NONLINEAR VISCO-HYPERELASTIC CONSTITUTIVE MODELING FOR FILLED ELASTOMERIC MATERIALS

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ABSTRACT

The mechanical behavior of filled elastomeric materials (rubber or rubber-like materials) is known to be incompressible, or nearly-incompressible, hyperelastic and time-dependent, or viscoelastic. This complex behavior of rubbery materials needs more understanding, and a good knowledge is required for such behavior in order to attain a constitutive modeling for better design of a rubber component for a specific application. To achieve this objective, theoretical and experimental works are presented in this paper.

Theoretical works are considered for modeling the hyperelastic and viscoelastic behaviors of rubber. The hyperelastic behavior is modeled using Mooney–Rivlin constitutive model. While the time-dependent behavior (viscoelasticity) was modeled by using Prony series. Modeling and parameters identification, for both hyperelastic and viscoelastic behaviors, were performed and compared with ANSYS 14. To do this, different tests were performed on filled rubber in the present work, all tests were performed on filled rubber material with three different kinds of carbon black, N326, N375, and N660 at room temperature.

Tensile stress-stretch curves were generated from the test data at strain rates 10 mm/min. Relaxation stress-time curves were generated from the test data at mean strain (200%) from the effective length of the specimen, at constant strain rate (200 mm/min).

From the work it is found that a two-term Mooney-Riviln adequately describes the hyperelasticity of the material. The numerical results, using ANSYS, exhibit good agreement with experimental data.

KEY WORDS: **Elastomer, rubber, rubber-like, constitutive model, carbon black, finite element.**

النمذجة اللاخطية المفرطة المرونة - اللزجة للمواد المطاطية المدعمة

الخلاصة

التصرف الميكانيكي للمواد المطاطية (المطاط والمواد الشبيهة به) المدعمة يعرف على انه غير قابل للانضغاط , او قريب من ذلك , مفرط المرونة , يعتمد على الوقت , او مرن لزج . هذا التصرف المعقد للمواد المطاطية يحتاج فهم اكثر , والمعرفة

الجيدة لهذا التصرف يمكن من الحصول على نمذجة لتصميم افضل تركيبة مطاط لتطبيق معين . وللوصول الى هذا الهدف , العديد من الاعمال النظرية والعملية قدمت في هذا البحث .

الاعمال النظرية شملت نمذجة التصرف المفرط المرونة والتصرف المرن اللزج للمطاط . تم نمذجة تصرف المرونة المفرطة باستخدام نموذج موني-ريفلن . بينما التصرف المرن اللزج نمذج باستخدام متسلسلة بروني . النمذجة ومعاملاتها التعريفية لكلا التصرفين المفرط المرونة والمرن اللزج تم عمله ومقارنته مع الانسز 14 . ولعمل ذلك , العديد من الاختبارات اجريت على المطاط المدعم في هذا العمل , جميع الاختبارات اجريت على مادة المطاط المدعم بثلاث انواع مختلفة من اسود الكاربون (N660,N375,N326) بدرجة حرارة الغرفة .

منحنيات الاجهاد – الاستطالة انشأت من البيانات التي تم الحصول عليها من اختبار الشد عند معدل انفعال 10mm⁄min.

منحنيات اختبار الاستراحة (الاجهاد – الزمن) انشأت من البيانات التي تم الحصول عليها عند مستوى انفعال (200%) من الطول الفعال للعينة , عند معدل انفعال ثابت (200 mm⁄min) .

من العمل وجد انه موديل موني-ريفلن ذو الثابتين تكون كافية لوصف المرونة العالية للمواد المطاطية. النتائج الحسابية باستخدام برنامج الانسز يظهر نتائج مطابقة مع النتائج العملية.

1. INTRODUCTION

Elastomers involve natural and synthetic rubbers, which are amorphous and are comprised of long molecular chains. Chains are highly twisted, coiled, and randomly oriented in an undeformed state. In applying load, these chains become partially straightened and untwisted; when load is removed the chains revert back to its original configuration. Their stress-strain relationship can be highly nonlinear.

The accurate modeling of this phenomenon is a key issue for a better understanding of the mechanical behavior of rubber. Most of the starting point for modeling of various kinds of elastomers is a strain energy function. The properties of a material are described by a constitutive model. Generally, this is a mathematical relation between the stress and the strain. As the stress, in some materials, is dependent on other factors rather than the strain, like strain rate, magnitude of strain, temperature, plasticity and strain amplitude and frequency in a case of cyclic loading, so rubber is a material which is dependent on most of the mentioned factors. Therefor; there are different kinds of constitutive models, which can be used to model rubber, have been developed.

Constitutive model is mathematical relation between the stress and the strain to find the material parameters. There are different kinds of constitutive models which can be used to model a rubber. The first successful model was due to Kuhn in (1936) [1], who derived a relation between the elastic modulus and the molecular weight of the chains. Flory and Rehner [2] proposed a four chain regular tetrahedron model.

The earliest work of large elastic deformation is due to Mooney theory [3], which, then developed by Rivlin and called Mooney-Rivlin model. Rivlin [4] showed that Mooney-Rivlin model, the earlier result of Mooney [3], can actually put in most general form by putting the strain energy function in terms of I_1 and I_2(the strain invariants).

An important development was introduced by Rivlin and Saunders [5], who adopted the more logical procedure of choosing the conjugate values of λ_1 and λ_2 (principle stretch)in the biaxial strain experiment in such a way that in any given test one of the two strain invariants I_1 and I_2 was held constant while the other was varied.

A number of scientists, who examined the reinforcement phenomenon (Alexandrov and Lazurkin [6], Dannenberg [7], Rigbi [8], Medalia [9], Edwards [10], Kilian et al. [11], Leblanc

[12], Kaliske and Rothert [13], attributed it to the surface mobility and dragged slippage of adsorbed segments of elastomer chains over the surface of filler particles, assuming that this process prevented molecules from premature breaking and thus increased the resistance of material to extension.

Another phenomenon was observed in elastomer behavior when it is subjected to cyclic loading which is characterized by an important loss of stiffness or a stress softening during the first few cycles. Bouasse and Carriere in (1903) first found this phenomenon in a test for a rubber vulcanized [14]. As a consequence of a more extensive experimental investigation by Mullins in (1947), the stress softening effect is now widely known as the Mullins effect. Lion [15], Septanika [16], Miehe and Keck [17], Drozdov and Dorfmann [18] and Besdo and Ihlemann [19] made their contributions to this field.

The main objective of the present paper is, first, considering hyperelastic constitutive modeling for filled rubber using Mooney–Rivlin, followed by examination of a viscoelastic constitutive model using Prony series. The two are then combined to yield a visco-hyperelastic constitutive relationship for rubber materials loaded at different strain rates. The modeling and parameters identification will be implemented in ANSYS 14.

2. EXPERIMENTAL WORK

2.1 Materials and Sample Preparation

The materials and samples preparing processes were done in Babylon Tire Factory laboratories. The gum material and additives were performed using the calendering machine, the mixing process continues till reaching a homogenous blend. The sulfur and accelerators are added at the end of the mixing process to avoid curing during calendaring processes. Electrical piston was used to cure the blend, when the piston temperature reach (145C°) the (70 gr) from the blend, **(Table 1)**, pressed by mold for (45 min) to produce a thin sheet of rubber which can be used later to make the dumbbell specimen. **(Table 1)** shows the blend with carbon black N375, the two other recipes are the same but with carbon black N326 and N660.

Dumbbell specimens were manufactured under ASTM D412 specifications for tensile test as shown in **(Figure 1)**.

2.2 Tensile Tests

These tests have been carried out by using the instrument showing in **(Figure 2)** type Monsanto Tensometor 10. To starting the test the instrument must be fed by input data, thickness, width and the strain rate (10 mm/min). The samples stretched to 300% from the original length which means (λ=3). The experiment has been repeated for all three blends.

The recorded values were used later to draw the stress-stretch curve, as shown in **(Figure 3)**.

2.3 Stress Relaxation Test

Rubbers are classified as viscoelastic materials, viscoelastic materials appear both elastic solid and a viscous fluid response when deformed. The main important method to study and compare the viscoelastic compounds properties is stress relaxation experiment method. Stress relaxation can be defined as continued decreasing in stress needed to maintain a given deformation or loss of stiffness with time. This test has been carried out by using Monsanto Tensometor 10 instrument showed in **(Figure 2)** which the same instrument used in tensile test before. Dumbbell specimen used in the relaxation test is shown in **(Figure 1)**.

The procedure started by holding both ends of the sample via clamps of the instrument. As in tensile test the effective length of the sample is the distance between the two holders which fixed at length (25 mm), then fed the instrument by input data which like, thickness, width of the sample and strain rate. Strain rate was fixed at 200mm/min for all experiment duration. After completing all those steps, the tensile stress was applied and continued till the deformation reached 200% and then stopped. The decreasing in stress was recorded by using video camera. The period of recording the data were continued for 5 minutes and the values taken were used for drawing the relationship between time and stress. Same steps were repeated for each blend used in this work.

(Figure 4) show the relationship between stress and time for different blends at 200% deformation which represent the relaxation phenomena.

3. CONSTITUTIVE MODEL

The properties of a material are described by a constitutive model. Generally, this is a mathematical relation between the stress and the strain. As the stress, in some materials, is dependent on other factors rather than the strain, like strain rate, magnitude of strain, temperature, plasticity and strain amplitude and frequency in a case of cyclic loading, so rubber is a material which is dependent on most of the mentioned factors. Therefore; there are different kinds of constitutive models, which can be used to model rubber, have been developed.

The purpose of the constitutive theories is to develop mathematical models for representing the actual behavior of matter. Past researches developed two approaches to obtain the strain energy functions in rubbery materials, or generally, elastomers. The first approach is based on the statistical thermodynamic, where the microscopic molecular structure of the material is taken into account. The second is a phenomenological one, which treats the material as a continuum.

3.1 Mooney-Rivlin Constitutive Model

The big challenge in non-linear elasticity theory is to come up with a reasonable and applicable elastic law (strain energy function), which is the crucial step to the development of dependable analysis tools. Despite of many attempts have been made to develop a theoretical stress-strain relation that can match the experimental results for hyperelastic materials, Mooney's theory was the most significant phenomenological theory of large elastic deformations, which has played a principal part in all later work in the field.

The compressible form of Mooney-Rivlin material model is [5]

$$W = c_{10}(\bar{I}_1 - 3) + c_{01}(\bar{I}_2 - 3) + \frac{1}{2}K(J-1)^2 \qquad (1)$$

where the third term is a compressible part, and J is the determinant of deformation gradient (or volume ratio). For incompressible material, $J = 1$ for that Mooney-Rivlin equation can be written as.

$$W = c_{10}(I_1 - 3) + c_{01}(I_2 - 3) \qquad (2)$$

where W is the strain energy function, $I_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2$, $I_2 = \lambda_1^2\lambda_2^2 + \lambda_2^2\lambda_3^2 + \lambda_3^2\lambda_1^2$, c_{01} and c_{10} are material constants, and λ_1, λ_2 and λ_3 are the principle stretches.

In the tensile test (the case of uniaxial tension), the change in strain energy can be expressed in variational form as:

$$dW = \left(\frac{\partial W}{\partial \lambda_1}\right) d\lambda_1 \quad (3)$$

which leads to the following form of Mooney-Rivlin constitutive relation:

$$\sigma_o = 2\left(\lambda^2 - \frac{1}{\lambda}\right)\left[c_{10} + \frac{c_{01}}{\lambda}\right] \quad (4)$$

where σ_o represents the engineering stress. In this case the cross sectional area changes with the deformation and can be expressed as [20]:

$$A = \frac{A_0}{\lambda} \quad (5)$$

By substitute equation (4) in equation (5), Cauchy true stresses obtained as:

$$\sigma = 2c_{10}\left(\lambda - \frac{1}{\lambda^2}\right) + 2c_{01}\left(1 - \frac{1}{\lambda^2}\right) \quad (6)$$

Equation (6) can be solved by using the least squares (linear regression) approach, the constitutive model coefficient obtained as:

$$Y = c_{10} + c_{01}X \quad (7)$$

where $Y = \frac{\sigma}{2(\lambda - \lambda^{-2})}$ and $X = \frac{1}{\lambda}$.

Solving the above equation to find the coefficient as:

$$\begin{aligned} c_{10}n + c_{01}\sum X &= \sum Y \\ c_{10}\sum X + c_{01}\sum X^2 &= \sum XY \end{aligned} \quad (8)$$

3.2 Viscoelastic Model

One of the basic rheological viscoelastic models which can anticipate relaxation behavior was proposed by James Clark Maxwell, Maxwell model, which consists of viscous Newtonian damper and elastic Hookian spring in series. The total strain would be equal to the summation of the strain in elastic and viscous elements, because they are in series. In relaxation test when the displacement applying instantaneously, the viscous part needs some time to move, while the spring could move instantaneously. The whole displacement will be compensated by the spring at the time zero. As the time goes on, the displacement in the spring will decrease, but increase in the damper. For infinity time, the strain in the elastic part would be zero, but this is a problem in modeling complicated elastomers, that is stress in these materials even in the long time would not lead to zero. Thus, a general Maxwell model for modeling relaxation behavior is needed.

When the material is assumed to be a general Maxwell solid, the relaxation function is typically modeled with a Prony series. Values of the shear and bulk modulus would be enough as the starting values of the material properties over the time, which are representative of deviatoric and volumetric parts of the stress, respectively, as expressed in the following equations [21]:

$$\sigma = \sigma_{deviatoric} + \sigma_{volumetric}\,, \quad (9)$$

$$\sigma = \int_0^t 2G(t-\tau)\frac{de}{d\tau}d\tau + \boldsymbol{I}\int_0^t K(t-\tau)\frac{d\Delta}{d\tau}d\tau\,, \quad (10)$$

where σ is Cauchy stress, e and Δ are deviatoric parts of the strains, G(t) and K(t) are shear and bulk modulus functions, respectively, t and τ are current and past time, respectively, and $\boldsymbol{I}$ is identity matrix.

Relating to the shear and bulk modulus over the time, Prony series can be proposed by the flowing formulas [21]:

$$G = G_0\left[\alpha_\infty^G + \sum_{i=1}^{n_G} \alpha_i^G \, exp\left(-\frac{t}{\tau_i^G}\right)\right] \quad (11)$$

$$K = K_0\left[\alpha_\infty^K + \sum_{i=1}^{n_K} \alpha_i^K \, exp\left(-\frac{t}{\tau_i^K}\right)\right] \quad (12)$$

where superscript shows belonging to shear or bulk modulus, and subscript indices the number of series component, $\alpha_i = \frac{G_i}{G_0}$ and τ_i are relaxation time constants for each Prony series component. α_i that can be calculated at *t* equal to zero will be obtained from equation (11), by writing the equation as

$$G_0 = G_0\left[\alpha_\infty^G + \sum_{i=1}^{n_G} \alpha_i^G\right] \quad (13)$$

it means that $\alpha_\infty = 1 - \sum_{i=1}^{n} \alpha_i$, so equation (11) can be written as :

$$1 = \alpha_\infty + \sum_{i=1}^{n} \alpha_i,$$

There are only two constants α_i and τ_i which should be determined by a relaxation test. In the series, the initial values of *G* and *K* would be taken into account at time equal to zero. For finding the coefficient of the bulk function, the same above procedure will be followed.

4. RESULTS AND DISCUSSION

4.1 Fitting Hyperelastic Material Parameters in ANSYS

In this research, stress-strain data obtained from the tensile test were used to determine the material parameters in hyperelastic models which in turn employed in the commercial FEA software ANSYS 14.0 to perform structural simulations of rubber components submitted to quasi-static loading under hyperelastic deformations.

There are some key assumptions related to the hyperelastic constitutive models in ANSYS, deformations are fully recoverable, thermal expansion is isotropic, materials nearly or fully incompressible and the constitutive hyperelastic models are defined through a strain energy density function.

ANSYS provides curve fitting tools to obtain material constants for hyperelastic models from the results obtained during the test. These results are fed into the ANSYS software in the form of a text file for defined stress-strain of the manipulated testing data for uniaxial tension.

Immediately after the data given to the ANSYS, the fitting process starts by choosing the Mooney-Rivlin strain energy function with two parameters. Based on above procedure, ANSYS analyzes the data, and the materials constants C_{10} and C_{01} become known under quasi-static strain rate (10 mm/min). These constants for the three blends used in the present work are presented **(Table 2)**.

(Figure 5) shows the experimental stresses vs. strains for the three rubber blends used in the current work and compared with that obtained from ANSYS 14 using the above mentioned fitting procedure for Mooney-Rivlin constitutive model. The results were very encouraging, since the ANSYS model was accurate for the approximately 75% range of deflections.

(Tables 3) summarizes the accuracy achieved in the FEA model of the dumbbell specimen of tensile test experiments with the adjusted Mooney-Rivlin function. The predicted stress at different strains and at strain rate 10 mm/min is relatively well compared to test data. The most minimum strain rate considered as equilibriums state in this case, so it is the minimum error ratio.

4.2 Fitting Relaxation Tests of Prony Series using ANSYS

The viscoelastic behavior for elastomer in consideration was studied under stress relaxation test by using stress vs. time curve obtained from the test, the curve which considered being as a milestone for further steps.

The shear modulus, *G*, may be calculated experimentally using the following equation [21]:

$$G = \frac{E}{2(1+v)} \tag{14}$$

The instantaneous modulus of elasticity, *E*, in the above equation, can be calculated by dividing the stress over the strain at a specific time *t*. The repetitive calculation is facilitated by the use of MS Excel. The final results of the shear modulus and time values obtained from above procedure were saved as a text file.

In ANSYS by having shear modulus over time, optimal parameters fitting of Prony series for shear could be found. The implemented materials were two components of the Prony series for modeling hyperelastic materials, **(Table 4)** shows the constants Prony obtained from ANSYS. The curve for shear modulus vs. time are plotted for the relaxation test conducted in the lab and compared with the results of the ANSYS model for three blends, as shown in **(Figure 6)**.

4.3 Rubber Sheet with Central Hole

To validate the current analysis a cyclic tension was applied on rubber sheet with a central hole, as shown in **(Figure 7a)**. The sheet manufactured from blend with carbon black N326, its dimensions are: 15cm long, 7.5cm wide, and ~0.25cm thick, the hole at the center is 2.6cm diameter. Two of clamps were made from steel plate fixed by three screws for each end. Purposes of using these clamps are to distribute the force evenly on the both ends of the specimen and to make it easy to be fixed between the two jaws of the computerized test machine, Testometric AX M500-25kN, as shown in **(Figure 7b)**. The test was carried out under strain rate of 100mm/min and deformation 100% with two numbers of cycles. The results of the test are plotted in **(Figure 8)**.

It is seen from **(Figure 8)** that rubber material exhibit hysteresis during cyclic loading, this indicates that the material has a significant amount of viscous behavior, and as the viscoelastic response of the materials increases the amount of hysteresis will increases. That is related to its time dependent characterizes behavior and for the same reason noticed that the uploading (increase the load) condition seemed stiffer than the unloading (decreasing the load) condition, this is an interesting behaviour and may the material relaxed during the unloading condition. Also, there is a significant difference between the response during the cycle one and cycle two during uploading, see **(Figure 8)**, while, in the unloading condition the response is less sensitive during the first two cycles. This feature significant clearly in filled rubber as used in this research, to broken the bonds between filled materials (carbon black) in the first cycle for the same strain rate.

This problem is solved by ANSYS, the model meshed with element type hyperelastic 8 nodes 183. The boundary conditions were applied as follows. The bottom edge of the model was fixed, while the upper edge is given a displacement values that makes the strain, first, 50%, and then 100% of strains. **(Figure 9)** showed the Von Misses stress contours with the exact deformed mesh at these strains. Observe that the inhomogeneous deformation is concentrated in the neighborhood of the hole, this localization gives a stress concentration at the sides of the hole, which is as expected in metallic solid materials, and the only difference here is that due to high deformation characteristics of the rubbery materials the stress concentration factor is lower.

5. CONCLUSION

1- By employing a fundamental approach to the formulation of constitutive relationships, a Mooney-Rivlin constitutive model with Prony series are anticipated to describe visco-hyperelastic large deformation behaviour of incompressible rubber-like materials under different strain rates.
2- It is found that a two-term Mooney-Rivlin adequately describes the hyperelasticity of the material. Another component in the equation, a generalised Maxwell model, is introduced to characterise viscoelastic response under these strain rates.
3- The total expression corresponds to a hyperelastic solid in parallel with a generalized Maxwell model, thus characterising not only hyperelasticity but also strain rate and strain history dependent viscoelasticity.
4- Stress vs. strain curves predicted by the model for three kinds of rubber blends are compared with the experimental data performed in the current research. The comparisons showed that the proposed model is well-suited for the description of visco-hyperelastic behaviour of rubber-like materials loaded at different strain rates.
5- The numerical results, using ANSYS, exhibit good agreement with experimental data, demonstrating that the model is suitable for prediction of visco-hyperelastic behaviour in situations different from that used to determine its parameters.

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Table 1: The recipe of the blend used in tread of farm tire

	Quantity in gm	*PHR**
SBR 1502(Strene Butadien Rubber)	**85.5**	**100**
SBR 1712(Strene Butadien Rubber)	**142.5**	
BR cis(Polybutadiene)	**56.4**	
ZnO(Zinc Oxide)	**7.2**	**2.53**
Stearic acid	**4.2**	**1.47**
6PPD(protection product)	**5.7**	**2**
TMQ	**3**	**1.05**
Wax	**5.7**	**2**
Paraffin oil	**56.7**	**19.93**
Sulfur	**4.5**	**1.58**
MBS(Morpholinothiobenzothiazole accelerators)	**3**	**1.05**
Carbon Black N375	**183.6**	**64.55**
Reclaimed Rubber	**42**	**14.76**

***** Parts per Hundred Rubbers

Table 2: Materials parameters obtained using ANSYS 14.

Blend with different carbon blacks	Mooney-Rivlin Coefficients	
	C_{10}	C_{01}
N326	0.598750473900	-0.969581769171
N375	0.797369060278	-1.384107044970
N660	0.783195721870	-1.421800766730

Table 3: Comparison of stresses at different strains between the experimental test and ANSYS.

	Strain %	Stresses at specific strains (MPa)		Error %
		Experimental	Predicted (ANSYS)	
Blend with N326	100	1.530	1.567	2.410
	200	3.140	2.800	10.82
Blend with N375	100	1.809	1.859	2.760
	200	4.028	3.530	12.36
Blend with N660	100	1.650	1.712	3.750
	200	4.000	3.370	15.75

Table 4: Prony series constants for three blends obtained from ANSYS.

Prony Series constants	Kind of blend		
	Blend with carbon black N326	Blend with carbon black N375	Blend with carbon black N660
α_0^G	0.907663954342	1.049103209860	1.127357967550
α_1^G	0.339863017171	0.445172377449	0.437216238454
α_2^G	0.354166922530	0.463290662996	0.405809181564
τ_1	96.15047225200	0.009380944487	46.29573533270
τ_2	13.31736723090	56.54316636330	0.286361264090

Figure 1: Dumbbell type rubber specimens.

Figure 2: Tensile test instrument, Monsanto Tensometer 10

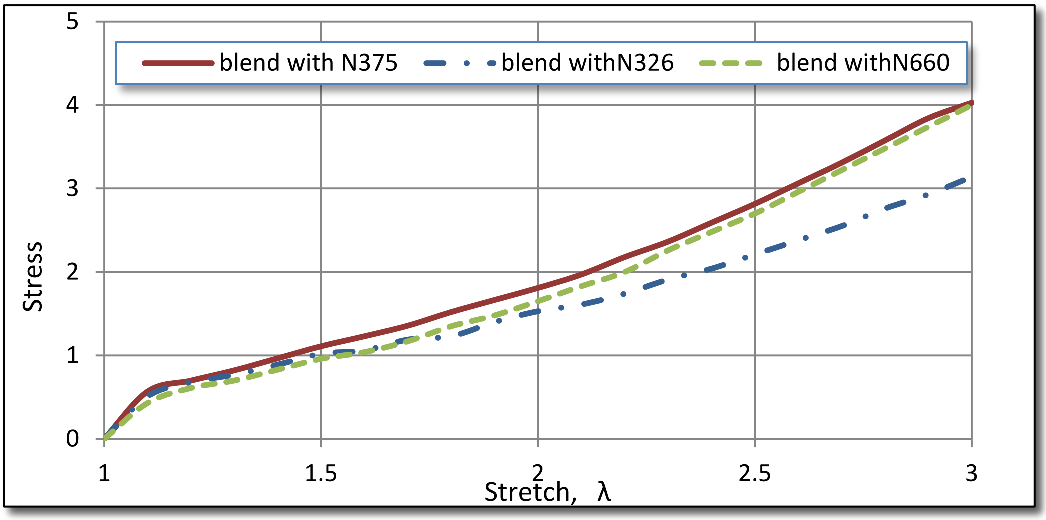


Figure 3: Stress-strain relationship for three blends at strain rate 10 mm/min

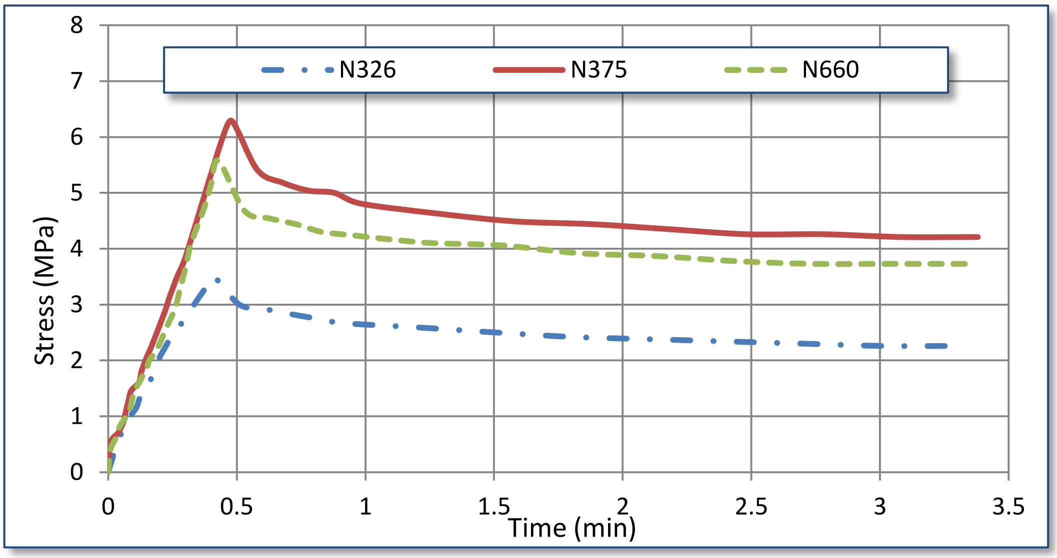


Figure 4: Stress relaxation for three blends at deformation 200%

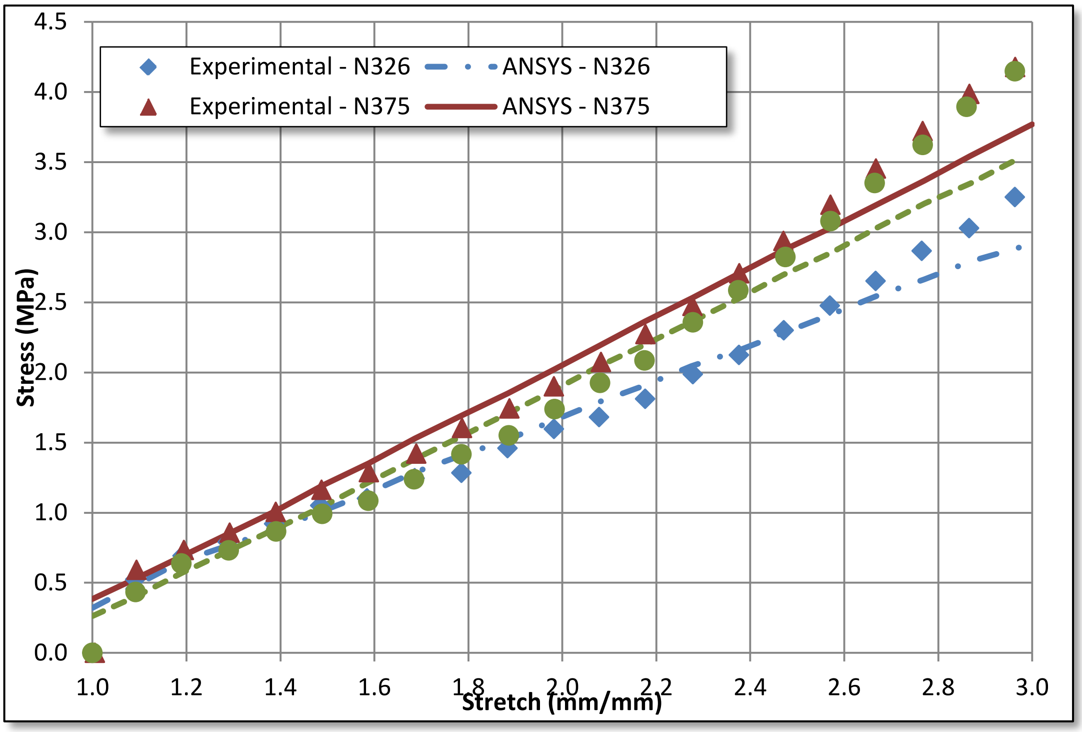


Figure 5: Experimental stresses-strains plots for three blends used as compared with ANSYS.

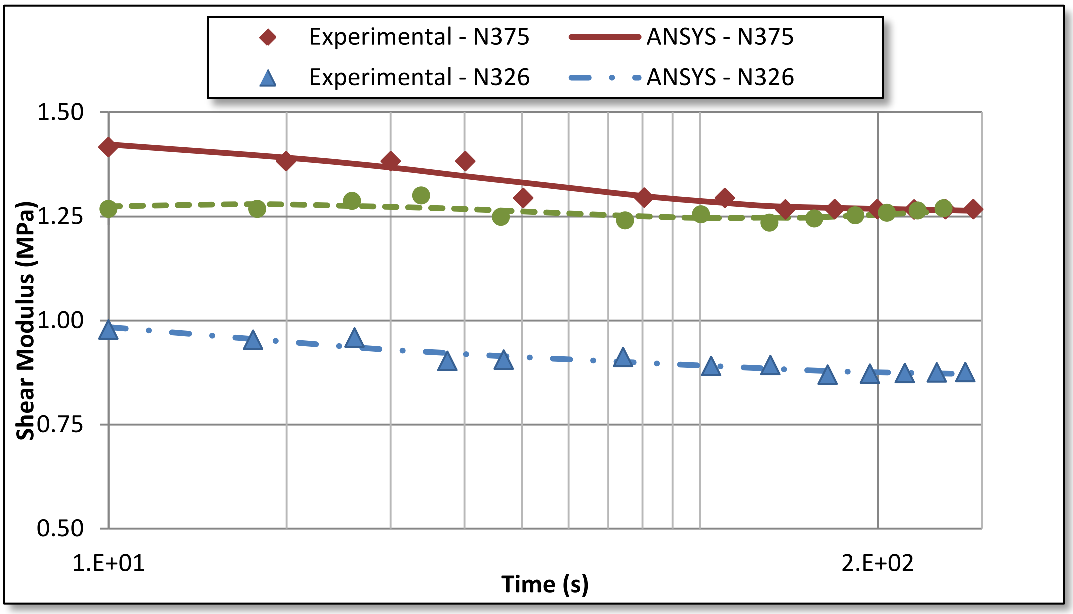


Figure 6: Plots of the experimental shear moduli vs. time for three blends as compared with ANSYS.

(a)

(b)

Figure 7: (a) Rubber sheet with central hole, (b) cyclic tensile test for rubber sheet specimen with central hole using a computerized Testometric AX M500-25kN.

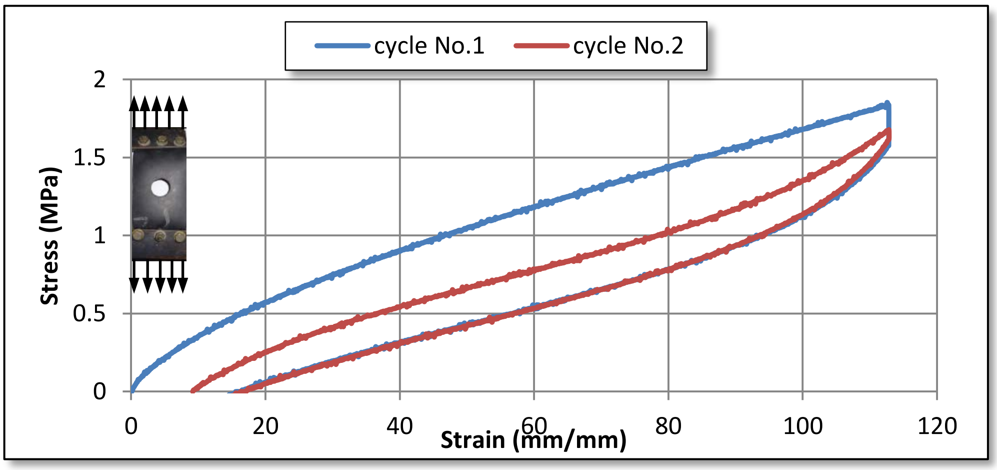
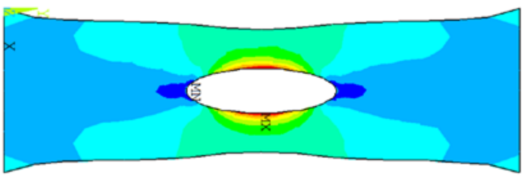
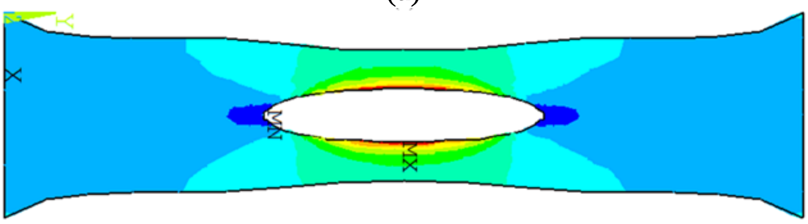


Figure 8: Two cycles for specimen with center hole and carbon black N326 at strain rate 100 mm/min.



(a)



(b)

Figure 9: Rubber sheet with central hole problem solved by ANSYS 14, (a) 50% applied final strain, (b) 100% applied final strain.