Thermal Effects of Heat-Resistant Rubber Blends. I. Joule Heating Effects in Carbon Black-Loaded NR / SBR Blends

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Synopsis

A heat-resistant rubber blend of natural rubber NR and styrene butadiene rubber (SBR-1502) was prepared according to the standard techniques. The rubber blend was loaded with different concentrations of LAMP black and general purpose furnace (GPF) black for obtaining conductive rubber composites. The obtained vulcanizates were subjected to electrical conductivity measurements over a wide range of temperature and electric field. It was found that all samples showed negative values of temperature coefficient of conductivity (TCC). The temperature developed inside the samples by the Joule heating effect was recorded as a function of time (T–t) at different values of the initial applied DC-electric power. The T–t dependence was found to obey an exponentially growing function with a time constant (τ), which was found to be dependent on the concentrations as well as the type of carbon black and the value of the initial applied power. The current–voltage characteristics of the samples showed a negative resistance behavior after a characteristic voltage (V^*). It was noticed that V^* decreases as the carbon black concentration increases.

INTRODUCTION

The electrical conductivity of carbon black-loaded rubber has attracted the attention of some investigators.[1,2] In our previous work[3-10] study of different parameters affecting the electrical conductivity of rubber containing different types of carbon black was carried out. In the present work a trial was made to investigate the electrical properties in accordance with Joule heating effect aiming at obtaining electrical rubber heaters. The conductive rubber samples used as heating elements possess the following advantages: (i) the source of heat is not localized as in the case of wire heaters, and the maximum temperature is therefore less for a given heat output. This may be a big safety factor in certain applications. (ii) The conductive rubber heaters if flexed will not break and give rise to incendiary sparks, as do resistance wires, which have relatively low fatigue resistance and will not stand up well to vibration. (iii) In many cases fabrication of conductive rubber elements is simpler than that of wire elements. In this work rubber blend consisting of natural rubber and styrene–butadiene rubber was selected as such blend is relatively thermally stable. Such blend was mixed with two different types of carbon black

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TABLE I
Composition of the Investigated (NR/SBR) Samples[a]

	ML8	ML9	ML10	MG8	MG9	MG10
NR	50	50	50	50	50	50
SBR-1502	50	50	50	50	50	50
Stearic acid	1.5	1.5	1.5	1.5	1.5	1.5
Clay[b]	70	70	70	70	70	70
Paraffin wax	1.5	1.5	1.5	1.5	1.5	1.5
TMTMS[c]	1	1	1	1	1	1
PBN[d]	1	1	1	1	1	1
Nonox BL[e]	0.5	0.5	0.5	0.5	0.5	0.5
Zinc oxide	15	15	15	15	15	15
LAMP	80	90	100	—	—	—
GPF	—	—	—	80	90	100
Sulfur	1	1	1	1	1	1

[a]The ingredients are arranged in the same way as used during their preparation.
[b]Hydrous aluminum silicate.
[c]Tetra-methyl-thiurammonosulfide.
[d]Phenyl-β-naphthylamine.
[e]Diphenylamine–acetone resin.

possessing different characteristics with respect to their effect as conducting fillers.

EXPERIMENTAL

Rubber blend of natural rubber and styrene butadiene rubber (SBR-1502) was prepared by mastication of each type then mixing for a period of 20 min with other ingredients. The ingredients were added according to their arrangement shown in Table I. The mixing procedure and measuring techniques have been described elsewhere.[3-6] The vulcanization process of the samples was conducted at 416 K under a pressure of about 4×10^6 N/m^2 for 20 min.

All tested samples were thermally aged at 70°C for 50 days before conductivity measurements to get reasonable stability and reproducibility of the measurements.[4] Besides, it is assumed that rubber samples will encounter similar conditions during their use as heating elements.

The temperature inside samples has been recorded by copper–constantan fine thermocouple inserted inside the sample before vulcanization.

RESULTS AND DISCUSSION

Figures 1 and 2 represent the temperature dependence of the electrical conductivity for conductive blends loaded with LAMP and GPF blacks, respectively. These composites showed a negative temperature coefficient of conductivity (TCC) over a wide range of temperature. The maximum values of the TCC are tabulated in Table II. This behavior of the heat-resistant rubber composite permits us to apply higher values of the electric power to increase the temperature of the samples by Joule heating effects. Figure 3 shows the temperature–time dependence (T–t) of LAMP black-loaded (NR/SBR) vulcanizates as a result of Joule heating for an initial power of 10 W. The sample

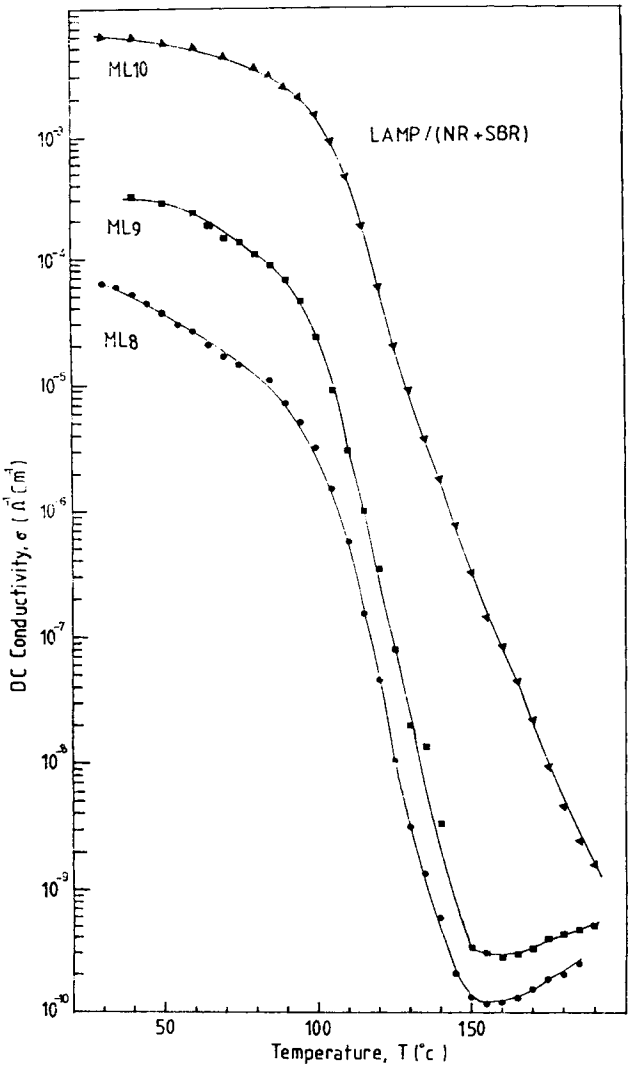

Fig. 1. Temperature dependence of the electrical conductivity of LAMP black-loaded (NR + SBR) vulcanizates.

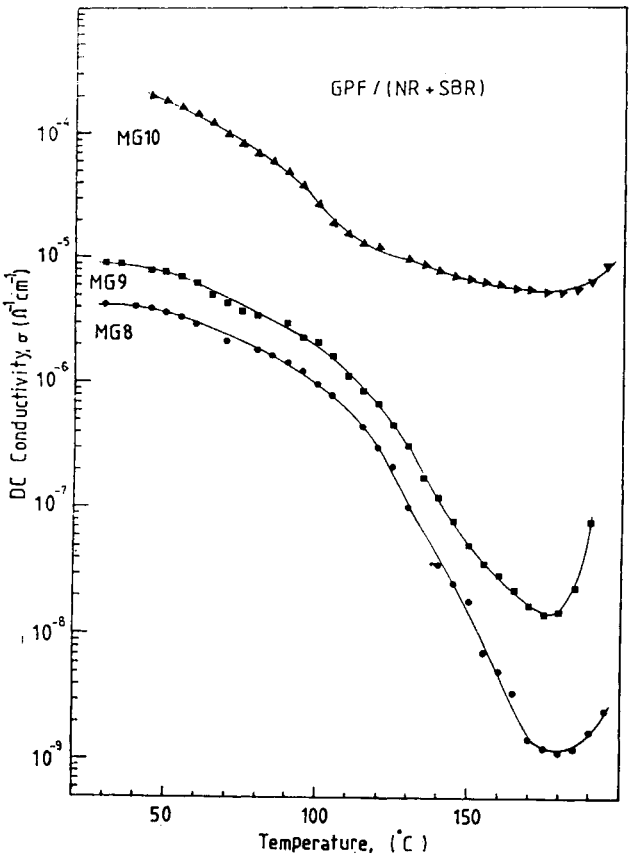

Fig. 2. Temperature dependence of the electrical conductivity of GPF black-loaded (NR + SBR) vulcanizates.

TABLE II
Dependence of TCC on the Black Type and Concentration

Sample	MG8	MG9	MG10	ML8	ML9	ML10
T (°C)	155	135	100	125	130	120
TCC (deg^{-1})	0.27	0.13	0.07	0.42	0.39	0.27

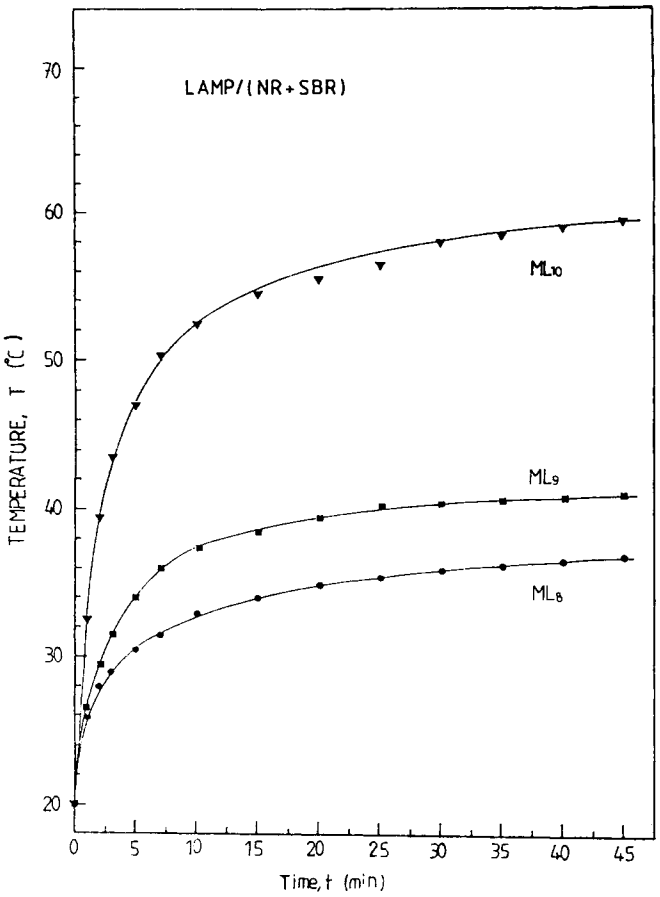

Fig. 3. Temperature–time dependence for LAMP/(NR + SBR) vulcanizates for an initial applied power of 10 W.

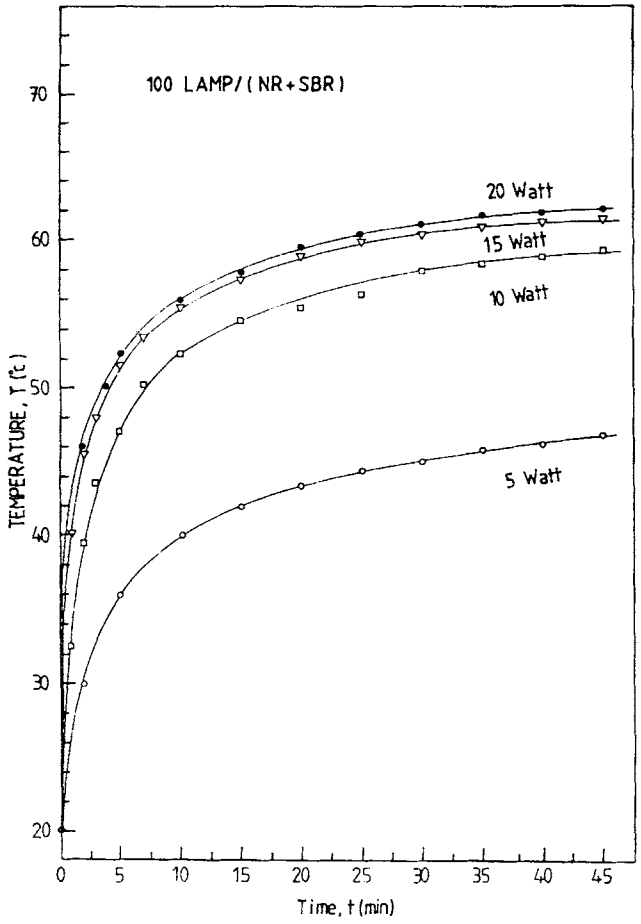

Fig. 4. Temperature–time dependence of 100 phr LAMP/(NR + SBR) vulcanizates at different values of the initial applied power.

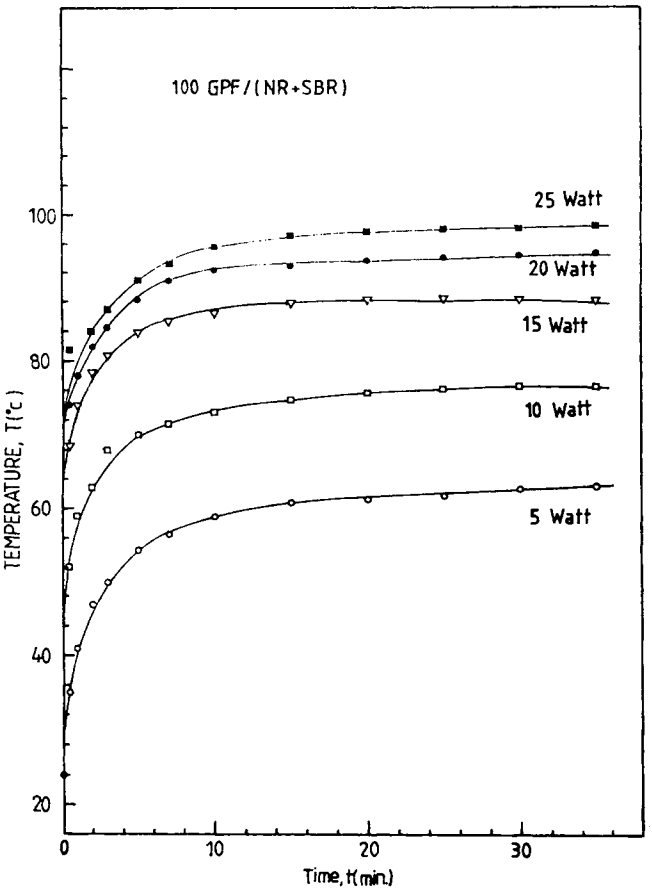

Fig. 5. Temperature–time dependence of 100 phr GPF/(NR + SBR) vulcanizates at different values of the initial applied power.

TABLE III
Dependence of τ on the Type and Concentration of Carbon Black as Well as the Applied Power

Sample	Power (W)	τ (min)
ML8	10	12
ML9	10	5.0
ML10	5	6.3
	10	4
	15	2.5
	20	2.3
ML10	5	3
	10	1.5
	15	0.5
	20	0.3
	25	0.2

temperature attains a steady value depending on the carbon black concentration.

The T–t dependence for samples loaded with 100 phr of LAMP black (Fig. 4) and 100 phr of GPF black (Fig. 5) at different values of the initial applied electric power shows that the variation in sample temperature with time obeys an exponentially growing function of the form

$$(T - T_0) = (T_m - T_0)[1 - \exp(-t/\tau)] \quad (1)$$

where T_0 is the ambient temperature, T_m is the ultimate temperature at-

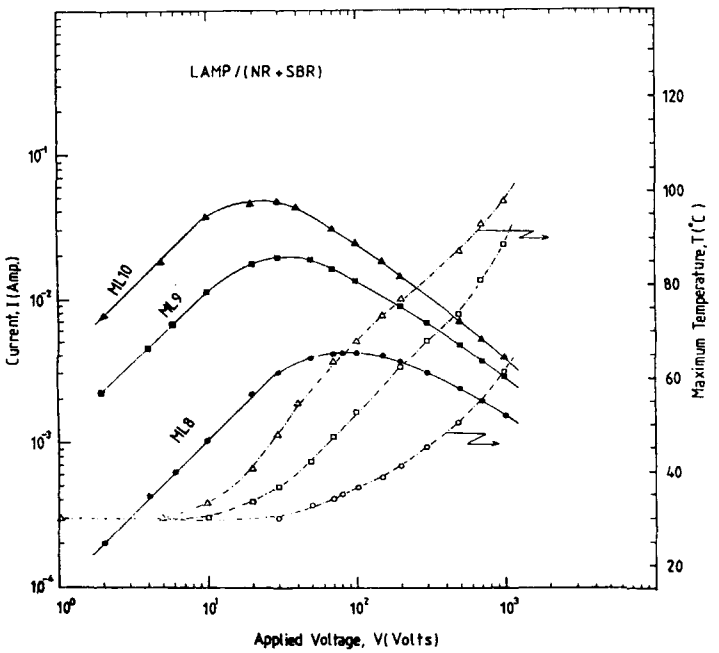

Fig. 6. $\ln I$ vs. $\ln V$ for LAMP black-loaded (NR + SBR) vulcanizates (solid lines). The dashed lines represent the maximum temperature attained inside the samples.

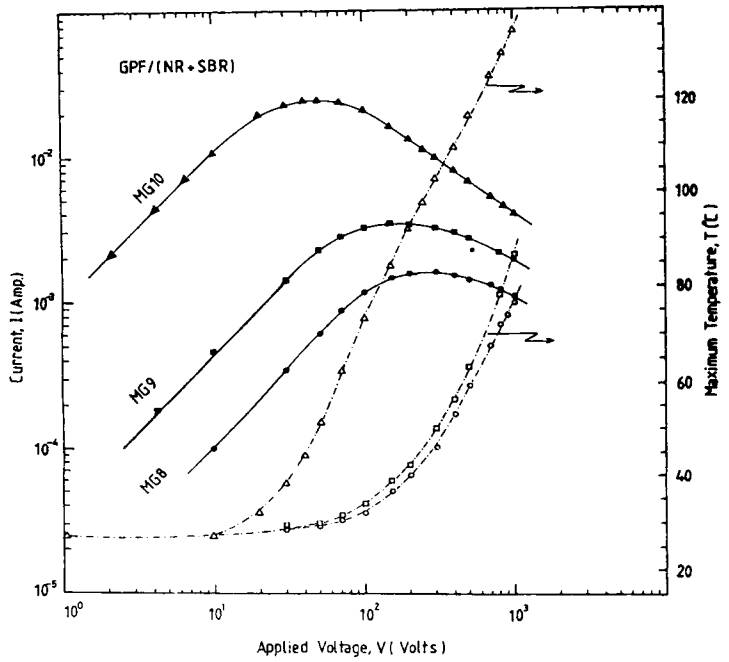

Fig. 7. $\ln I$ vs. $\ln V$ for GPF black-loaded (NR + SBR) vulcanizates (solid lines). The dashed lines represent the maximum temperature attained inside the samples.

tained, and τ is a characteristic time depending on the type and concentration of carbon black as well as on the value of the initial applied power. Table III presents the values of τ for the above samples at different concentrations and applied electric power. From the table it is shown that τ decreases with increasing carbon black concentration as well as the applied electric power.

Figures 6 and 7 illustrate $\log I$ vs. $\log V$ (solid curves) for NR/SBR vulcanizates loaded with LAMP and GPF black, respectively; the dashed curves represent the ultimate temperature attained inside the samples. At low

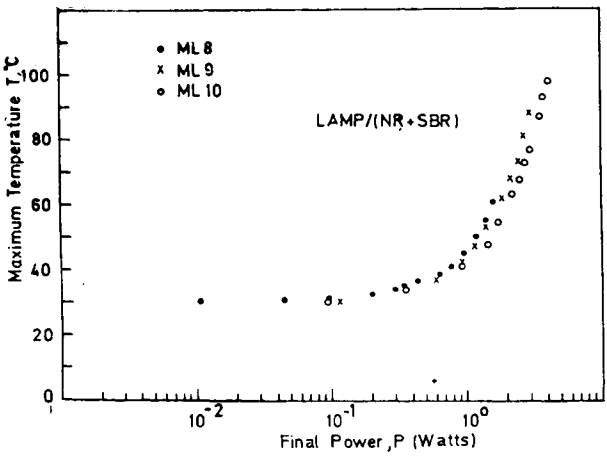

Fig. 8. Ultimate temperature vs. final power for LAMP/(NR + SBR) vulcanizates.

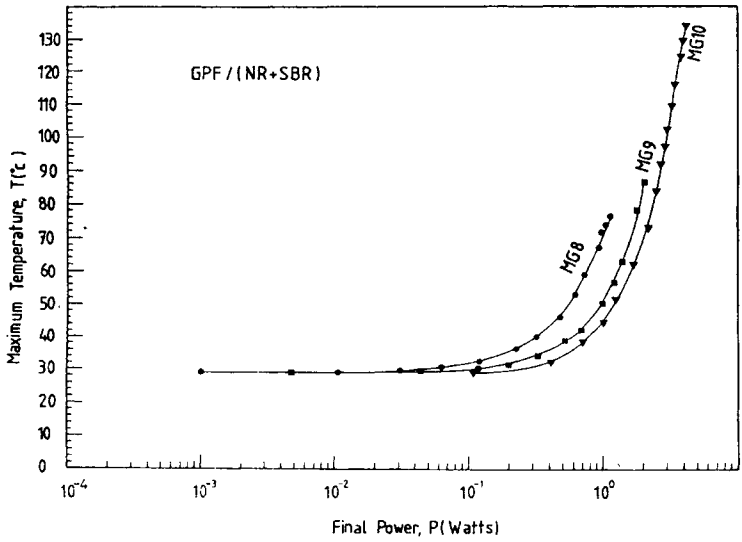

Fig. 9. Final temperature vs. final power for GPF/(NR + SBR) vulcanizates.

values of applied electric field, I increases linearly with V (ohmic behavior). As the power increases, a deviation from ohmicity is observed at a turn over voltage V^*. After this potential, the current decreases with increasing V leading to the so-called negative resistance behavior. It is worth mentioning that the value of V^* decreases with increasing carbon black concentration.

Figures 8 and 9 summarize the relationship between the working power and the ultimate temperature developed inside the sample. It was observed that samples containing 80 phr carbon black consumed a lower electric power to attain the same ultimate temperature compared with other concentrations.

CONCLUSIONS

From the above results it may be concluded that:

1. Negative values of the temperature coefficient of conductivity (TCC) were observed for NR/SBR blend heavily loaded with LAMP and GPF black. For rubber blend containing 80 phr LAMP the TCC value reaches -0.4 deg^{-1} at 125°C.
2. The temperature developed inside the sample by Joule heating effect as a function of time was found to obey an exponentially growing function with a characteristic time τ. This time is dependent on the type and concentration of carbon black as well as the value of the initial electric power applied on the sample.
3. The I–V characteristic showed a negative resistance behavior after a turn over potential V^* which was found to be dependent on both type and concentration of carbon black.
4. The sample containing 80 phr carbon black was found to consume less power to attain the same ultimate temperature compared with other carbon black concentrations.

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