

## Modeling the Hyperviscoelastic Behavior of a Tire Tread Compound Reinforced by Silica and Carbon Black

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**ABSTRACT:** This research study was devoted to the modeling of the mechanical behavior of three carbon black/silica natural rubber/butadiene rubber (NR/BR) compounds under a tensile load. These compounds were prepared on a two-roll mill, and the tensile testes were carried out on a dumbbell-shaped specimen and three rubber-strip specimens with different widths. Heat buildup tests were also performed, and the temperature rise was determined. The time-independent behavior of the rubber was described by traditional hyperelastic models, including those of Marlow and Yeoh. The viscoelastic behavior was studied with two linear (Prony series) and nonlinear [Bergstrom–Boyce (BB)] models. A previously developed methodology by the authors, which was based on the finite element modeling of the tension of the rubber strips, was used to determine the parameters of the mentioned equations. It was shown that neglecting the viscoelasticity would have given rise to large errors in predicting the mechanical deformation in the rubbers. However, the linear viscoelastic model failed to predict the correct behavior at a large strain, particularly for wider samples. On the other hand, not only could the combination of the Yeoh hyperelastic model with the BB equation describe the mechanical behavior at low to medium strains, but also the large strains were taken into consideration. Both the linear and nonlinear hyperviscoelastic models accurately described the hysteresis in rubbers, and this could be used for the evaluation of the rolling resistance in tires. © 2012 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 000: 000–000, 2012

**KEYWORDS:** modeling; rubber; theory; viscosity; viscoelastic properties

Received 2 May 2012; accepted 20 June 2012; published online

**DOI:** 10.1002/app.38242

### INTRODUCTION

The proper description of the mechanical behavior of vulcanized rubber compounds is a crucial step during finite element analysis of elastomeric products. Not only must this behavior be described by an appropriate hyperelastic model, but also, in several cases, time effects such as viscoelasticity should also be included in the material model. For example, the modeling of tire rolling resistance needs a hyperelastic model to be employed in conjunction with a viscoelastic equation to accurately simulate the dissipation of the energy during the analysis. However, although many studies have been carried out to develop hyperelastic<sup>1–11</sup> and viscoelastic models, few attempts have been made to study hyperviscoelasticity in cured rubbers.<sup>12–15</sup> Recently, a combination of the hyperelastic models with the Prony series model, which resembles the hyperviscoelasticity in rubbery materials, has been studied.<sup>16</sup> A new method was developed, which was based on the numerical fitting of the experimental

data of the tension of three rubber-strip samples into a hyperviscoelastic model by the use of the finite element method. In that study, an natural rubber/butadiene rubber (NR/BR) rubber compound reinforced by carbon black was selected. However, the application of the Prony series is limited to the linear viscoelasticity. This is a serious problem because filler–filler and filler–polymer interactions and also the difficulties associated with large deformation should be taken into consideration by nonlinear viscoelastic models, especially when silica is used in conjunction with carbon black in tread compounds.

In this study, the hyperviscoelastic behavior of an NR/BR tread compound (used for truck tires) reinforced by different amounts of silica and carbon black were studied with a hybrid numerical/experimental method that was developed and reported previously.<sup>16</sup> The novel aspect of this research was the examination of the applicability of two commonly used hyperelastic models (Marlow<sup>11</sup> and Yeoh<sup>9</sup>) in conjunction with two

linear and nonlinear viscoelastic models for the description of the mechanical behavior of these compounds. Furthermore, for the first time, the effect of the silica content in the compound on the viscoelastic properties via the change in the material parameters was also studied. The well-known Prony series<sup>17</sup> equations were selected for linear viscoelasticity, whereas the Bergstrom–Boyce (BB)<sup>18</sup> model was used to study the nonlinear viscoelasticity. These models are well-implemented in ABAQUS/Standard.<sup>19</sup> Therefore, this code was used to carry out the numerical calculations. It should be emphasized here that as determined by a literature survey, there are no specific published works on the combination of the Prony series and BB models with hyperelastic equations to describe the mechanical behavior of silica-reinforced tread compounds.

In the following section, the mathematical background of the selected hyperelastic models and two linear and nonlinear viscoelastic equations for rubbery materials are briefly discussed. Then, in the Experimental section, the formula and preparation techniques of the rubber compounds are given. Later, the tests carried out on the samples are presented. The finite element models developed in this work are described in the subsequent section. The results and discussion are then presented, and finally, the conclusions are given.

## MATHEMATICAL BACKGROUND

The time-independent behavior of elastomers is normally described by hyperelastic models. There have been a large number of hyperelastic models proposed thus far that are based on either a phenomenological or mechanistic viewpoint. The former models were developed by the use of direct observation of the stress versus strain curves and the proposal of a semi-empirical model. For examples, polynomial (e.g., Mooney–Rivlin, second order, etc.),<sup>4</sup> Yeoh,<sup>9</sup> and Ogden<sup>10</sup> models are a few examples of such relations. Conversely, mechanistic models are based on molecular theory to find a logical relation between the structure or configuration of polymer chains and the macroscopic behavior of the rubber. The oldest hyperelastic model, that is, the Neo–Hookean<sup>4</sup> and Arruda–Boyce<sup>20</sup> models, are categorized in this group. In addition, there are also a few models, such as the Gent equation,<sup>21</sup> that are hybrids of the phenomenological and mechanistic viewpoints. Regardless of whatever the basis of each model is, hyperelastic constitutive equations are expressed in the form of a relation between the strain energy density ( $W$ ) and invariants of the strain tensor ( $I_1$ ,  $I_2$ , and  $I_3$ ), such as in polynomials and the Yeoh and Arruda–Boyce models, or principle stretching ratios ( $\lambda_1$ ,  $\lambda_2$ , and  $\lambda_3$ ), such as in the Ogden model. Full descriptions of these models and their mathematical relations can be found in several references<sup>4,9,10</sup> and, thus, are not repeated here. In a different approach, Marlow<sup>11</sup> proposed a new method to take the hyperelasticity behavior of rubber compounds into account via an integral approach rather than giving an explicit form of  $W$  as a function of invariants or stretching ratios. It is assumed that  $W$  is only a function of  $I_1$ , which can be given as follows:

$$W = W(I_1) = \int_0^{\lambda(I_1)-1} \sigma(\varepsilon) d\varepsilon \quad (1)$$

where  $\varepsilon$  and  $\sigma$  are the nominal strain and stress, respectively, in the uniaxial tension test. These values are directly substituted in eq. (1). With the Marlow model, the number of experiments required to accurately describe the material behavior is reduced to a simple uniaxial tension test. In other words, experimental data obtained from biaxial, planar, and other complicated tests are not obligatory with this model. The experiments that we carried out showed that the Marlow model gave the most accurate results among the different hyperelastic constitutive models. However, the ABAQUS/Standard code could not accurately work with the Marlow model and nonlinear viscoelastic models simultaneously. Consequently, the Yeoh model, in which  $W$  is a function of  $I_1$ , was also selected and was expressed as follows:

$$W = C_{10}(I_1 - 3) + C_{20}(I_1 - 3)^2 + C_{30}(I_1 - 3)^3 \quad (2)$$

Where  $C_{10}$ ,  $C_{20}$  and  $C_{30}$  are the first, second and third coefficients of the Yeoh model, respectively.

To include the viscoelasticity in the material model, two linear and nonlinear equations were selected in conjunction with the aforementioned hyperelastic models (Marlow and Yeoh). The Prony series equations<sup>17</sup> were used for linear hyperviscoelasticity. The combined forms of the Prony series with the Marlow and the Yeoh models are, respectively, given as follows:

$$W^R(t) = W^0 \left[ 1 - \sum_{i=1}^N g_i \left( 1 - e^{-t/\tau_i} \right) \right] \quad (3)$$

and

$$C_{i0}^R = C_{i0}^0 \left[ 1 - \sum_{i=1}^N g_i \left( 1 - e^{-t/\tau_i} \right) \right] \quad (4)$$

where the  $W^R(t)$  and  $W^0$  are the relaxation form and initial value, respectively, of the strain energy density in the Marlow model and  $C_{i0}^R$  and  $C_{i0}^0$  are the relaxation form and initial value, respectively, of the parameters of the Yeoh model [see eq. (2)]. Also,  $g_i$  is a material parameter and  $\tau_i$  and  $N$  are the relaxation time and number of Prony series terms, respectively.

For the nonlinear viscoelasticity, the BB hysteresis model<sup>18,22,23</sup> was selected. In this model, the mechanical response of the rubber to applied loads can be attributed to two networks within the molecular structure of the material. The first one is an equilibrium network (A), which corresponds to the time-independent behavior of the rubber, and the second network (B) is responsible for the nonlinear rate-dependent part of the response. We also assumed that the total response was the sum of the responses of each network. The mechanical behavior of the first network (A) was described by the hyperelastic models, such as the Yeoh model, whereas the strain rate in network B was given by the following equation:

$$\dot{\varepsilon}_B = A[\lambda_B - 1]^c (\sigma_B)^m \quad (5)$$

where  $\dot{\varepsilon}_B$  is the effective creep strain rate,  $A$  is a material constant,  $\lambda_B - 1$  is the nominal creep strain,  $\sigma_B$  is the effective

**Table I.** Ingredients and Sample Codes of the Compounds

Ingredient	Sample code		
	NR60	NR605	NR61
NR	75	75	75
BR	25	25	25
Carbon black	60	55	50
Silica	0	5	10
Silane	0	0.4	0.8
Aromatic oil	10	10	10
Stearic acid	2	2	2
ZnO	4	4	4
Wax	2	2	2
Sulfur	1.5	1.5	1.5
CBS	0.75	0.75	0.75
TMQ	1	1	1
6PPD	1.5	1.5	1.5

Abbreviations: CBS, N-Cyclohexylbenzothiazole2-sulfenamide; TMQ, 2,2,4-trimethyl-1,2-dihydroquinoline; 6PPD, N-(1,3-dimethylbutyl)-N'-phenyl-1,4-Benzenediamine.

stress,  $m$  is a positive value greater than 1 representing the effective stress dependence of the effective creep strain rate, and  $c$  is a value between  $-1$  and  $0$  representing the dependency of the creep strain on the effective creep strain rate. To complete the BB model, a stress scaling factor ( $S$ ) is also required; it defines the ratio of the stress carried by network B to the stress carried by network A under instantaneous loading.<sup>19</sup> It should be noted that all of these values were defined over network B. It is also worth noting that the parameter  $S$  is not included in eq. (5), but it is used internally by finite element code to accurately distribute the stress between networks A and B.

## EXPERIMENTAL

Table I lists the compounding ingredients that were used in this work and their specifications and also gives the formulas and designated codes for the prepared compounds. A series of rubber compounds based on an NR/BR blend was prepared in this study. For this blend, three compounds were designed in which the total filler contents were selected to be equal to 60 phr. The first compound in the formula had 60 phr carbon black. The carbon black content was then reduced and substituted with silica filler in the consequent formulas.

To prepare rubber compounds, the ingredients were mixed in a two-roll mill (Hiwa Machinery Co., Tehran, Iran) at a friction ratio of 1 : 2 according to a standard mixing sequence. The reinforcing fillers (carbon black and silica/silane) were first added along with the process oil and paraffinic wax. Then, CBS (accelerator) and zinc oxide were added, respectively, and finally, the sulfur and antioxidant were mixed. The curing properties of the prepared compounds were measured with an MDR Rheometer 900 (Hiwa Co.) on the basis of ASTM D 5289 at a temperature of 160°C. The rubber compounds were then compression-molded into sheets with thicknesses of 2 mm at 160°C.

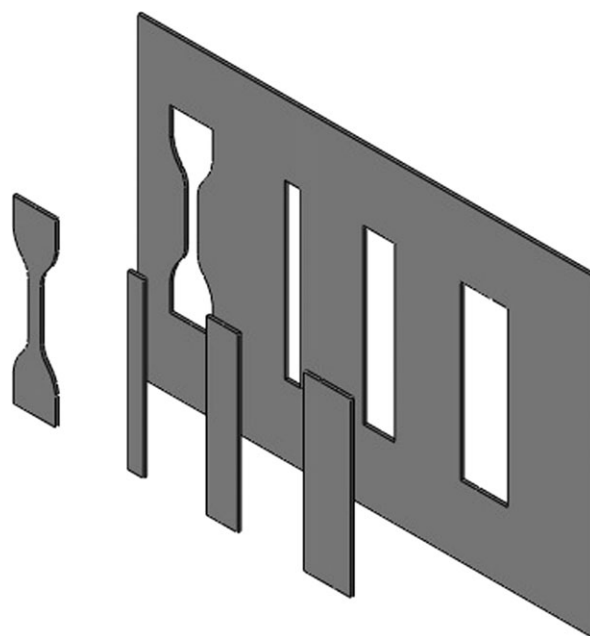
## Tests of the Specimens

To calibrate the hyperelastic material parameters, dumbbell-shaped samples were cut from vulcanized sheets, and tensile testing was carried out on the basis of ASTM D 412 with a Hiwa universal testing machine. In each test, the nominal stress and strain were measured and recorded.

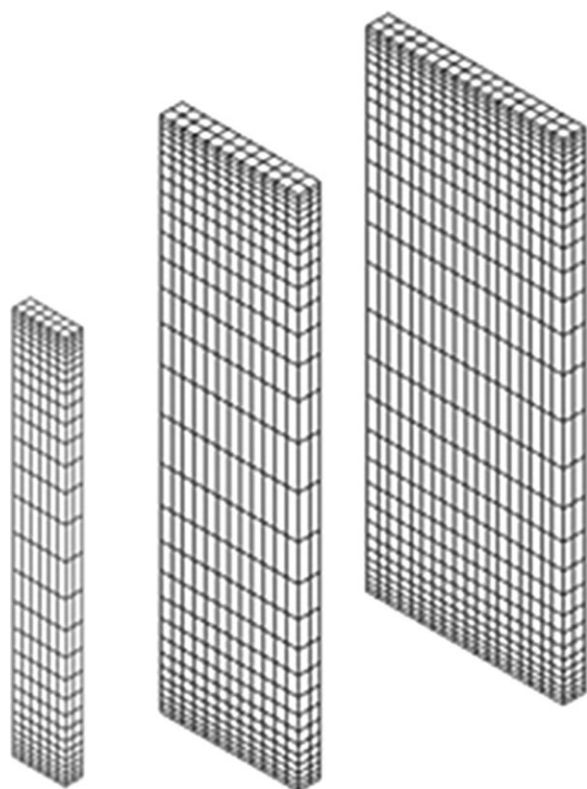
In addition to previous standard tests, tensile tests were also performed on three rubber-strip samples with a length of 11 cm and widths of 10, 20, and 30 mm, respectively, as shown in Figure 1. These tests were innovatively designed to assess the stress relaxation behavior the rubber during the tensile experiments. It should be noted that all tests were carried out on at least three samples, and the statistical calculations proved their accuracy to be 95% or more. The hysteresis and heat buildup behavior of these compounds were also evaluated with a Goodrich Flexometer according to ISO 4666.

## Finite Element Model

To properly describe the hyperviscoelasticity, the parameters of the hyperviscoelastic models had to be determined. To accomplish this task, the uniaxial tests carried out on the three rubber strips (as mentioned before) were individually modeled by the finite element method. The analyses were displacement-controlled, and the force required to achieve a 200-mm extension at a fixed rate of 500 mm/min was computed. This extension was applied on one side of the model, and the other side was completely fixed. The geometries of the three rubber strips were discretized into 360-element, 20-nodded hexahedral hybrid elements, as shown in Figure 2. The adaptive mesh technique implemented in the ABAQUS/Standard code was used in conjunction with the algorithm described in a previous work<sup>16</sup> to determine the convergent and accurate mesh.

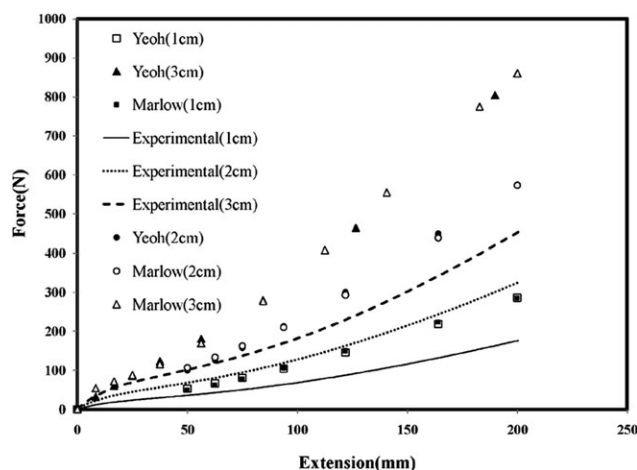


**Figure 1.** Standard sample (ASTM D 412) and three rubber-strip specimens cut from a vulcanized rubber sheet.<sup>16</sup>



**Figure 2.** Finite element meshes designed for 1-, 2-, and 3-cm samples (from left to right).

The main finite element calculations were carried out on the three rubber samples with the ABAQUS/Standard code in two steps. In the first step, hyperelastic models without time effects were considered, and thus, the analyses were carried out with a general nonlinear static assumption. In the second step, the viscoelastic material model was used. The analyses were considered to be time-dependent, in which the effects of the loading history on the material deformation were taken into account by the use of a combination of hyperelastic and Prony series



**Figure 3.** Force versus extension as predicted by the Yeoh and Marlow hyperelastic models, along with the corresponding experimental data for 1-, 2-, and 3-cm samples of NR60.

models [eqs. (11)–(13)]. The time period of each analysis was set to 24 s; this corresponded to a 200-mm extension on the basis of an extension rate of 500 mm/min (8.3333 mm/s).

## RESULTS AND DISCUSSION

### Hyperelastic Modeling

The mechanical models used in this study were implemented in ABAQUS/Standard code, which could be applied for the simulation of the rubbers under mechanical loading. The finite element calculations were carried out with both the Marlow and Yeoh equations to check the validity of the hyperelastic models without time effects. Figure 3 illustrates the variations of the experimentally measured and predicted force versus extension for the three rubber strips (1, 2, and 3 cm) prepared from the NR60 compound. There was a significant discrepancy between the experimental and model prediction, which developed at high extensions; this was due to neglecting the viscoelasticity, as reported previously.<sup>16</sup> In other words, the combined effect of the stress relaxation and creep phenomena generated severe deviations from the experimental data in the calculated results. Figure 3 also shows that both the Marlow and Yeoh models predicted almost identical results. This was due to the polynomial form of the Yeoh model, in which  $W$  is only a function of  $I_1$  [eq. (2)]. The Marlow model also assumed that  $W$  is a function of  $I_1$ . It could be concluded, therefore, that behaviors of these models, at least in tension mode, were similar.

### Linear Hyperviscoelastic Modeling

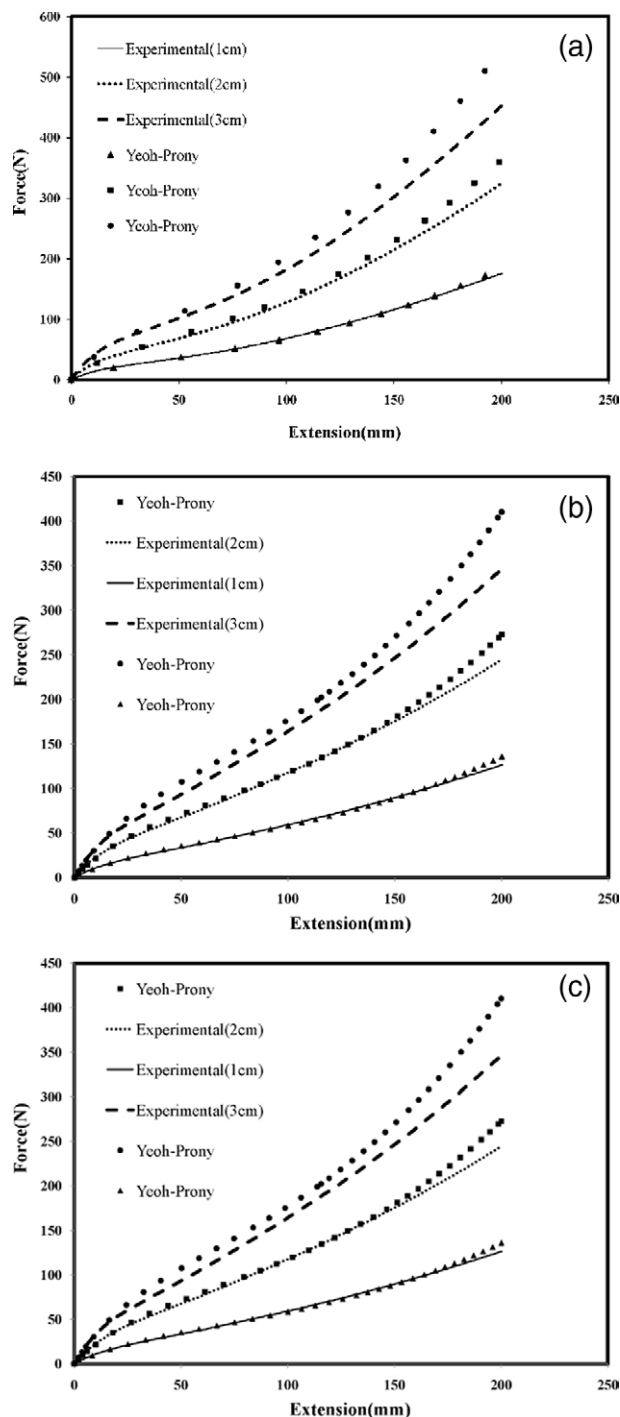
To overcome this problem, the time effect or viscoelasticity had to be taken into consideration. The analyses were carried out with the Marlow and Yeoh hyperelastic models and a linear viscoelastic model, in which the changes in material parameters were considered with the Prony series model given by eqs. (3) and (4). To determine the parameters of the Prony series (i.e.,  $g_i$  and  $\tau_i$ ), the initial values were assumed for these variables, and the analysis was performed for the 1-cm model. The predicted force was then checked against the experimentally recorded forces, and the error was determined as follows:

$$\text{Error} = \sum_{i=1}^n \left( 1 - \frac{F_i^{\text{Calc}}}{F_i^{\text{Exp}}} \right)^2 \quad (6)$$

where  $F_i^{\text{Exp}}$  and  $F_i^{\text{Calc}}$  are the experimentally measured and calculated forces, respectively. With the optimization technique described in a previous work,<sup>16</sup> the updated values of the parameters were determined, and analysis was repeated consecutively until the calculated error became very small ( $<0.001$ ). Table II gives the parameters of the Prony series model for the compounds. Moreover, to verify the accuracy of the computed

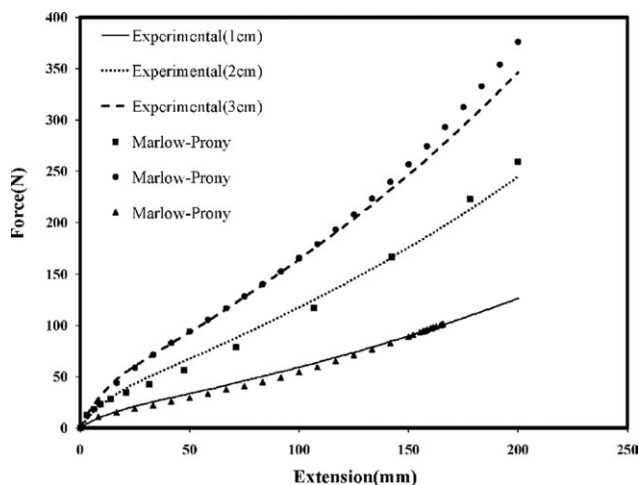
**Table II.** Parameters of the Prony Series for Each Compound

Sample code	$g_i$	$\tau_i$
NR60	0.5	2.5
NR605	0.4	2.5
NR61	0.38	4.5



**Figure 4.** Force versus extension as predicted by the Yeoh and Prony hyperviscoelastic models, along with the corresponding experimental data: (a) NR60, (b) NR605, and (c) NR61.

values of  $g_i$  and  $\tau_i$ , the analyses were also carried out for the 2- and 3-cm samples. Figure 4(a–c) shows the variations of the predicted and experimentally measured force versus extension of the three rubber samples for compounds NR60, NR605, and NR61, respectively. The hyperelastic model in the results shown in these figures was the Yeoh model. The results show that these values were accurate, and the method was, thus, applicable.



**Figure 5.** Force versus extension as predicted by the Marlow and Prony hyperviscoelastic model, along with the corresponding experimental data for NR60.

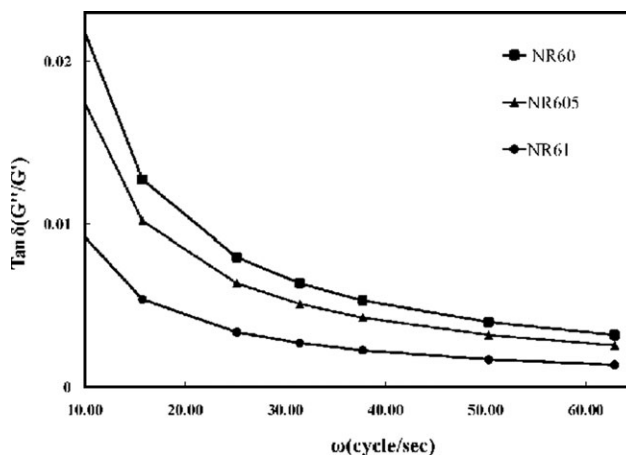
Similar to hyperelastic modeling results presented in previous section, the Marlow model also showed the same behavior as the Yeoh model because both models consider  $I_1$  to describe the  $W$  function. Figure 5 shows the force versus extension for the NR60 compound with Marlow and Prony series equations.

To verify the method and obtained results in a more applied sense, the parameters of the Prony series equations were used in conjunction with following relations<sup>17</sup> to compute the storage modulus ( $G'$ ) and loss modulus ( $G''$ ):

$$G'(\omega) = G_0 \left[ 1 - \sum_{i=1}^N g_i \right] + G_0 \sum_{i=1}^N \frac{g_i \tau_i^2 \omega^2}{1 + \tau_i^2 \omega^2} \quad (7)$$

$$G''(\omega) = G_0 \sum_{i=1}^N \frac{g_i \tau_i^2 \omega}{1 + \tau_i^2 \omega^2} \quad (8)$$

Where  $\omega$  and  $G_0$  are the frequency and an instantaneous shear modulus, respectively. The loss factor ( $\tan \delta$ ), calculated as  $G''(\omega)/G'(\omega)$ , is shown in Figure 6. On the other hand, the



**Figure 6.** Predicted  $\tan \delta$  values for three compounds.

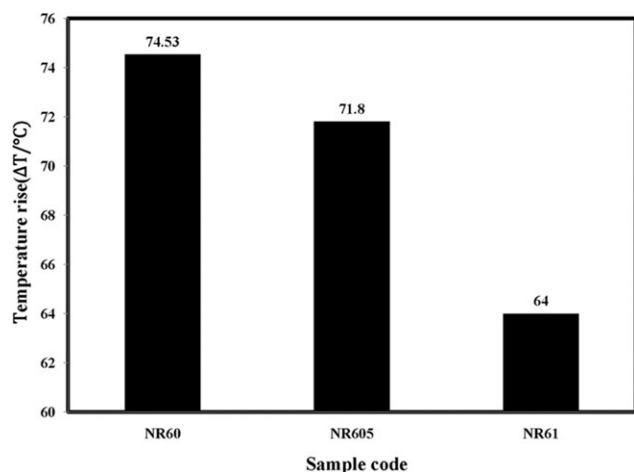


Figure 7.  $\Delta T$  as measured by heat buildup tests.

results of the heat buildup experiments, which were increases in the temperature of the sample, are shown in Figure 7. These tests were carried out at an approximate  $\omega$  of 60 cycle/s. A comparison of the temperature rise ( $\Delta T$ ) of the compounds shown in Figure 7 with the  $\tan \delta$  values at  $\omega \approx 60$ , illustrated in Figure 6, revealed that the higher the values of  $\tan \delta$  were, the higher the  $\Delta T$  value in the heat buildup experiments was obtained. As expected, the compound NR60, which had no silica in its formula, had the maximum  $\tan \delta$  and  $\Delta T$  values. When the carbon black in the formulations was substituted with silica, the  $\tan \delta$  and  $\Delta T$  decreased so that compound NR61 had the lowest values of both  $\tan \delta$  and  $\Delta T$ . We concluded that accurate determination of the Prony series parameters took advantage of heat buildup prediction and was experimentally well worth it.

### Nonlinear Hyperviscoelastic Modeling

As it can be seen in Figure 4(a–c), deviation between experimental data and calculated results using linear hyperviscoelastic model (i.e., Yeoh or Marlow with Prony series equations) becomes more prominent at higher extension. It is even more significant when the width of the samples increases. The main reason that can describe the failure of the combined hyperelastic and Prony series equations is the nonlinearity in material behavior at higher deformation. Furthermore, increasing of the sample width (moving from 1 cm to 2 cm and 3 cm) will change the state of the stress and strain from simple one-dimensional to more complicated two-dimensional. Therefore, the nonlinear viscoelastic model proposed by BB (see eq. (5)) was used to take the nonlinear viscoelasticity into account. In

Table III. BB Parameters for Each Compound

Sample code	S	m	A	c
NR60	0.95	6	0.185	-1
NR605	1.1	5	0.32	-1
NR61	0.5	4	0.555	-1

other words, combination of a hyperelastic model (e.g., Marlow or Yeoh) with Prony series equation only considers the nonlinearity in time-independent part of the simulation. However, in this approach both time-independent (hyperelastic) and dependent (viscoelastic) parts were considered simultaneously. A similar optimization approach used for the determination of the parameters of the Prony series was also used and the parameters of the BB model were obtained and recorded in Table III. Figure 8(a–c) shows the variations of the experimentally measured and

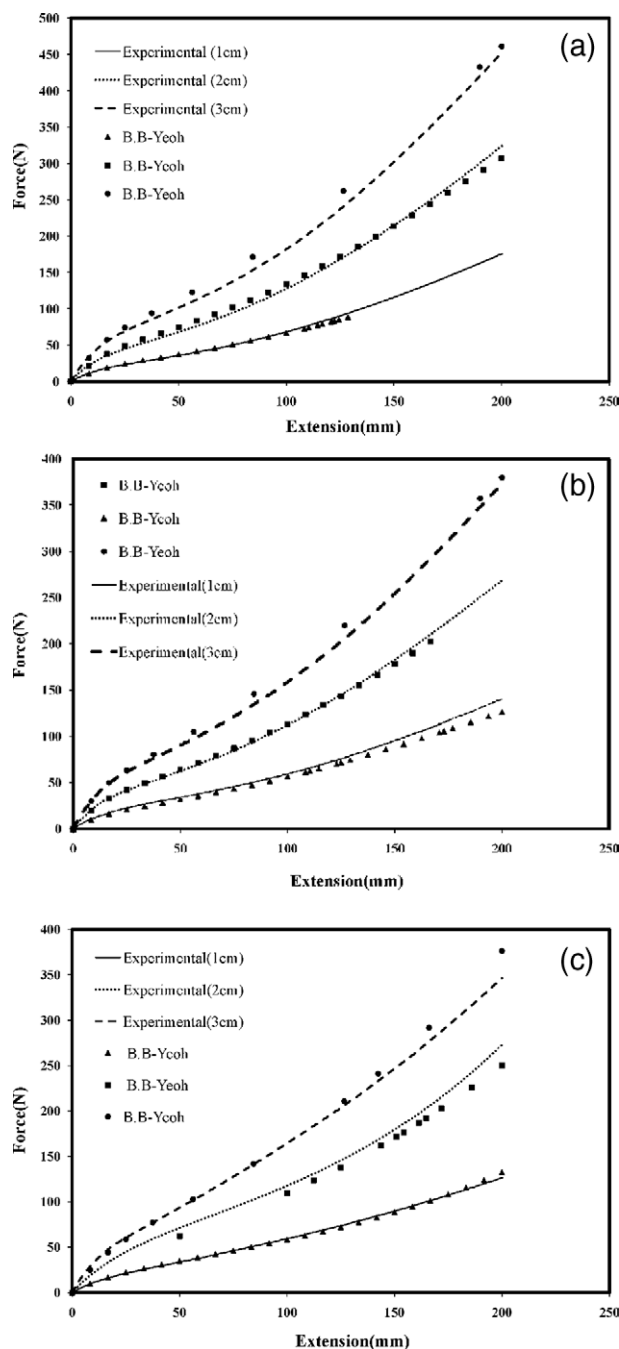


Figure 8. Force versus extension as predicted by the Yeoh and BB hyperviscoelastic models, along with the corresponding experimental data: (a) NR60, (b) NR605, and (c) NR61.

calculated forces of the three samples for compounds NR60, NR605, and NR61, respectively. Very good agreements were seen between the experiments and simulations with the combined hyperelastic and material models.

The classical background or theory of the BB model implies that an increase in  $S$  [see eq. (5)] would increase the dissipation of energy, and thus, a larger value for  $\Delta T$  would be obtained. On the other hand, a very relatively large or a very small value of  $A$  means that the compound would show a low hysteresis behavior. It is quite well-known that the addition of silica into carbon-black-filled rubber compounds imposes a dual role on the elastomer matrix. First, the hysteresis or energy dissipation during cyclic loading decreases, and second, the mechanical properties, such as the tensile properties, decrease; this is attributed to the large particles size of silica and weak interaction between the filler and polymer matrix.<sup>24,25</sup> Consequently, as shown in Table III, the incorporation of silica into the compound changed all of the parameters ( $S$ ,  $A$ ,  $c$ , and  $m$ ) in the BB, and a simple interpretation of the model parameters could not be accomplished. However, the model was quite capable of predicting the mechanical behavior of the compounds at both low and high strain values.

## CONCLUSIONS

With a previously developed methodology, the hyperviscoelasticity behavior of an NR/BR blend developed for the tread compound of truck tires was studied. Two linear (Prony series) and nonlinear (BB) viscoelastic models were combined with two available hyperelastic models (Marlow and Yeoh). It was shown that neglecting the viscoelasticity gave rise to large errors in the prediction of the mechanical deformation in the rubbers. However, the linear viscoelastic model failed to predict the correct behavior at large strain, especially for wider samples. On the other hand, not only did the combination of the Yeoh hyperelastic model with the BB equation describe the mechanical behavior at low to medium strains, but also, the large strains were taken into consideration. Both the linear and nonlinear hyperviscoelastic models accurately described the hysteresis in rubbers and could be used for the evaluation of the rolling resistance in tires.

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