubber; composites

INTRODUCTION

During the past decade, the study of composites[1–3] and blends[4] has gathered momentum and has become an important area of study. This is because of the wide range of applications of these materials in science and technology. Composites can offer a combination of properties, which are not obtainable by single element. By reinforcing a polymer matrix by a variety of fillers, we are now able to tailor materials according to our requirement.[5] Present study deals with the elastomeric composites, which are of much interest from many points of view. The styrene butadiene rubber (SBR) compounds filled with natural rubber condom waste particles are the result of a successful effort to develop a cost-effective technique to convert waste and used rubber into a processable form.[6,7] The reuse of waste rubber is important not only from the point of view of economy but ecology also, because natural rubber takes decades to decompose. Various types of studies on morphology, mechanical propreties,[8,9] and rheological behavior of the NR latex waste-filled SBR compounds have already been made. However, no serious effort has been made so far to study the thermal properties such as thermal conductivity and thermal diffusivity of these composites.

Heat transfer process through these systems in general can be studied in terms of their charac-
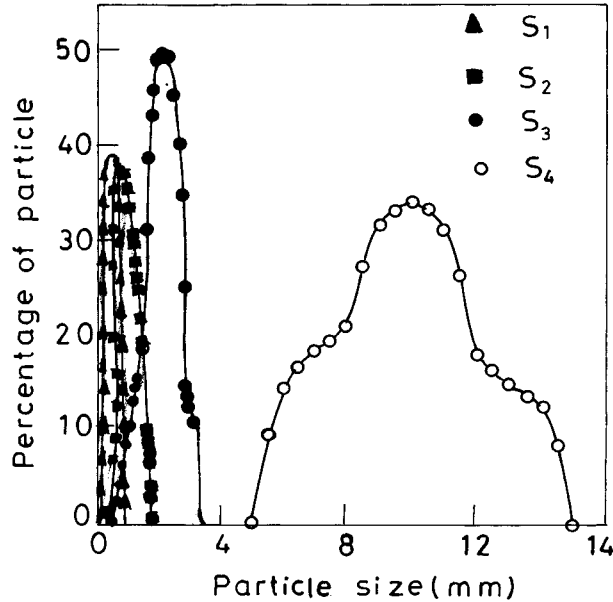
Figure 1 Particle size distribution curve of natural rubber.

Table I Composition of Composites

Material	Loading
SBR	100
Zinc oxide	5
Stearic acid	2
CBS	1
TDQ	1
Sulfur	2.2
Filler	Variable

teristic thermal properties. In this study, we have successfully measured the effective thermal conductivity and thermal diffusivity of SBR composites using the transient plane source (TPS) technique at room temperature and thermal conductivity of the filler NR has been evaluated by a theoretical model.

SAMPLE PREPARATION AND EXPERIMENTAL SETUP

The powdering of lightly crosslinked NR latex waste is done by a mechanical grinding process in a toothed wheel mill to get a poly dispersed powder. The particle size distribution of NR latex waste is shown in Figure 1. A powdered form of precipitated silica and English-Indian clay was used in the preparation of the composites. Compounding of SBR, NR, silica and SBR, NR, clay was done in a two roll mixing mill and curing was done in an electrically heated hydraulic press at 150°C using a conventional vulcanization curing system. A sample of 28-mm diameter and 13-mm thickness was prepared by compression moulding technique. Other details regarding the composites are presented in Table I.

The TPS technique has proved to be a precise and convenient method for measuring the thermal transport properties of electrically insulating materials. The TPS method consists of an electrically conducting pattern (Fig. 2) in the form of a bifilar spiral, which also serves as a sensor of the temperature increase in the sample. In Figure 2, K-4521 is the design number of the sensor and ′K′ stands for kapton. The sensor is sandwiched between the thin insulating layers of kapton. Assuming the conductive pattern to be in the Y-Z plane of a coordinate system, the rise in the temperature at a point Y-Z at time t due to an output power per unit area Q is given by[10,11]:

$$\Delta T(y,z,\tau) = \frac{1}{4\pi^{3/2}a\lambda} \int_o^\tau \frac{d\sigma}{\sigma^2} \int_A dy'dz'$$

$$\times \ Q\left(y'z't - \frac{\sigma^2 a^2}{\kappa}\right) \exp\left[\frac{-(y-y')^2 - (z-z')^2}{4\sigma^2 a^2}\right]$$

$$(1)$$

where $\kappa(t-t') = \sigma^2 a^2$, $\theta = a^2/\kappa$, $\tau = (t/\theta)^{1/2}$, a is the radius of the hot disc which gives a measurement of the overall size of the resistive pattern, and θ is known as the characteristic time. σ is a constant variable and λ is the thermal conductivity in the units of W/mK. The temperature increase $\Delta T(\tau)$ because of flow of current through the sensor gives rise to a change in the electrical resistance $\Delta R(t)$ which is given as[11]:
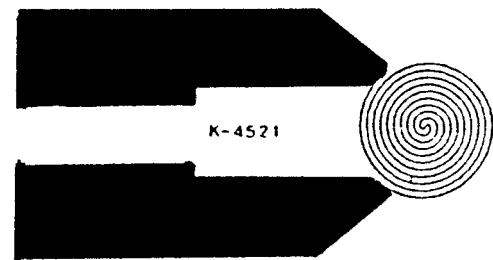

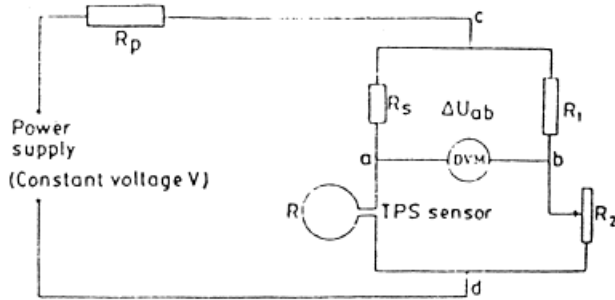**Figure 2** Schematic diagram of the TPS Sensor.

Figure 3   Bridge circuit diagram for TPS technique. R_1 and R_2 are the resistances, R_p is the effective resistance of the wires outside the arms of the bridge, R is the resistance of the TPS element, and DVM is the digital voltameter.

$$\Delta R(t) = \alpha R_0 \overline{\Delta T(\tau)} \qquad (2)$$

where R_0 is resistance of the TPS element before the transient recordings has been initiated, α is the temperature coefficient of resistance (TCR), and $\overline{\Delta T(\tau)}$ is the mean value of the time dependent temperature increase of the TPS element. $\overline{\Delta T(\tau)}$ is calculated by averaging the increase in temperature of the TPS element over the sampling time because the concentric ring sources in the TPS element have different radii and are placed at different temperatures during the transient recording[11]:

$$\overline{\Delta T(\tau)} = \frac{P_o}{\pi^{3/2} a \lambda} D_s(\tau) \qquad (3)$$

where

$$D_s(\tau) = [m(m+1)]^{-2}$$

$$\times \int_0^\tau \frac{d\sigma}{\sigma^2} \left[\sum_{l=1}^m l \left\{ \sum_{k=1}^m k \, \exp \frac{-(l^2+k^2)}{2\sigma^2 m^2} L_o \left(\frac{lk}{2\sigma^2 m^2} \right) \right\} \right]$$

$$(4)$$

P_0 is the total output power, and L_0 is the modified Bessel function. To record the potential difference variations, which normally are of the order of a few millivolts during the transient recording, a simple bridge arrangement as shown in Figure 3 was used. If we assume that the resistance increase will cause a potential difference variation $\Delta U(t)$ measured by the voltmeter in the bridge, the analysis of the bridge indicates that

$$\Delta E(t) = \frac{R_s}{R_s + R_o} I_o \Delta R(t)$$

$$= \frac{R_s}{(R_s + R_o)} \frac{I_o \alpha R_o P_o}{\pi^{3/2} a \lambda} D_s(\tau) \qquad (5)$$

where

$$\Delta E(t) = \Delta U(t)[1 - C.\Delta U(t)]^{-1} \qquad (6)$$

and

$$C = \frac{1}{R_s I_o \left[1 + \dfrac{\gamma R_p}{\gamma(R_s + R_o) + R_p} \right]} \qquad (7)$$

The definition of various resistances is found in Figure 3. R_s is a standard resistance with a current rating that is much higher than I_o, which is the initial heating current through the arm of the bridge containing the TPS element. γ is a constant, which is chosen to be 100 in the present measurement. Calculating $D_s(t)$ using a computer program and recording the change in potential difference $\Delta U(\tau)$, one can determine λ. Thermal conductivity and thermal diffusivity of all the samples were measured using the TPS method, which are reproducible within 2%.
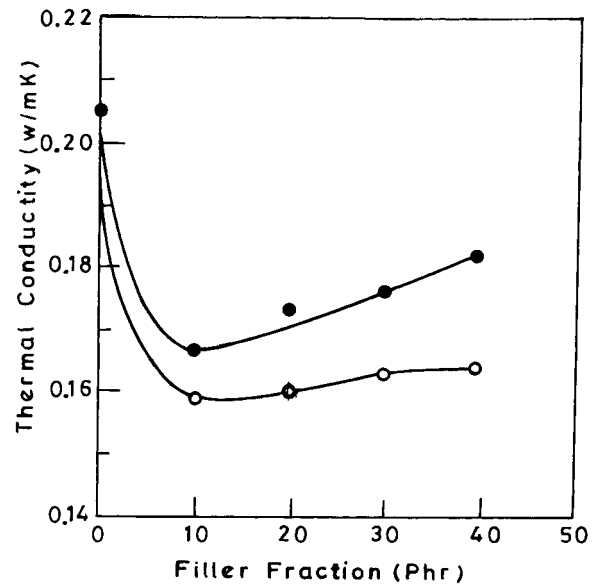

Figure 4   Variation of experimental values of effective thermal conductivity with filler volume percentage: (●) silica; (○) clay.
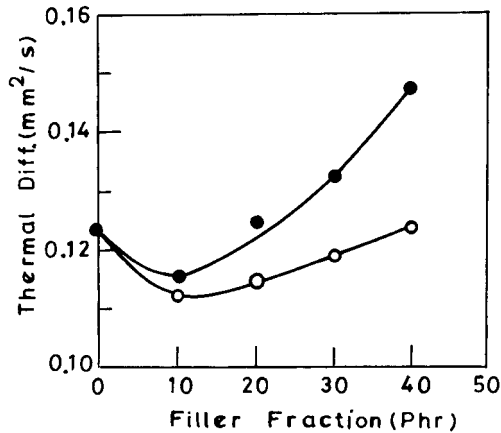
Figure 5 Variation of thermal diffusivity with volume percentage of filler: (●) silica; (○) clay.

RESULTS AND DISCUSSION

The effective thermal conductivity and thermal diffusivity of the SBR composites obtained experimentally are plotted in Figures 4 and 5, respectively as a function of filler fraction and are presented in Table II.

From the plots, it is clear that the thermal conductivity as well as diffusivity of the composites decreases until 10 phr of the filler loading and then starts increasing linearly as the filler fraction increases. To explain this behavior of the composite, thermal conductivity of the filler was evaluated, because the effective thermal conductivity of the composites depend on the thermal conductivity of the constituent elements.[12]

The thermal conductivity of the filler NR has been evaluated using the Agari model[13–15] for the composites containing silica as well as clay. According to the model, the thermal conductivity of the composites is linearly related to the volume percentage of the filler. The straight line fit of the
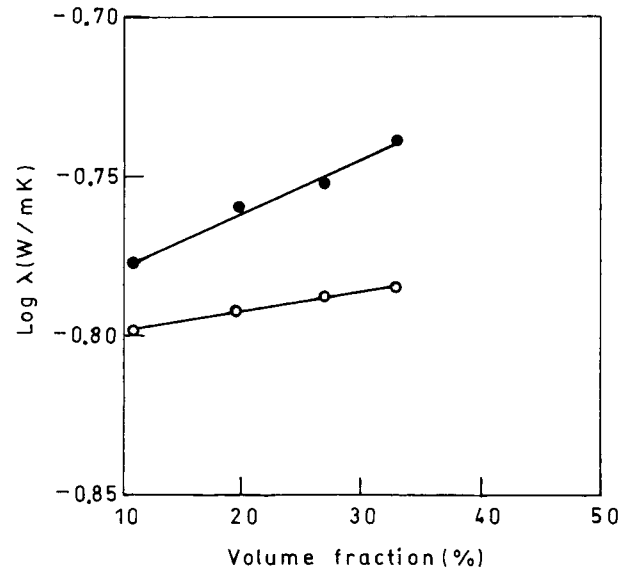

Figure 6 log λ versus volume percentage of NR: (●) silica; (○) clay.

experimental values is shown in Figure 6. Thermal conductivity results of NR obtained by the model are shown in Table III. It is clear from Table III that the thermal conductivity of NR in silica composites is slightly more than the thermal conductivity of NR in clay composites.

It should be noted that in the present article, the SBR/latex waste of the NR system has been studied, not the SBR/NR blend containing raw NR, because the raw NR does not contain certain unreacted accelerated fragments as latex waste contain.

The lowering in the effective thermal conductivity value of the composite could be traced to the blending effect of NR particles in SBR with NR being less thermally conductive, hence thermal conductivity decreases at this particular composition. At higher filler concentrations, the linear

Table II λ and κ Represent the Thermal Conductivity and Thermal Diffusivity of the SBR Composites, Respectively

Filler Fraction	λ^{a} $(W/m - K)$	κ^{a} (mm^2/sec)	λ^{b} $(W/m - K)$	κ^{b} (mm^2/sec)
10	0.167	0.116	0.159	0.113
20	0.174	0.125	0.161	0.115
30	0.177	0.133	0.163	0.119
40	0.182	0.148	0.164	0.124

[a] Represents silica composites.
[b] Represents clay composites.

Table III Thermal Conductivity of NR in Silica and Clay Composites Using the Agari Model

Composites	λ_{NR}	C_p	C_f
SBR + NR + silica	0.160	0.183	4.9
SBR + NR + clay	0.157	0.765	1.0

increase in the thermal conductivity values of the composites has been observed, which may be due to the fact that as the filler fraction increases, more and more particles are available for the conductive chain formation in the SBR matrix and the thermal conductivity of the composite starts increasing. The trend of the thermal diffusivity is the same as that of the thermal conductivity. Our earlier studies[16] also show a similar type of behavior for SBR + NR blends. The only difference is the presence of the third phase, i.e., silica and clay, respectively, in the SBR composites. From Figures 4 and 5 it is obvious that the thermal conductivity and thermal diffusivity values for silica composites are larger than that of the clay composites. This is due to the fact that thermal conductivity of silica (λ_{silica} = 1.2 $W/m - K$) is larger than that of clay[17] (λ_{clay} = 0.9 $W/m - K$). In the literature, it has been found that if a matrix is reinforced with filler of greater thermal conductivity from that of the matrix thermal conductivity, the effective thermal conductivity of the composite increases, and vice versa. It has to be noted that the fraction of NR in both types of composites is the same. So the factor affecting the conductivity of the composites is the third phase, i.e., silica and clay respectively.

Besides, it is also interesting to note that the filler materials are unable to increase the thermal conductivity of the composites above the matrix thermal conductivity (0.205 W/mK) value. This may be due to the porosity factor which results either from the poor wetout of the filler by the rubber phase or by the generation of moisture in the bonding.

CONCLUSIONS

Thermal conductivity of SBR composites was measured using the TPS method at room temper-ature. Thermal conductivity values show a sharp decrease until 10 phr of filler loading and then starts increasing linearly with filler loading.

Thermal conductivity values of silica composites are larger than clay composites; predicting clay is less conductive than silica.

Thermal conductivity of NR determined using the Agari model are in quite good agreement with our earlier results.

It is found that SBR blends with 40 phr of filler loading has the maximum thermal conductivity among the measured values.

The presence of the filler is unable to enhance the thermal conductivity value above the thermal conductivity value of the matrix.

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