

# Effect of Rubber Ratio, Carbon Black Level, and Accelerator Level on Natural Rubber/Bromobutyl Rubber Blend Properties

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**ABSTRACT:** Statistical experimental design, that is, response surface methodology, was used to predict and explain the effects of rubber ratio, carbon black, and accelerator level on the cure characteristics and physical properties of natural rubber/bromobutyl rubber (NR/BIIR) blends. With these three independent variables, 20 designed compounds were mixed by a two-roll mill and the scorch time, cure time, cure rate index, together with physical properties (hardness, tensile property, and compression set) were all determined by one operator. Multiple linear regression analysis was used to obtain response equations and thus contour plots, which illustrate the effects of the three independent variables on each property, as shown in detail by the diver-

sity of interactions between independent factors and each property. It was found that the carbon black level is the most significant influential factor on scorch time, cure time, tensile properties, hardness, and compression set. The difference in reactivity toward sulfur vulcanization of NR and BIIR resulted in cure behavior and physical properties that are dominated by the NR content in the rubber ratio factor. Finally, the response equations were shown to be useful for making accurate predictions. © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 90: 3059–3068, 2003

**Key words:** rubber; blends; curing; response surface methodology; physical properties

## INTRODUCTION

Blends of natural rubber (NR) with other synthetic rubbers have been widely studied.<sup>1–10</sup> The main reason is understandably the desire to achieve a balance of unique properties and cost. Bromobutyl rubber (BIIR) is a modified butyl rubber (IIR, isobutylene isoprene rubber), which has greater cure versatility than the unmodified rubber because of the bromine atoms attached to the carbon bond.<sup>11</sup> Its faster cure rate results in cure compatibility with other diene rubbers, thus enabling the development of useful blends. For example, BIIR has been blended with NR to produce a compound with higher damping and thermal stability.<sup>12–14</sup> However, there is no report of any statistically designed experiments that have been carried out to determine the effects of varying specific factors on the cure characteristic and physical properties of this blend compound. A statistically designed experiment has been one of the useful tools with which quantitative relationships may be defined by a mathematical equation. Many informative studies

have been published on experimental designs.<sup>15–25</sup> Krakowski and Tinker's work<sup>24,25</sup> is one such example that has given an excellent introduction and discussion on using a response surface methodology designed experiment examining NR/BR blends. The present work uses the same designed experimental method, but rather than detailing the statistical method used, it investigates the effect of a wide range of NR/BIIR ratios and both carbon black and accelerator levels on cure characteristics and general physical blend properties.

## EXPERIMENTAL

Response surface methodology using central composite rotatable designs is based on a complete two-level factorial design, which is then supplemented by additional points to enable the curvature of the response surface and the experimental error to be estimated. The experimental points are identified by code values, which assign five levels to each variable. The coded five levels are  $-a$ ,  $-1$ ,  $0$ ,  $+1$ , and  $+a$ , in which  $a = 2^{k/4}$  and  $k$  is the number of independent variables in the experiment. In this work, three variables were investigated: rubber blend ratio, carbon black level, and accelerator level. The range of selected data was defined as 0.25 to 4, 0 to 60, and 0.75 to 1.50 for rubber

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TABLE I  
Relationship Between Real and Code Values

Code value	STR5L/BIIR ratio	Carbon black level (phr)	TBBS level (phr)
-1.682	0.25 (20/80)	0	0.75
-1	1.01 (50/50)	12.16	0.90
0	2.12 (68/32)	30	1.12
+1	3.24 (76.4/23.6)	47.84	1.35
+1.682	4.00 (80/20)	60	1.50

blend ratio, carbon black level, and accelerator level, respectively. The real values are related to the assigned coded experimental points and scale of variable ( $S$ ) according to eqs. (1) and (2):

$$S = \text{Range}/(2 \times a) \quad (1)$$

$$R = (S \times \text{code}) + \text{Mean} \quad (2)$$

Table I shows the real values in relation to the code values. An example of a calculation is shown as follows:

$$\begin{aligned} \text{Scale of NR/BIIR ratio} &= (4.00 - 0.25)/(2 \times 2^{(3/4)}) \\ &= 1.115/\text{coded unit} \end{aligned}$$

for -1 on the coded scale,

$$\text{Real NR/BIIR ratio} = (-1)(1.115) + 2.125 = 1.01$$

Table II gives the full experimental design and real value for each point. The design provides for eight factorial compounds (mix numbers 1-8), which enable modeling of linear and second-order interactive ef-

fects; seven star points,<sup>9-14</sup> which allow for modeling of quadratic curvature; and replication of the center point six times,<sup>15-20</sup> which provides an assessment of error and model adequacy. Through the use of multi-variable linear regression analysis, the data are fitted to a second-order response surface equation of the general form

$$Y = b_0 + \sum_{i=1}^k b_i X_i + \sum_{i=1}^k b_{ii} X_i^2 + \sum_{i=1}^k \sum_{j=1}^k b_{ij} X_i X_j \quad (3)$$

in which  $Y$  is a response or dependent variable to be fitted,  $X_i$  and  $X_j$  are independent variables,  $b_0$  is the constant term,  $b_i$  is the linear coefficient,  $b_{ii}$  is the quadratic coefficient,  $b_{ij}$  is the interaction coefficient ( $i \neq j$ ), and  $k$  is the number of variables. Therefore, for a three-variable experimental design, the equation is

$$\begin{aligned} Y = & b_0 + b_1 X_1 + b_2 X_2 + b_3 X_3 + b_{11} X_1^2 + b_{22} X_2^2 + b_{33} X_3^2 \\ & + b_{12} X_1 X_2 + b_{13} X_1 X_3 + b_{23} X_2 X_3 \quad (4) \end{aligned}$$

An estimate of the variance was obtained from the center points and the usual formulation for standard deviation (SD), that is

TABLE II  
Experimental Design

	Mix no.	Code			Real value		
		NR/BIIR	C-Black	TBBS	NR/BIIR	C-Black (phr)	TBBS (phr)
Factorial points	1	-1	-1	-1	1.01	12.16	0.90
	2	+1	-1	-1	3.24	12.16	0.90
	3	-1	+1	-1	1.01	47.84	0.90
	4	+1	+1	-1	3.24	47.84	0.90
	5	-1	-1	+1	1.01	12.16	1.35
	6	+1	-1	+1	3.24	12.16	1.35
	7	-1	+1	+1	1.01	47.84	1.35
	8	+1	+1	+1	3.24	47.84	1.35
Star points	9	-1.682	0	0	0.25	30	1.12
	10	+1.682	0	0	4.00	30	1.12
	11	0	-1.682	0	2.12	0	1.12
	12	0	+1.682	0	2.12	60	1.12
	13	0	0	-1.682	2.12	30	0.75
	14	0	0	+1.682	2.12	30	1.50
Center-point replicates	15-20	0	0	0	2.12	30	1.12

**TABLE III**  
**Based Formulation**

Ingredient	phr
STR5L/BIIR	Variable (0.25–4)
HAF N-330	Variable (0–60)
TBBS <sup>a</sup>	Variable (0.75–1.5)
Zinc oxide	3
Stearic acid	2
6PPD <sup>b</sup>	1
Sulfur	1

<sup>a</sup> TBBS, *N-tert-butyl-2-benzothiazyl sulfenamide*.

<sup>b</sup> 6PPD, *N-phenyl-N'-1,3-dimethylbutyl-p-phenylenediamine*.

$$SD = \sqrt{\frac{\sum (x_i - \bar{x})^2}{n - 1}} \quad (5)$$

where  $n$  is the number of center points. A test of significance was made on each term in the regression equation using the following standard errors (SE)<sup>18</sup>:

$$SE(b_i) = 0.271(SD) \quad (6)$$

$$SE(b_{ii}) = 0.263(SD) \quad (7)$$

$$SE(b_{ij}) = 0.354(SD) \quad (8)$$

where  $SE(b_i)$  is the standard error of  $b_1$ ,  $b_2$ , and  $b_3$ ;  $SE(b_{ii})$  is the standard error of  $b_{11}$ ,  $b_{22}$ , and  $b_{33}$ ; and  $SE(b_{ij})$  is the standard error of  $b_{12}$ ,  $b_{13}$ , and  $b_{23}$ .

From Table II, 20 compounds were prepared based on formulations in Table III. The rubbers used in this study were NR (STR5 L) manufactured by Tavorn Manufacturing Ltd. (Songkla, Thailand) and BIIR from Polysar Bromobutyl X2, (Bayer, Bangkok, Thailand) with 1.89% bromine content. Rubber chemicals were standard commercial-grade materials. Com-

pounding was performed by mixing in a laboratory two-roll mill (diameter 6 in.; length 20 in.; Chaijarean-kanchang Co. Ltd., Bangkok, Thailand) at room temperature. The friction speed ratio of front roll to back roll was 1.25 : 1. Two rubbers were first masticated and blended for 5 min. The remaining ingredients were zinc oxide, antioxidant [*N-1,3-dimethylbutyl-N'-phenyl-p-phenylenediamine* (6PPD)], accelerator [*N-tert-butylbenzothiazole-2-sulfenamide* (TBBS)], carbon black (HAF N330), stearic acid, and sulfur (all obtained from Pedthai Chemical Products, Bangkok, Thailand), which were mixed for another 15–30 min depending on the carbon black level.

After thorough mixing to ensure good dispersion, the compound was removed from the mill and stored at room temperature for 24 h before testing. The cure behavior of the compounds was determined at 160°C by a Monsanto oscillating disc rheometer (Model ODR 2000, Monsanto, St. Louis, MO) at 1° arc (ASTM D 2084-88). Tensile properties, hardness, and compression set were determined according to ASTM D 412-98, ASTM D 2240-97, and ASTM D 395-98, respectively. Statistical results were analyzed by multiple linear regression analysis as detailed in Diamond.<sup>18</sup> Contour curves were displayed using Maple V Release 4 software (Waterloo Maple Software, Ontario, Canada).

## RESULTS AND DISCUSSION

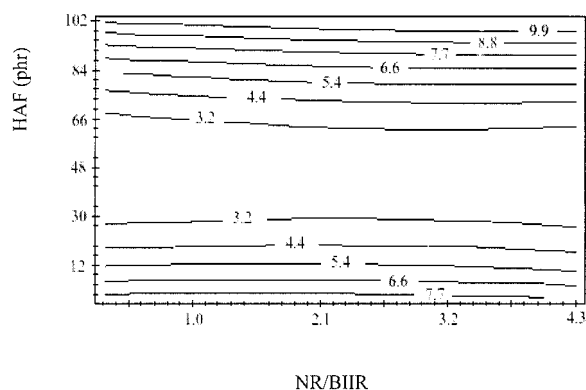
### Cure behavior

Table IV shows the calculated response equations for the parameters measured in the entire experiment. A resulting response equation for the relation between the three variables and the scorch time is therefore

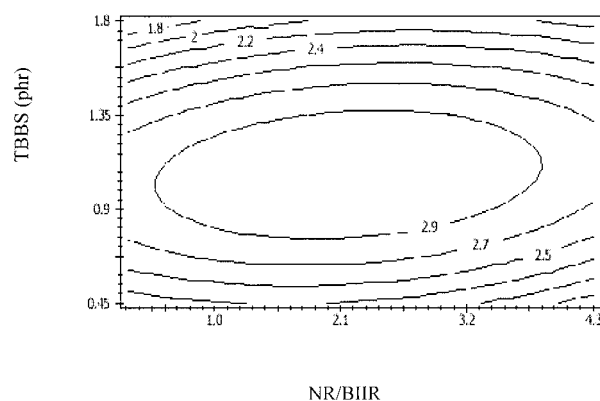
**TABLE IV**  
**Coefficients for Response Equations of Cure Characteristics**

Parameter <sup>a</sup>	$T_2$ (min)	$T_{90}$ (min)	CRI (%)	$M_L$ (lb.-in.)	$M_H$ (lb.-in.)	$M_H - M_L$ (lb.-in.)
$b_0$	3.109	6.166	33.063	5.156	23.989	18.833
$b_1$	0.018	-1.283	6.326	-0.517	1.541	2.058
$b_2$	-1.540	-1.730	1.806	2.047	7.008	4.961
$b_3$	-0.057	-0.432	3.661	0.031	1.674	1.643
$b_{11}$	-0.083	0.987	-2.876	0.598	-0.268	-0.866
$b_{22}$	0.867	0.747	-0.454	0.271	0.818	0.546
$b_{33}$	-0.104	-0.309	0.445	-0.061	-0.280	-0.219
$b_{12}$	0.073	0.034	0.768	-0.062	0.181	0.244
$b_{13}$	0.030	0.081	0.768	-0.020	-0.341	-0.321
$b_{23}$	0.090	0.119	0.034	0.345	1.084	0.739
SD	0.1157	0.1949	1.5047	0.7921	1.1008	0.4118
$SE(b_i)$	0.0314	0.0528	0.4078	0.2147	0.2983	0.1116
$SE(b_{ii})$	0.0304	0.0513	0.3957	0.2083	0.2895	0.1083
$SE(b_{ij})$	0.0410	0.0690	0.5327	0.2804	0.3897	0.1458

<sup>a</sup>  $b_0$ , constant;  $b_1$ , NR/BIIR;  $b_2$ , HAF level;  $b_3$ , TBBS level.



**Figure 1** Effect of HAF level and NR/BIIR ratio on scorch time at TBBS coded 0 (1.12 phr).



**Figure 2** Effect of TBBS level and NR/BIIR ratio on scorch time at HAF coded 0 (30 phr).

$$\begin{aligned}
 T_{s2} = & 3.109 + 0.018(\text{NR/BIIR}) - 1.540(\text{HAF}) \\
 & - 0.057(\text{TBBS}) - 0.083(\text{NR/BIIR})^2 + 0.867(\text{HAF})^2 \\
 & - 0.104(\text{TBBS})^2 + 0.073(\text{NR/BIIR})(\text{HAF}) \\
 & + 0.030(\text{NR/BIIR})(\text{TBBS}) + 0.090(\text{HAF})(\text{TBBS}) \quad (9)
 \end{aligned}$$

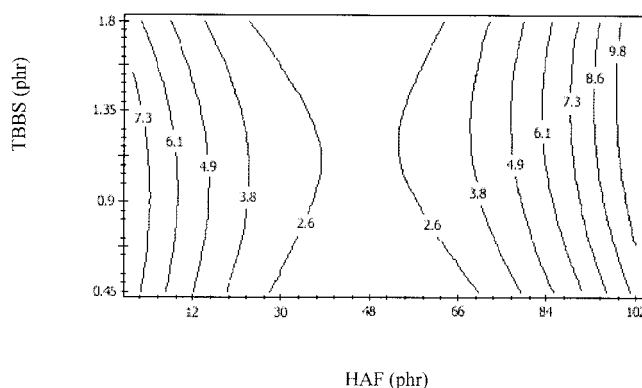
where NR/BIIR, HAF, and TBBS can be any value of NR/BIIR ratio, HAF, and TBBS level in coded terms, respectively.

The coefficients for the rubber ratio, HAF level, and TBBS level are relative magnitude at 0.018,  $-1.540$ , and  $-0.057$ , respectively. They indicate the comparative effect of each of those factors on the scorch time response, together with the direction of the effect. That is, the HAF level is the most significant factor affecting the scorch time of the compound followed by the level of TBBS and the NR/BIIR ratio, respectively. Figure 1 clearly illustrates the contour plot of the fitted equation indicating the effect of HAF and NR/BIIR ratio on the scorch time of the compound (where ratio of TBBS is constant at coded 0, 1.12 phr). An increase in the amount of HAF up to 30 phr resulted in a decrease in the scorch time. This is because the surface chemistry and the pH of carbon black have greater influence on the extent of vulcanization. Channel blacks, which contain a number of surface oxygen functional groups, such as quinones, hydroquinones, phenolic hydroxyls, carboxylic acids, and lactones, have been reported to be acidic, which tend to retard cure. Furnace blacks are characterized by a neutral pH or slightly alkali with low oxygen content have an acceleration effect.<sup>26</sup> Consequently, the furnace black used (HAF with pH 8.7) accelerates the cure behavior and decreases the scorch time. However, increasing the level of HAF to more than 66 phr lowered the volume of rubber that delayed the curing.

The effect of increasing the TBBS level up to 0.9 phr delayed the scorch, whereas levels higher than 1.3 phr caused a decrease in scorch time (Fig. 2). TBBS is a delayed-action accelerator in a benzothiazole sulfen-

amide group. The accelerator becomes active as the tertiary butyl amine splits off during vulcanization.<sup>27</sup> The base activates 2-mercaptobenzothiazole as it is formed. Consequently, TBBS delays the start of vulcanization. However, the high amount of TBBS ( $>1.3$  phr) that decreased scorch time can be attributed to the high amount of both amine and MBT, which together accelerate the start of vulcanization. Figure 3 additionally shows that at a fixed ratio of NR/BIIR, TBBS has little effect on the scorch time compared to the HAF level. The result also shows a shaped surface with maximum scorch time in the direction of low HAF, low TBBS, high HAF, and high TBBS levels.

Because every property is analyzed over the same ranges of the variables (i.e., over the same grid), they can be superimposed. Two cure characteristics of 90% cure time ( $T_{90}$ ), and cure rate index (CRI) can be seen in Figure 4. The most influential factor affecting cure time in the linear coefficient is the carbon black level, followed by the rubber ratio and the TBBS level, respectively. The coefficient for HAF is 1.3 and 4 times higher than that of the rubber ratio and the TBBS level, respectively, whereas the NR/BIIR ratio has the greatest significance in the quadratic coefficient. When the



**Figure 3** Effect of carbon black and TBBS level on scorch time at NR/BIIR coded 0 (68/32).

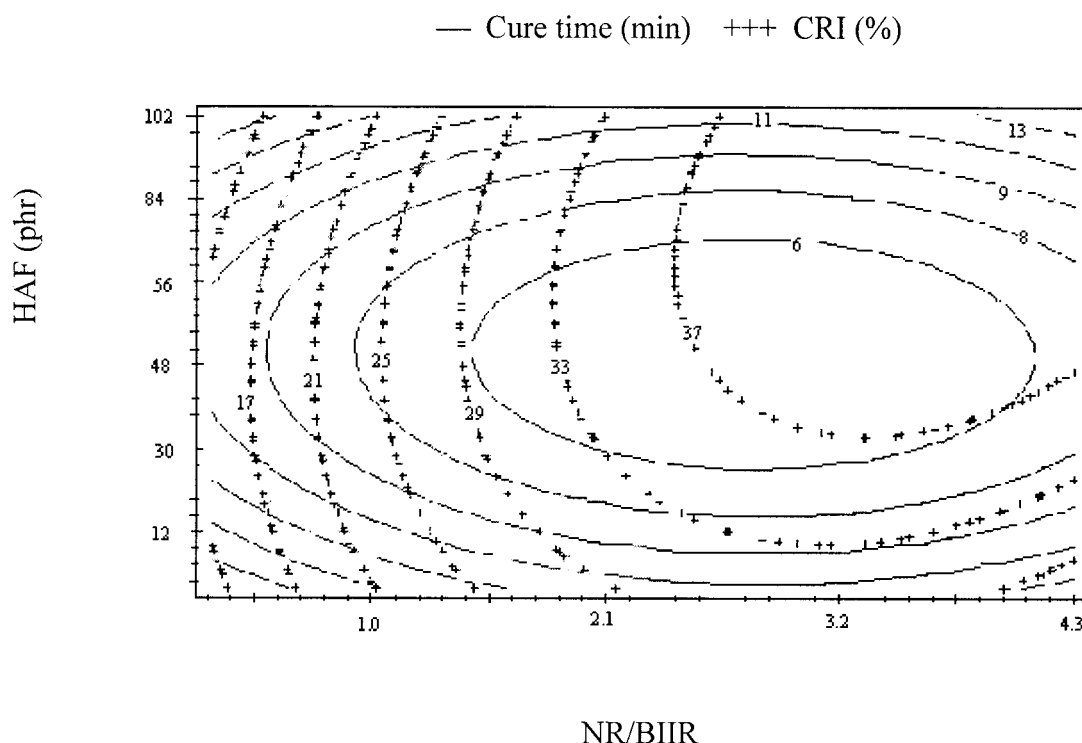


Figure 4 Effect of HAF level and NR/BIIR ratio on cure time and cure rate index at TBBS coded 0 (1.12 phr).

level of TBBS is constant at 1.12 phr, the effect of increasing the level of HAF on the cure time (Fig. 4) is, interestingly, very similar to its effect on the scorch time. An increase in HAF up to 30 phr resulted in a shorter cure time, but values greater than 66 phr increased the cure time possibly because of the low volume fraction of rubber. Increasing the NR/BIIR ratio or increasing the NR content caused a decrease in the cure time because of the greater reactivity of carbon double bonds on a natural rubber molecule. Figure 5 shows the dependency of cure time on the NR/BIIR ratio and of the TBBS level when the HAF level is at the center point of the design, 30 phr. The figure indicates that TBBS has little significant effect on the cure time when the NR/BIIR ratio is less than 2.1 (NR/BIIR, 68/32), but that it is a significant factor, by reducing the cure time, when the NR/BIIR ratio is more than 2.1. As expected, high levels of both TBBS and NR content produced the shortest cure time.

The CRI, which indicates the rate of cure of the compounds, is defined as  $100/(T_{90} - T_2)$ . A higher value means a higher rate of vulcanization. It can be seen from Table IV and Figures 4 and 5 that the NR/BIIR ratio has the greatest effect: it has about 1.7 and 3.5 times the effect of TBBS and HAF level, respectively, and TBBS is about twice as effective as HAF. Increasing the NR content and the TBBS level increased the CRI as expected, whereas increasing the HAF level resulted in little change in the cure rate.

Table IV also shows the coefficients of response equations for minimum torque ( $M_L$ ), maximum torque

( $M_H$ ) and their difference ( $M_H - M_L$ ). The minimum torque roughly indicates the compound viscosity and is most affected by the HAF level. The NR/BIIR ratio and TBBS have only a slight effect on  $M_L$ .  $M_H - M_L$  also indicates the state of cure of the compound and the HAF level is the most influential factor followed by the rubber ratio and TBBS, respectively. Increasing the HAF level, rubber ratio (increase in NR content), and TBBS level all increase the  $M_H - M_L$ , suggesting an increase in modulus of the vulcanizates.

### Physical properties

Table V and Figure 6 show that the stress required to achieve 300% strain, ultimate tensile strength, and the elongation at break were all affected most significantly by the reinforcing carbon black HAF level. As expected, HAF reduces the elongation at break and increases the stress required to achieve 300% strain and the ultimate tensile strength. However, increasing the HAF level above 66 phr lowered the ultimate tensile strength because of the reduction of rubber volume fraction. Increasing the NR content also served to dramatically affect the ultimate tensile strength: the variable of the NR/BIIR ratio is 8.5 times greater than that of TBBS. The average effect of increasing the NR level was to increase the ultimate tensile strength by about 29% in the range used in the experiment (Fig. 7). The strain crystallization of NR contributes to the high tensile strength of the vulcanizates. It may also be observed from Table IV that HAF and TBBS level have

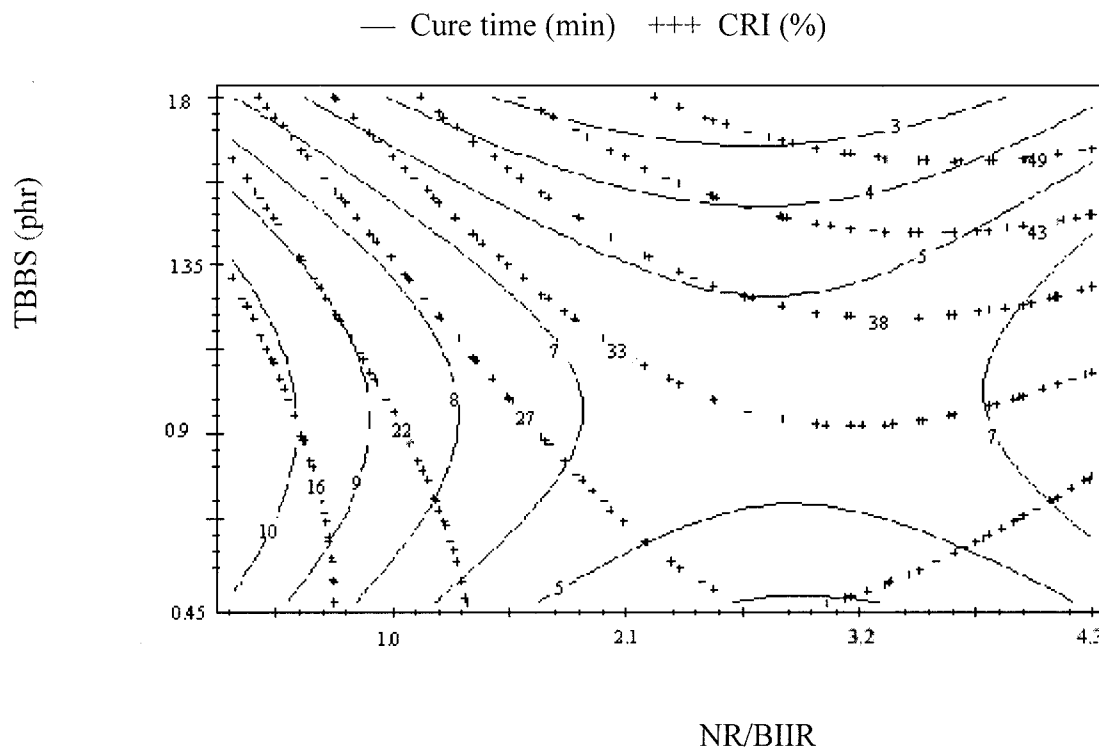


Figure 5 Effect of TBBS level and NR/BIIR ratio on cure time and cure rate index at HAF coded 0 (30 phr).

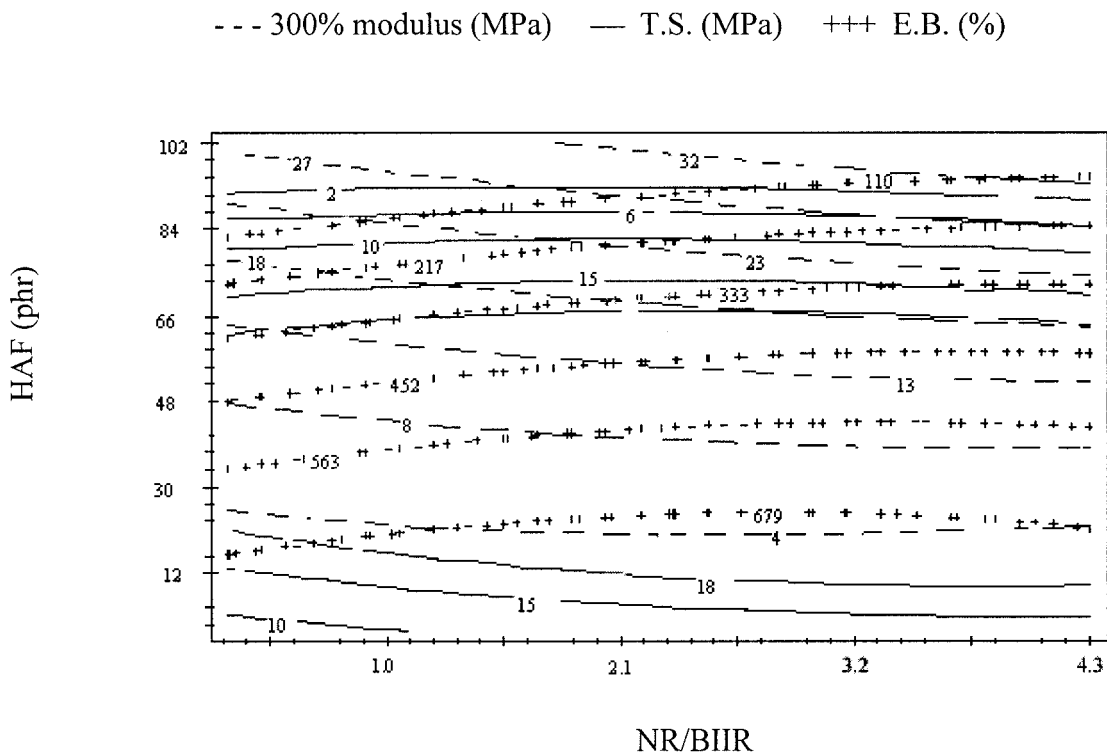
greater influence on 300% modulus than in ultimate tensile strength comparing the coefficient of 4.252 to 2.576 and 0.615 to 0.276 for HAF and TBBS levels, respectively. This supports an important role of the reinforcement carbon black on modulus and hardness of the filled vulcanizates. For accelerated-sulfur vulcanization, an increase in accelerator concentration (TBBS level) results in an increase in crosslink precursors, given that the higher amount of accelerator can react with sulfur to give more monomeric polysul-

fides. The higher monomeric polysulfides then interact with rubber to form higher polymeric polysulfides (crosslink precursors). Finally, the rubber polysulfides react to give more crosslinks.<sup>28</sup> A higher modulus vulcanized rubber with a higher number of crosslinks should then be found with increasing amount of TBBS. However, NR/BIIR has a greater influence on tensile strength (2.410) higher than 300% modulus (0.428) and hardness (1.115). This indicates that the rubber type and content, and the strength of raw rubber have a

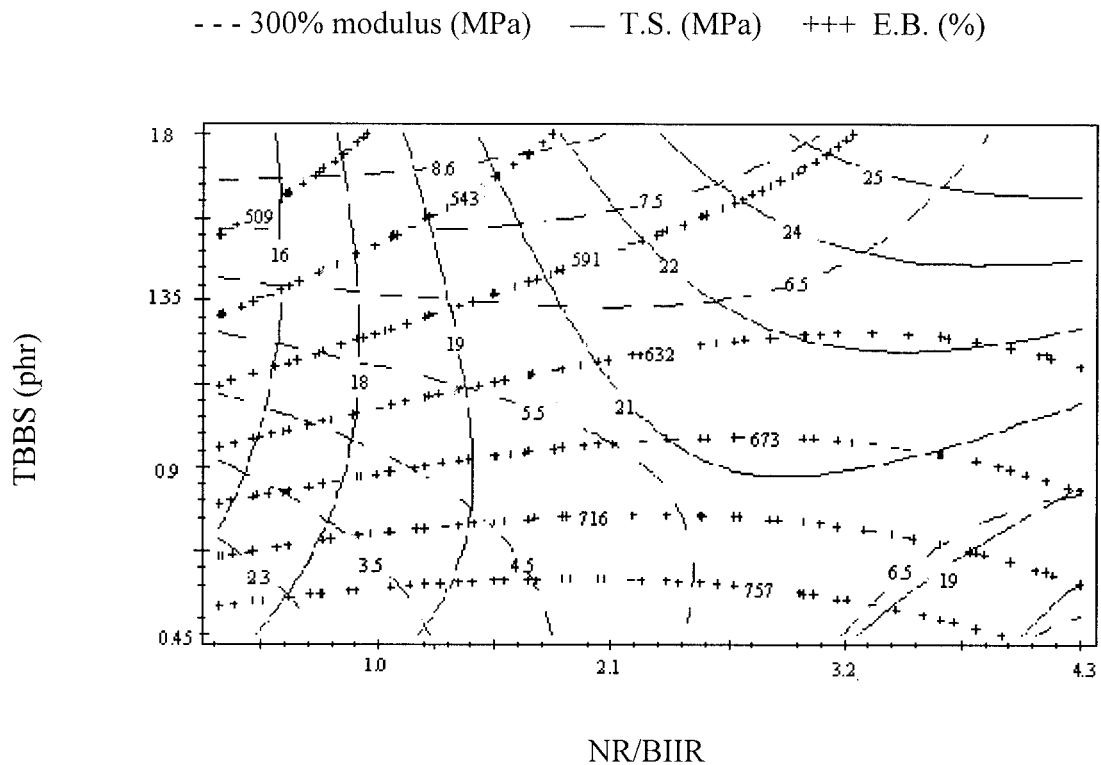
TABLE V  
Coefficients of Response Equations for Physical Properties

Parameter <sup>a</sup>	Hardness (Shore A)	300% Modulus (MPa)	Tensile strength (MPa)	Elongation at break (%)	Compression set (%)
$b_0$	45.398	5.902	20.775	642.749	54.091
$b_1$	1.115	0.428	2.410	15.644	2.574
$b_2$	11.122	4.252	2.576	-111.255	5.073
$b_3$	1.839	0.615	0.276	-38.860	-2.120
$b_{11}$	-0.619	-0.197	-0.450	-9.476	1.555
$b_{22}$	-0.708	0.647	-2.478	-13.896	-1.548
$b_{33}$	-0.354	0.099	0.046	3.786	2.419
$b_{12}$	-0.188	0.503	-0.356	7.813	-2.122
$b_{13}$	-0.438	-0.467	0.299	7.813	1.023
$b_{23}$	-0.188	0.636	-0.441	-17.188	-4.187
SD	1.1584	0.7414	0.8031	20.4157	8.4699
SE ( $b_i$ )	0.3139	0.2009	0.2176	5.5327	2.2953
SE ( $b_{ii}$ )	0.3047	0.1950	0.2112	5.3693	2.2276
SE ( $b_{ij}$ )	0.4101	0.2625	0.2843	7.2272	2.9983

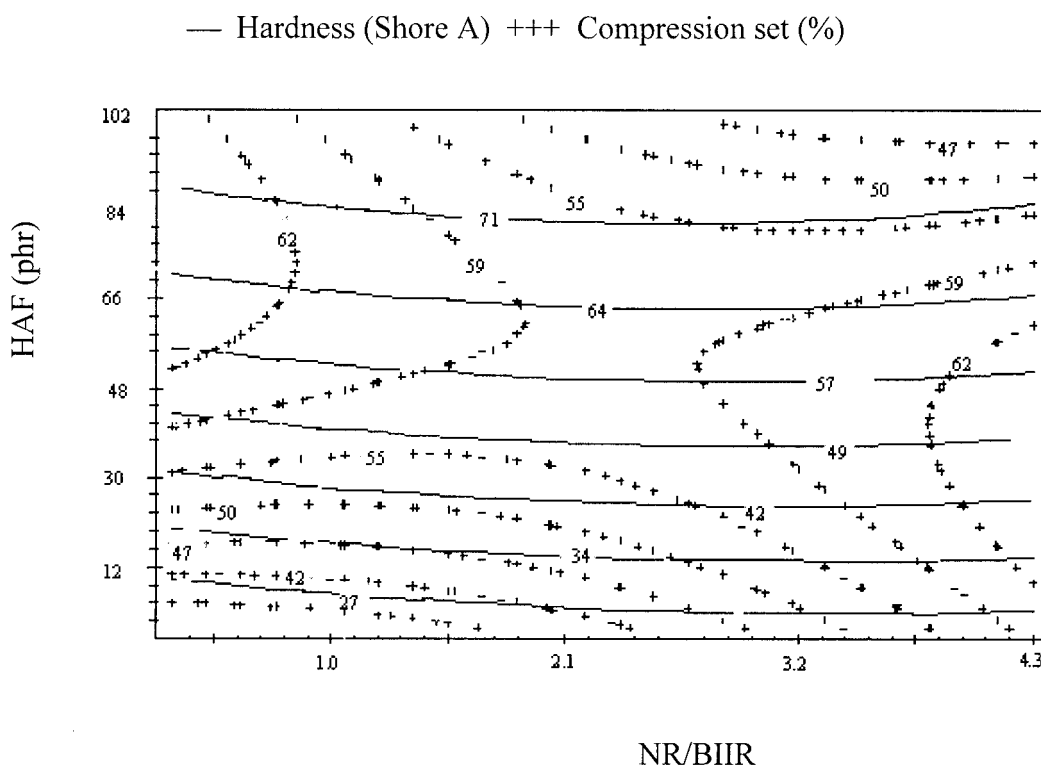
<sup>a</sup> See Table IV for parameter definitions.



**Figure 6** Effect of HAF level and NR/BIIR ratio on 300% modulus, tensile strength, and elongation at break at TBBS coded 0 (1.12 phr).



**Figure 7** Effect of TBBS level and NR/BIIR ratio on 300% modulus, tensile strength, and elongation at break at HAF coded 0 (30 phr).



**Figure 8** Effect of HAF level and NR/BIIR ratio on hardness and compression set at TBBS coded 0 (1.12 phr).

significant influence on the tensile strength of the vulcanized rubber. Thus the important role of each parameter to meet required properties should be well recognized. The elongation at break was also found to be more affected by the TBBS level than by the NR/BIIR ratio. This can easily be seen from the opposite directions of linear coefficients:  $-38.86$  for the TBBS level and  $15.64$  for the NR/BIIR ratio. Increasing the level of TBBS serves to decrease the elongation at break because it helps to form a greater number of crosslinks, as mentioned before. A tighter molecular network with less extension was thus found. Nevertheless, increasing the rubber ratio tended to increase the elongation at break especially at the high level of TBBS (Fig. 7), most probably because of the high elasticity of natural rubber. However, the reverse effect was observed after the rubber ratio reached about 2.8, the elongation at break decreasing with increasing NR content. This can be related to the possibly greater number of carbon-carbon double bonds on NR molecules, which give more crosslink sites toward sulfur vulcanization reaction resulting in a higher crosslink formation with a tighter network and less extend.

Figure 8 shows the dependency of hardness and percentage of compression set on the three variable factors. The hardness property, like the 300% modulus, was found to be affected most by the level of HAF, followed by the TBBS level and the NR/BIIR ratio, respectively, which were attributed to the same reasons. The compression set property provides insight

into the ability of rubber compounds to retain elastic properties after the prolonged action of compressive stresses. The most influential factor affecting compression set was the HAF level. It had an influence of 2 and 2.5 times greater than the NR/BIIR ratio and the TBBS level, respectively. The contour curve in Figure 8 interestingly reflects the response of compression set to the HAF level and the NR/BIIR ratio, when the TBBS is constant at the center point of 1.12 phr. It shows distinctly different response regions. First, an increase in the carbon black level to more than 60 phr, while increasing the NR content, tended to produce a lower compression set. Because the high active surface of carbon black could interact more with the NR molecules to combine with a tight crosslinking network, the high reactivity of the double bond on the NR (at the same time, the high load reinforcing the carbon black) will enhance the elastic ability to bear the loaded stresses and recover on the stresses released. This will enable desirable low compression set formulations to be produced. When the amount of HAF was lower than 60 phr, increasing the NR content resulted in an increase in the compression set. It indicates the poor heat resistance of NR with a less tight crosslinking network at a lower level of carbon black. The TBBS level is also expected to exercise a significant effect on compression set as shown in Figure 9. At a fixed amount of 30 phr carbon black, increasing amounts of TBBS up to the center point of 1.12 phr reduced the compression set, possibly because of an increase in



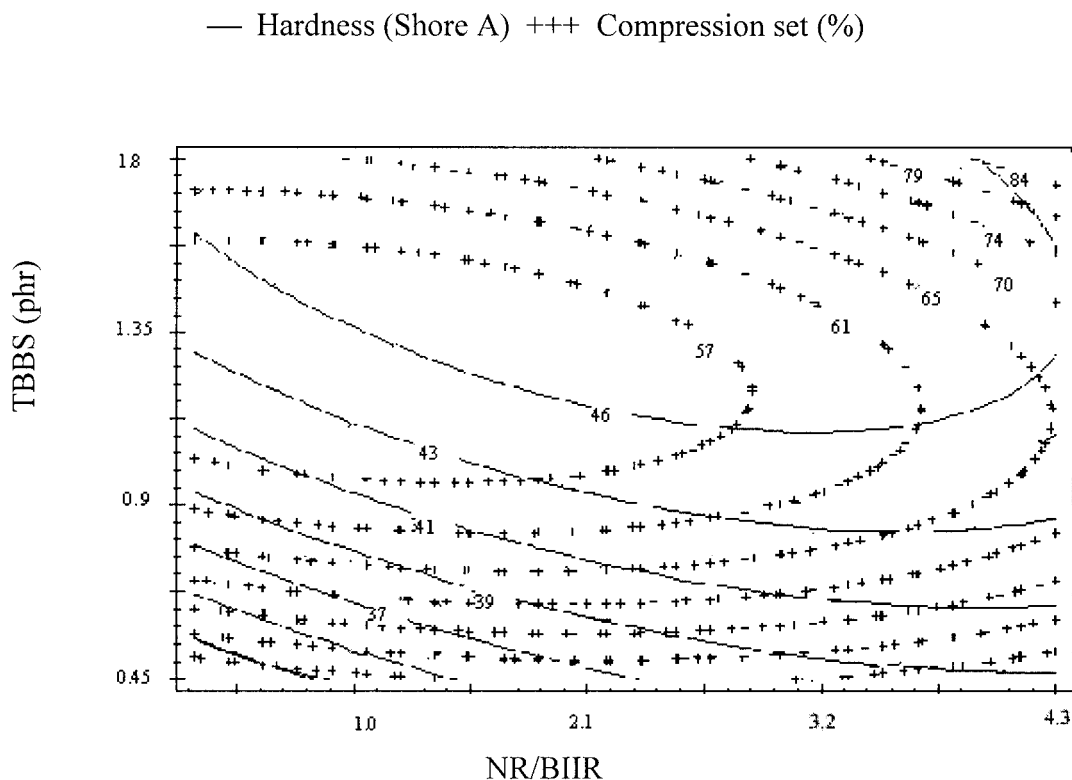


Figure 9 Effect of TBBS level and NR/BIIR ratio on hardness and compression set at HAF coded 0 (30 phr).

crosslink precursors to form more crosslinks. This increases the retractile force and reduces the amount of permanent deformation remaining after removal of the deforming force, whereas increasing the rubber ratio tended to slightly increase the compression set. Increasing the level of TBBS higher than 1.4 phr, however, increased the compression set. This is because TBBS is also a sulfur-donor accelerator, which at high levels may produce more polysulfidic crosslinking networks. This network can be easily broken down by heat, causing poor elastic recovery and thus increasing the compression set. However, at this high level of TBBS a reduction trend of compression set with increasing rubber ratio was observed, suggesting that a higher elasticity of higher NR content would overrule the effect.

Figure 10 illustrates the use of superimposing contour plots to determine compound composition with specific properties. The unshaded area provides many combinations of the rubber ratio and TBBS level at a constant HAF level of 30 phr in compounds that will give 5.5 MPa minimum 300% modulus, 19 MPa minimum tensile strength, and 57% maximum compression set. The two formulations based on TBBS, NR/BIIR at 1.35 : 2.1 (68/32) and 1.13 : 2.5 (71.5/28.5), were compounded, vulcanized, and determined all the properties. These fitted the desired properties well, which shows that the equations are useful for making accurate predictions.

CONCLUSIONS

Contour plots were generated from response equations in this study and were found to be very useful as an indication of the change in any particular property with change in any parameter. Optimum properties can easily be obtained, with an acceptable degree of

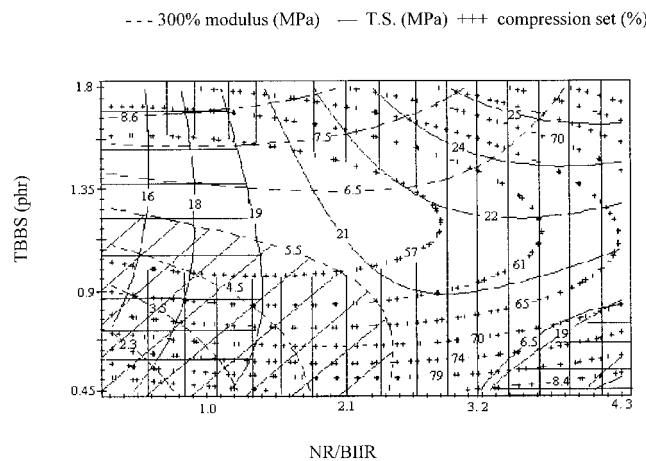


Figure 10 Use of superimposing contour plots to determine compound composition with specific properties. Unshaded areas provide many combinations of the rubber ratio and TBBS level at a constant HAF level of 30 phr in a compound that will give 5.5 MPa minimum 300% modulus, 19 MPa minimum tensile strength, and 57% maximum compression set.

accuracy, by superimposing the contour plots. The carbon black level had the most significant influence on scorch and cure time and all of the physical properties.

Because NR is more reactive to sulfur vulcanization and has a higher strength than that of BIIR it is strongly reflected in the response of cure behavior and most physical properties to the rubber ratio. Compression set is more dependent on the sulfur crosslinking network: an increase in the TBBS level up to a one-point decrease in compression set, but at a higher level, may produce an increase in polysulfide crosslinks, which in turn would increase the compression set.

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