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A Stress-Strain Relationship for Filled Rubber

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A semi-empirical relationship relating stress to strain has been developed based on a model which expresses nominal stress as a function of strain invariant I_1. The relationship has been observed to be applicable to various types of black-filled rubber subjected to three different modes of deformation, namely uniaxial extension, compression and simple shear. At equal I_1, the moduli in tension, compression and simple shear were observed to be the same and prediction of the stress-strain values for one mode of deformation could be made using parameter constants for the relationship developed from another mode of deformation.

The stress-strain relationship is a fundamental property of a rubber-like material, and the ability to predict the response of an elastomer under a known load or deformation facilitates the design of engineering rubber products.

There are two main approaches in defining the constitutive equation to describe the elastic nature of rubber-like materials. The molecular approach considers the response of the molecular network to deformation. Typically, this is the statistical or Gaussian theory where the parameters are calculated from such quantities as finite molecular length and molecular weight between crosslinks[1]. There is also the phenomenological approach where the elasticity theory was derived from entirely mathematical consideration. The models of Mooney[2], Rivlin[3,4] and Valanis-Landel[5] are among those which are derived from or based on the phenomenological approach.

Neither the statistical nor the phenomenological theory can satisfactorily describe the stress-strain behaviour of filled rubber because these theories are based on the assumption that the stresses are uniquely determined by the strain imposed. The assumption is valid provided there is a complete reversibility in the stress-strain behaviour of rubber-like materials, and no hysteresis occurs. With rubber-like materials, in particular filled rubbers, the assumption is not valid. Thus, the existing statistical and phenomenological theories can not describe the stress-strain behaviour of filled rubber. This paper describes an alternative form of a stress-strain relationship which is applicable to filled rubber subjected to moderate deformation.

EXPERIMENTAL

Rubbers

All samples were based on natural rubber, using the formulation given in *Table 1*. The rubbers were vulcanised at 150°C for the time required to develop maximum torque on the Monsanto rheometer. Fillers used ranged from N110 (SAF) to N762 (SRF) with the mean particle diameter of about 10–20 mm for N110 and 60–100 mm for N762.

Stress-Strain Measurements

Tension. Measurements were made on an Instron 1122 tensile testing machine using parallel-sided dumbells (or bongo shape) 100 mm long, 2.0 mm thick held in spring-loaded grips. The strain rate was 20%/min,

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TABLE 1. FORMULATION

Compound	Sulphur system	Peroxide system
Natural rubber (SMR L)	100	100
Carbon black	5 – 60	5 – 60
Zinc oxide	5	–
Stearic acid	1.5	–
Flectol H[a]	1.0	–
Process oil[b]	0.5 – 6.0	
Sulphur	0.17 – 3.75	
CBS[c]	0.3 – 7.5	
DCP[d]	–	1 – 4.5

[a]Poly - 2, 2, 4 - trimethyl - 1, 2 - dihydroquinoline

[b]Dutrex

[c]N - cyclohexyl benzothiazole - 2 - sulphenamide

[d]Dicumyl peroxide

and samples were extended to a maximum extension of 150% (where possible).

Compression (lubricated). Compression measurements were carried out on cylinders having 25.4 mm diameter, 9.0 mm thickness using the Instron testing machines. The samples were compressed to a maximum of 50% strain at a strain rate of about 20%/min. The loaded surfaces of the rubber cylinders were lubricated using silicone oil.

Simple shear. Shear measurements were carried out on two rubber disks (6 mm thick and 25.4 mm diameter) bonded between three metal pieces. The stress-strain values were obtained by shearing the sample to 100% shear at a rate of 20%/min and all measurements were for 'static' condition.

RESULTS AND DISCUSSION

A Form of Stress-Strain Relationship

The theory of Mooney[3,4] expresses the stored energy function, W, as a function of strain invariants I_1 and I_2 where

$$I_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2$$
$$I_2 = \lambda_1^2 \; \lambda_2^2 \; \lambda_3^2 \qquad \ldots 1$$

and λ_1, λ_2 and λ_3 are the extension ratios in the three principal directions. If W is strongly dependent on both strain invariants, I_1 and I_2, then a simple mathematical equation relating the modulus in tension, compression, H ($H = \sigma/\lambda-\lambda^{-2}$, σ is the nominal stress) and simple shear, G ($G = \sigma/\gamma$, γ is the shear strain) cannot be obtained because the contributions due to I_1 and I_2 will be different for different types of strain. Only if W is independent of either I_1 or I_2 are these moduli likely to be related.

Previous work by Gregory[6] which has been restricted to filled rubber, suggested that the stored energy function for the three simple modes of deformation, *viz.* tension, compression and simple shear, was dependent only on I_1 and the contribution due to I_2 was small. This has been shown by comparing the modulus at equal value of I_1 *(Figure 1)*. If

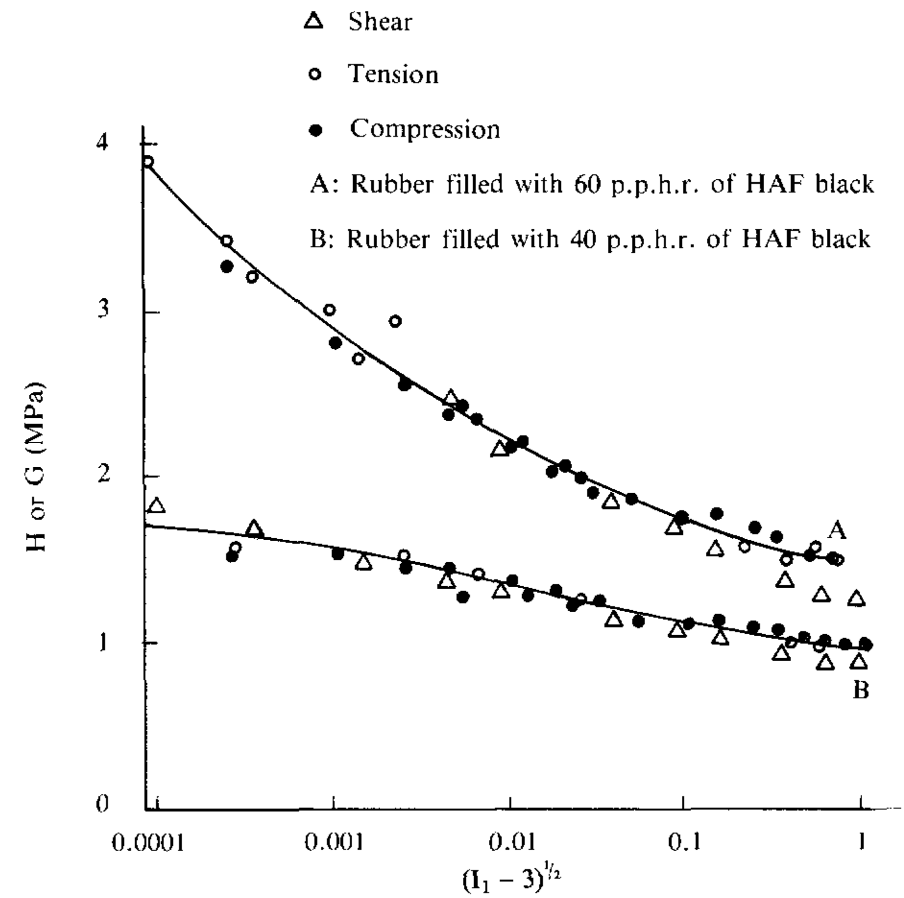

Figure 1. Variation of H and G with strain invariant I_1 for rubber filled with 60 p.p.h.r. and 40 p.p.h.r. of HAF (N330) black[6].

the partial derivative $\frac{\delta W}{\delta I_1}$ is dependent on I_2 and the contribution due to $\frac{\delta W}{\delta I_2}$ is not small, differences in the values of tensile/compression and shear modulus at equal I_1 will be observed. However, the agreement observed in practice between H and G at moderate strain suggests that serious errors are not introduced if one assumes that the moduli in tension, compression and shear are only a function of I_1.

Thus assuming that, at low to moderate strains, the moduli in tension, compression and simple shear are a function of I_1 only, then we may write, for simple extension or compression

$$\sigma_T = F(I_1)(\lambda - \lambda^{-2}) \qquad \dots 2$$

and for simple shear

$$\sigma_s = F(I_1)\,\gamma \qquad \dots 3$$

where σ_T and σ_s are the nominal tensile and shear stresses respectively, γ is the shear strain and $F(I_1)$ is a term which is a function of the strain invariant I_1. It follows that, if $F(I_1)$ is known, the stress-strain behaviour in tension, compression and simple shear can be predicted.

Let us consider a simple shear deformation. When the shear stresses are plotted against shear strain, the relationship is linear at moderate strains (*Figure 2*). The linear region may be expressed as,

$$\sigma_s = A\gamma + k \quad \text{... 4}$$

where A is the modulus at moderate strain and k the intercept. However, at low strain, *Equation 4* cannot be applicable since the predicted shear stress does not approach zero at limiting shear strain. To accommodate the behaviour at low strains, the value of k may take the form,

$$k = f(\gamma).\gamma \quad \text{... 5}$$

where $f(\gamma)$ is a function which decreases with shear strain. The simplest possible form of $f(\gamma)$ which gives the required decrease in $f(\gamma)$ with increasing γ, but which provides a finite value of $f(\gamma)$ at zero strain is:

$$f(\gamma) = \frac{1}{B\gamma + C} \quad \text{... 6}$$

where B and C are constants.

Shear strain is related to the strain invariant[1] I_1, by

$$\gamma = (I_1 - 3)^{1/2} \quad \text{... 7}$$

Substitution of *Equation 7* into *Equation 6* gives,

$$f(\gamma) = \frac{1}{B(I_1 - 3)^{1/2} + C} \quad \text{... 8}$$

From *Equations 4, 5* and *8*, it follows that the shear stress may be expressed as:

$$\sigma_s = \left[A + \frac{1}{B(I_1 - 3)^{1/2} + C} \right] \gamma \quad \text{... 9}$$

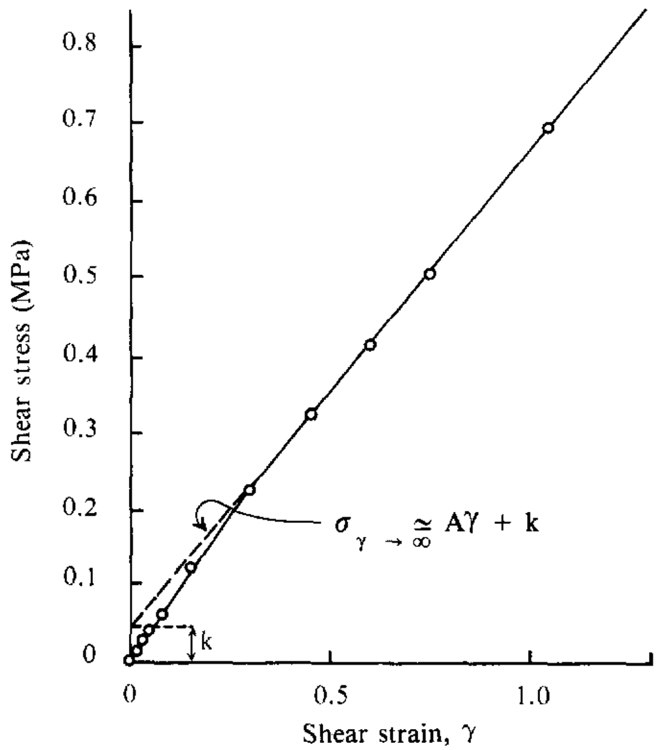

Figure 2. Shear stress as a function of shear strain for rubbers filled with 25 p.p.h.r. N330 black.

The plots of $\frac{\sigma_T}{\lambda-\lambda^{-2}}$ *versus* $(I_1-3)^{1/2}$ were to be approximately the same as the corresponding plots of $\frac{\sigma_S}{\gamma}$ *versus* $(I_1-3)^{1/2}$ for low to moderate strains, suggesting that the function $F(I_1)$ for simple extension and compression is the same as that for simple shear[6]. It follows that a more appropriate form of *Equation 9*, which is applicable to the three modes of deformation considered will be

$$\sigma = \left[A + \frac{1}{B(I_1-3)^{1/2}+C} \right] f(e) \quad \ldots 10$$

where $f(e) = \lambda - \lambda^{-2}$ for tension or compression and $f(e) = \gamma$ for simple shear.

Verification of the Stress-Strain Relationship

Correlation between stress and strain. The relationship given by *Equation 10* is applicable to tension, compression and simple shear, but verification of the expression is carried out on rubbers subjected to tension deformation because, experimentally, tensile tests are much easier to perform than shear or compression. Furthermore, repeat tests can be carried out for tubbers orginating from the same source (*i.e.* same moulded sheet), thus reducing variability.

In tension, *Equation 10* may be written as,

$$H = A + \frac{1}{B(I_1-3)^{1/2}+C} \quad \ldots 11$$

where $H\ (= \sigma/\lambda-\lambda^{-2})$ is an elastic modulus which depends on the three unknown constants, A, B, and C. Rearranging *Equation 11* gives,

$$(H-A)^{-1} = B\,(I_1-3)^{1/2} + C \quad \ldots 12$$

If *Equation 12* is valid, then plots of $(H-A)^{-1}$ *versus* $(I_1-3)^{1/2}$ should be linear with a slope B and an intercept C.

In principle, A can be estimated from the limiting value of $\frac{d\sigma}{d(\lambda-\lambda^{-2})}$ at high strain. For lightly filled (<20 p.h.r. black) rubbers, a fairly good estimation of A can be obtained graphically, but for unfilled rubbers estimation of A becomes difficult and inaccurate because the slope of σ *versus* $\lambda - \lambda^{-2}$ plots at high strains does not reach a limiting value. For heavily filled rubbers on the other hand, the on-set of non-affine deformation at relatively lower strain makes determination of A difficult.

Taking $\frac{d\sigma}{d(\lambda-\lambda^{-2})}$ from the linear portion of σ *versus* $\lambda - \lambda^{-2}$, curves as the value of A, plots of $(H-A)^{-1}$ *versus* $(I_1-3)^{1/2}$ were made. Typical results are shown in *Figures 3* and *4* and the good straight line obtained suggests that *Equation 11* gives a good description of the stress-strain behaviour for the low to moderate strain region.

It may be noted that straight lines were only obtained from the plots of $(H-A)^{-1}$ *versus* $(I_1-3)^{1/2}$ in the region before the upturn in the stress-strain curve, or in the region of affine deformation because the proposed relationship (*Equation 11*) was derived based on the stress-strain behaviour in this region.

The linearity of the plots of $(H-A)^{-1}$ *versus* $(I_1-3)^{1/2}$ is sensitive to changes in A, particularly at high strains where H approaches A. An accurate determination of A is therefore required in order to get accurate values of B and C. Since graphical determination of A was inaccurate, a statistical technique using the method of least squares was subsequently used to obtain parameters A, B and C. In the analyses, variables H and $(I_1-3)^{1/2}$ were known, but parameter A was unknown. In order to obtain parameters A, B and C by this method, values of A were fed into *Equation 12* at an incremental step of 0.01 MPa. As a first approximation, a limiting value of $\frac{d\sigma}{d(\lambda-\lambda^{-2})}$ was taken as the value A. The values of A, B and C which correspond to the maximum correlation coefficients were taken to be the best fit to the experimental data.

Typical values of parameters A, B and C and the corresponding maximum correlation coefficients obtained are shown in *Tables 2–5*.

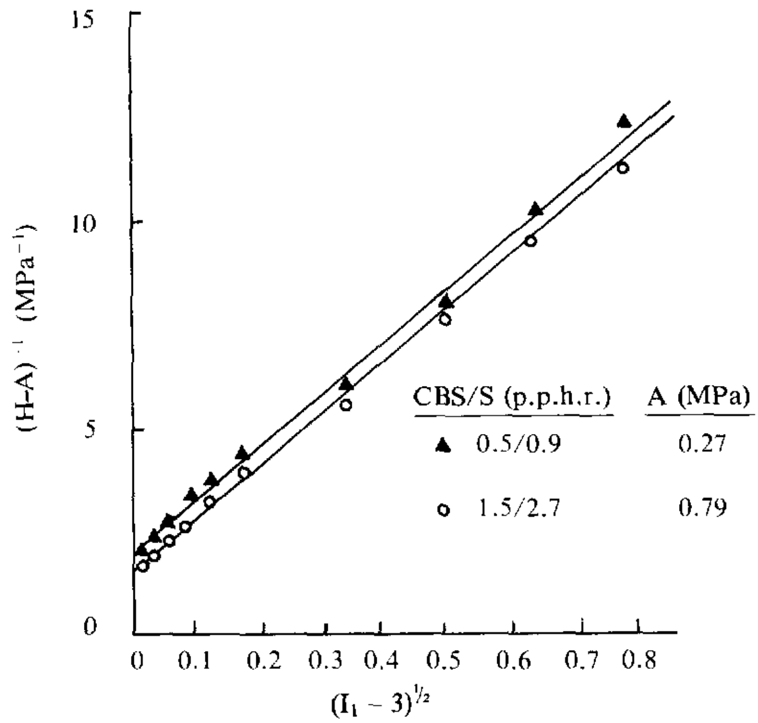

Figure 3. $(H - A)^{-1}$ *as a function of* $(I_1 - 3)^{1/2}$ *for rubbers filled with 20 p.p.h.r. N347 black.*

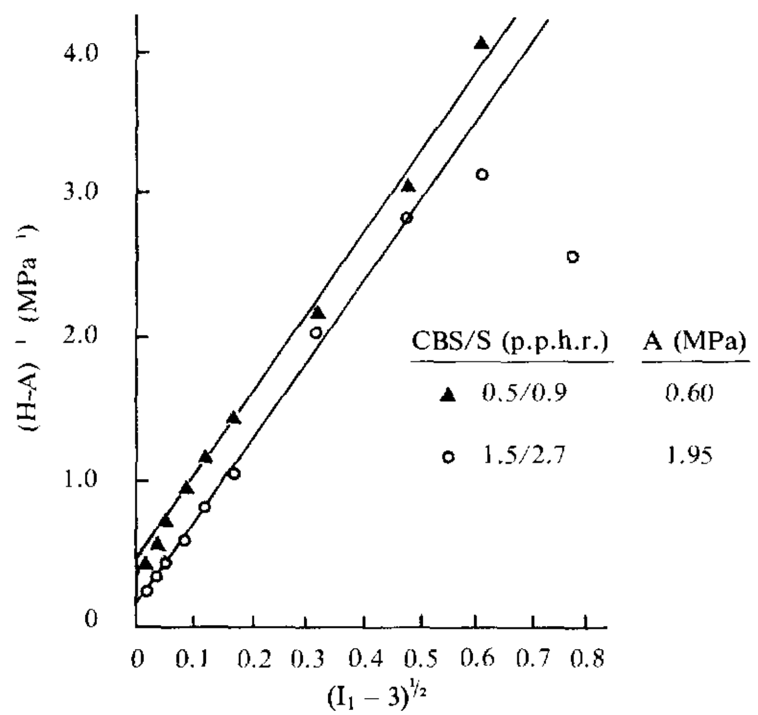

Figure 4. $(H - A)^{-1}$ *as a function of* $(I_1 - 3)^{1/2}$ *for rubbers filled with 60 p.p.h.r. N347 black.*

TABLE 2. VALUES OF A, B AND C FOR UNFILLED RUBBERS (SULPHUR AND PEROXIDE SYSTEMS)

Sulphur/Peroxide system	A (MPa)	B (MPa^{-1})	C (MPa^{-1})	Maximum correlation coefficient
CBS/S (p.p.h.r.)				
0.25/0.45	0.08	5.78	5.55	0.9990
0.5/0.9	0.15	4.73	5.37	0.9983
0.75/1.35	0.22	3.30	5.07	0.9983
1.0/1.8	0.27	2.86	5.06	0.9983
1.25/2.25	0.36	4.29	5.73	0.9976
1.5/2.7	0.450	6.83	5.74	0.9934
DCP (p.p.h.r.)				
1	0.21	7.31	6.41	0.9962
2	0.33	3.40	5.35	0.9973
3	0.50	6.67	7.23	0.9964
4	0.70	6.56	5.90	0.9965

TABLE 3. VALUES OF A, B AND C FOR SULPHUR-CURED N550 BLACK FILLED RUBBERS

Black loading (p.p.h.r.)	CBS/S (p.p.h.r.)	A (MPa)	B (MPa^{-1})	C (MPa^{-1})	Maximum correlation coefficient
20	0.25/0.45	0.08	7.17	2.89	0.9984
	0.5 /0.9	0.26	9.79	2.71	0.9988
	0.75/1.35	0.35	4.91	3.53	0.9944
	1.0 /1.8	0.56	11.40	3.90	0.9987
	1.25/2.25	0.63	11.22	3.56	0.9983
	1.5 /2.7	0.79	15.12	3.61	0.9981
40	0.25/0.45	0.15	8.47	1.97	0.9986
	0.5 /0.9	0.41	12.04	1.50	0.9975
	0.75/1.35	0.59	10.08	1.49	0.9976
	1.0 /1.8	0.79	11.82	1.51	0.9986
	1.25/2.25	0.97	11.18	1.52	0.9988
	1.5 /2.7	1.13	14.8	2.29	0.9975
60	0.25/0.45	0.21	8.74	1.25	0.9992
	0.5 /0.9	0.54	8.6	1.162	0.9992
	0.75/1.35	0.93	11.06	0.794	0.9991
	1.0 /1.8	1.10	7.45	0.918	0.9987
	1.25/2.25	1.48	8.73	0.688	0.9997
	1.5 /2.7	1.51	10.22	0.613	0.9990

TABLE 4. VALUES OF A, B AND C FOR SULPHUR-CURED N347 BLACK FILLED RUBBERS

Black loading (p.p.h.r.)	CBS/S (p.p.h.r.)	A (MPa)	B (MPa^{-1})	C (MPa^{-1})	Maximum correlation coefficient
20	0.25/0.45	0.10	10.46	2.99	0.9995
	0.5 /0.9	0.28	9.30	2.52	0.9994
	0.75/1.35	0.46	10.22	2.33	0.9991
	1.0 /1.8	0.52	9.33	2.26	0.9997
	1.25/2.25	0.68	13.93	2.34	0.9989
	1.5 /2.7	0.80	12.94	2.08	0.9993
40	0.25/0.45	0.09	8.63	1.61	0.9993
	0.5 /0.9	0.36	8.15	0.93	0.9989
	0.75/1.35	0.58	7.65	0.71	0.9996
	1.0 /1.8	0.76	8.07	0.77	0.9990
	1.25/2.25	0.93	8.09	0.57	0.9995
	1.5 /2.7	1.13	9.09	0.41	0.9982
60	0.25/0.45	0.32	7.63	0.53	0.9996
	0.5 /0.9	0.62	5.94	0.43	0.9985
	0.75/1.35	0.97	5.38	0.31	0.9995
	1.0 /1.8	1.35	7.13	0.17	0.9981
	1.25/2.25	1.57	5.00	0.19	0.9995
	1.5 /2.7	2.05	7.67	0.068	0.9940

For unfilled rubbers of different crosslink densities (*Table 2*), the maximum correlation coefficients of the range 0.9934 – 0.9990 were obtained. For sulphur-cured rubbers, the values of the maximum correlation coefficients improved with decreasing crosslink density (*i.e.* lower sulphur content) averaging about 0.9975. For peroxide-cured rubbers, the average maximum correlation coefficient was about 0.9966, which was comparable to that of the corresponding sulphur system.

Typical values of the correlation coefficients for filled rubbers are given in *Tables 3–5*. For rubbers filled with N550 black and crosslinked using the sulphur vulcanising system (semi-EV), maximum correlation coefficients from 0.9944 to 0.9992 were obtained. The correlation coefficients appeared to be generally better for those containing 60 p.h.r. black than those containing 20 p.h.r. black. The maximum correlation coefficients of rubbers filled with N347 black and crosslinked using the sulphur vulcanising system (semi-EV) ranged from 0.9940 to 0.9998, with less heavily filled rubbers (e.g. 20 p.h.r. black) having higher correlation coefficients than the heavily filled rubbers. The peroxide-cured rubbers showed equally good maximum correlation coefficients, ranging from 0.9943 to 0.9999 (*Table 5*).

Generally, the lightly filled and unfilled rubbers gave relatively poorer maximum correlation coefficients compared to heavily filled rubbers. For over two hundred rubbers tested, the maximum correlation coefficients obtained varied from 0.9934 to 0.9999, which are fairly good and these results showed that the plots of $(H-A)^{-1}$ *versus* $(I_1 - 3)^{1/2}$ were linear and that *Equation 11* is valid.

TABLE 5. VALUES OF A, B AND C FOR PEROXIDE-CURED N550 AND N347 BLACK FILLED RUBBERS

Black loading	Dicumyl peroxide (p.p.h.r.)	A (MPa)	B (MPa^{-1})	C (MPa^{-1})	Maximum correlation coefficient
20 p.p.h.r. FEF	1	0.25	14.75	4.39	0.9982
	2	0.47	23.43	4.53	0.9991
	3	0.62	15.30	4.93	0.9998
	4	0.82	25.75	3.67	0.9998
40 p.p.h.r. FEF	1	0.38	13.45	2.42	0.9991
	2	0.67	17.85	2.01	0.9995
	3	0.92	17.15	2.01	0.9997
	4	1.21	19.07	1.50	0.9995
60 p.p.h.r. FEF	1	0.51	10.45	1.23	0.9996
	2	0.91	12.06	0.80	0.9997
	3	1.29	15.25	0.70	0.9997
	4	1.64	16.36	0.57	0.9998
20 p.p.h.r. HAF–HS	1	0.26	10.60	3.09	0.9997
	2	0.46	12.93	3.30	0.9997
	3	0.60	17.39	2.63	0.9986
	4	0.87	27.92	2.40	0.9998
40 p.p.h.r. HAF–HS	1	0.37	10.13	1.20	0.9993
	2	0.64	12.54	0.87	0.9943
	3	0.95	12.80	0.72	0.9999
	4	1.18	14.15	0.69	0.9997
60 p.p.h.r. HAF–HS	1	0.58	8.25	0.63	0.9989
	2	1.01	8.23	0.43	0.9993
	3	1.37	8.61	0.43	0.9994
	4	1.74	10.21	0.30	0.9994

Comparison between experimental and predicted values. Equation 10 gives the value of H or G as a function of $(I_1 - 3)^{1/2}$, with parameters A, B and C as constants. For either tension, compression or simple shear, values of A, B and C for a particular rubber would be the same if H and G are identical at equal $(I_1 - 3)^{1/2}$. Hence, knowing the values of A, B and C for any one mode of deformation may allow prediction of stress-strain values in other modes of deformation to be made.

Values of A, B and C were obtained using the method of least squares from data obtained in simple extension. With these values of A, B and C, prediction of tensile, compressive and shear moduli were made using *Equation 11*. Typical results are shown in *Figures 5–7*, where the continuous lines represent the predicted values and the points are the experimental values.

In tension *(Figure 5)*, good agreement was obtained between the experimental and predicted values, with deviation differing with the former by not more than 3% for both unfilled and filled rubbers at low to moderate strains.

Comparisons between the experimental and predicted compressive moduli are shown in *Figure 6*. The agreement observed was also

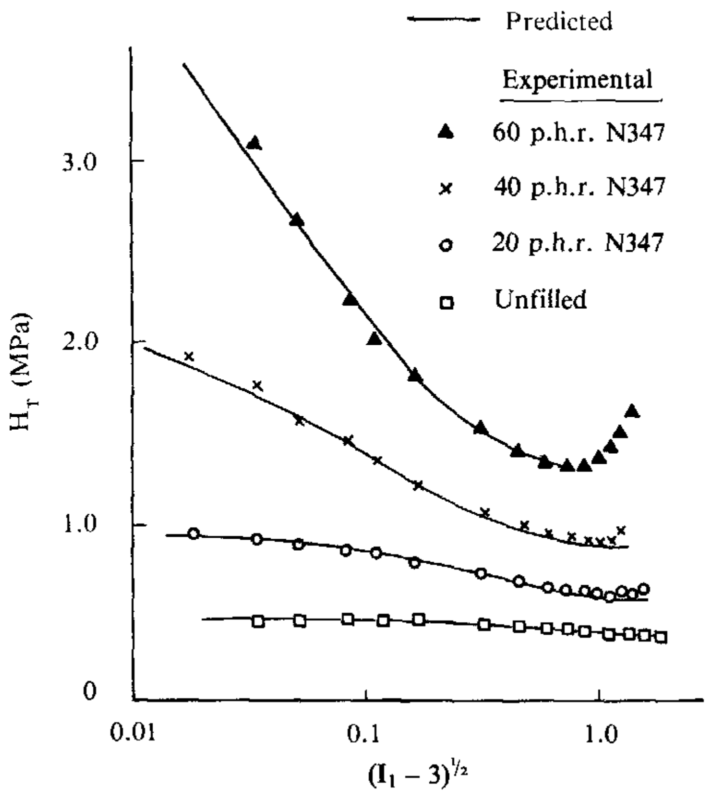

Figure 5. Comparison between experimental and predicted tensile moduli.

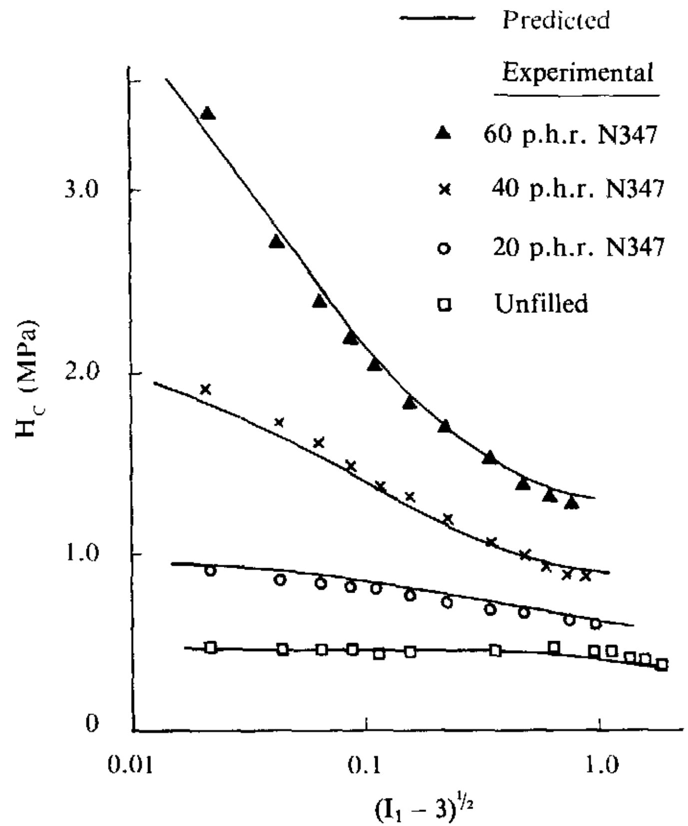

Figure 6. Comparison between experimental and predicted compressive moduli.

reasonably good, with the predicted values differing by not more than 5% from the experimental values for all rubbers tested.

For simple shear deformations *(Figure 7)*, the agreement observed between the experimental and predicted moduli was similar to those observed with compression, with not more than 5% deviation. The agreement appears to be better with heavily filled rubbers (60 p.h.r. N347) than with lightly filled rubbers.

Thus, at low to moderate strains (*i.e.* before the upturn in stress-strain curve) *Equation 10* has been shown to give a good description of the stress-strain behaviour of both filled and unfilled rubbers crosslinked with different vulcanising systems. The shear and compressive moduli were able to be predicted to within 5%, using the data from simple extension, and this will enable tests in different modes of deformation to be rationalised and simplified. The proposed equation is also more useful than the available stress-strain relationship because it is applicable to filled rubbers and it correctly predicts the non-linear stress-strain behaviour in simple shear.

Physical significance of parameters A, B and C. The proposed relationship between stress and strain takes the form

$$H \text{ or } G = A + \frac{1}{B(I_1-3)^{1/2}+C} \qquad \ldots 13$$

where $H = \sigma / \lambda - \lambda^{-2}$, $G = \sigma / \gamma$ and A, B and C are constants. The limiting conditions for the proposed relationship are:

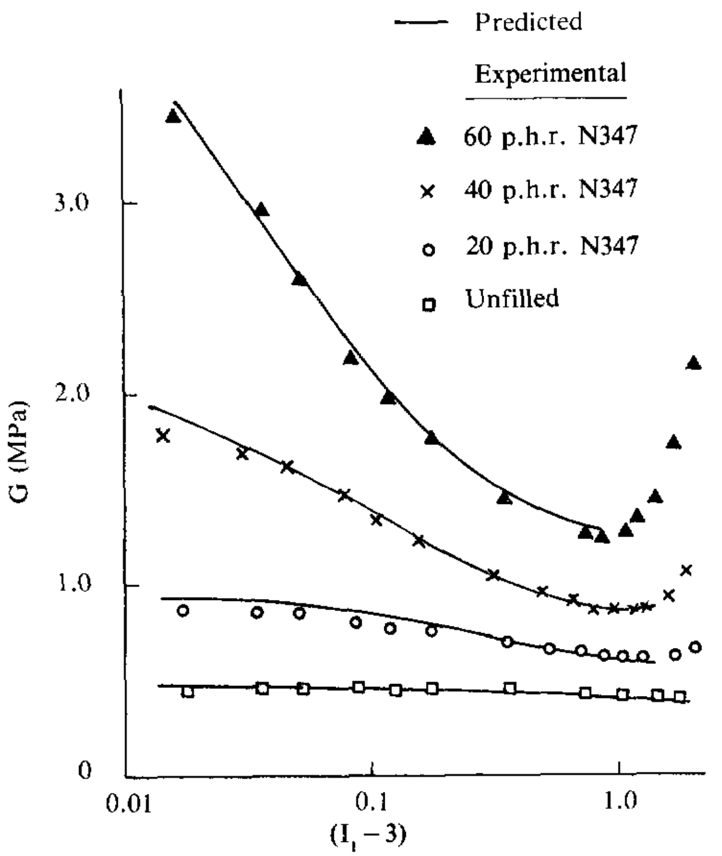

Figure 7. Comparison between experimental and predicted shear moduli.

$$\text{as } (I_1 - 3)^{1/2} \dashrightarrow 0,\ H_0 = A + \frac{1}{C} \qquad \ldots 14$$

$$\text{and as } (I_1 - 3)^{1/2} \dashrightarrow \infty,\ H_\infty = A$$

The parameter A is equivalent to H_∞ or G_∞, the modulus at high strain. According to Payne[7,8], G_∞ is the value of shear modulus which is independent of strain at sufficiently high strains *i.e.* at strains greater than those needed to breakdown any structure of carbon black.

The differrence in modulus, $H_0 - H_\infty = \frac{1}{C}$ gives the value of the change in modulus with strains[7,8], which is normally expressed as $G_0 - G_\infty$. This term has been attributed to the structural effect of carbon black agglomeration *i.e.* $G_0 - G_\infty$ arises from the breakdown of the carbon black agglomerate structures. Since $\frac{1}{C}$ is equal to $G_0 - G_\infty$, the former describes the extent of breakdown of carbon black structure due to the effects of strains.

From *Equations 13* and *14*, it is clear that the parameter B is non-contributing at the limiting strains. However, in between the two strain limits, parameter B gives a significant effect since it is associated with the strain invariant, I_1. The modulus contribution from the $B(I_1 - 3)^{1/2}$ term decreases with strain while that of A and $\frac{1}{C}$ are constant. Thus, parameter B may be associated with the manner in which H_0 changes to H_∞.

CONCLUSION

A semi-empirical relationship which describes the stress-strain behaviour of filled rubber has been developed. The relationship, which expresses nominal stress as a function of strain invariant I_1, is developed based on the assumption that the shear modulus is independent of strain at high strain.

The relationship is applicable to various types of filled rubbers subjected to low to moderate strains. It relates a modulus H in tension/compression or G in simple shear to a strain invariant I_1 and three parameters A, B, C, *viz.*

$$H \text{ or } G = A + \frac{1}{B(I_1 - 3)^{1/2} + C}$$

where $H = \sigma / \lambda - \lambda^{-2}$, $G = \sigma/\gamma$, $I_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2$ and σ, λ, γ are the nominal stress, extension ratio and shear strain, respectively. At equal I_1, the modulus H is observed to be identical to G.

The relationship enables the prediction of stress-strain behaviour of filled rubber in different modes of deformation to be made using known values of parameters A, B and C obtained from a simple mode of deformation. This enables the testing of rubber to be simplified and rationalised.

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