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The Elastic Behavior of Vulcanized Rubber

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Up to extensions of about 270 per cent the elastic behavior of vulcanized rubber can be represented analytically by a very simple function. The stress-strain diagrams for tension and compression are quite different; nevertheless this asymmetrical behavior is satisfactorily taken care of by employing only two elastic coefficients.

The mental attitude of the engineer to an organic compound such as rubber is a very skeptical one if its elastic behavior has to be compared with that of metals; nevertheless experiments with rubber belts furnished by the Goodyear Tire and Rubber Company to the Massachusetts Institute of Technology have convinced the author of this paper that a comparison of the elastic properties of rubber with those of metals is justified and at the same time very instructive as regards the mechanics of finite deformations in general.

Naturally rubber is more easily influenced by temperature and by surrounding liquid and gaseous substances which eventually cause chemical changes. However, vulcanized rubber can now be manufactured of such excellent quality that the way is opened to uses hitherto not exploited. Of these may be mentioned the use of rubber tires for the wheels of steam locomotives, enabling the latter to climb steep grades, and the wide field of application on damping vibrations and changing unfavorable critical speeds, already exemplified in the automotive industry.

Such applications can only be industrially exploited if the elastic stresses in rubber obey a known, mathematically defined law. In this paper the author therefore develops the law for large deformations and shows its agreement with the results of experiments made with the two rubber compounds that were furnished.

General Law of the Ideal Elastic Body

We call a material ideally elastic if, even after a considerable deformation, it assumes its original shape after removal of the constraints introduced and if the process of loading and unloading is not accompanied by transformations of mechanical energy into heat (cf. *J. Rheology*, 2, No. 2, 169). Nearly all materials are ideally elastic if the deformation is infinitesimal, but in the case of most substances finite deformations are accompanied by losses of mechanical energy. It is not the aim of this paper to study these losses, but it may be mentioned that there is even in the realm of ideal elasticity an unexplored region indicated by the experimental fact that the law for infinitesimal elastic deformations does not hold good for finite deformations. It is only necessary to subject a rubber cylinder to tension and compression at comparatively small strains, when the lack of agreement becomes obvious.

At the foundation of all elastic theories lies the definition of strain, and before introducing a new law of elasticity we must explain how finite strain is to be measured.

Infinitesimal strain is measured by the ratio of the increase in length to the length itself. In defining a considerable strain λ of a fiber we assume this strain to be divided into small portions $d\lambda$. Each of these portions we define, as in the case of an infinitesimal strain, by the ratio:

$$d\lambda = \frac{dx}{x} \tag{1}$$

where x is the length of a fiber in a certain state of strain. Integrating and taking the length of the fiber before stressing as a and after stressing as b, we get

$$\lambda = \log_e \frac{b}{a} \tag{1a}$$

Our measure of finite strain is therefore the natural logarithm of the ratio of the two lengths.

On the basis of the general theory developed in the Appendix we obtain the law of the simple *uniaxial* tension and compression test of rubber in the following two forms:

Stress per Unit Area Referred to the Deformed State:

$$\sigma = 2G\lambda\left(e\lambda + \frac{1}{2}e^{-\lambda/2}\right) \tag{2a}$$

Stress per Unit Area Referred to the Undeformed State:

$$\sigma_0 = 2G\lambda\left(1 + \frac{1}{2}e^{-3\lambda/2}\right) \tag{2b}$$

If P is the load, A_0 the area of cross section of the unloaded specimen, and A the area after the load is applied, we have, for incompressible material, $A = A_0e^{-\lambda}$. The stress σ_0 is the observed stress, because all testing machines measure directly the force applied and not the true physical stress σ.

The coefficient G is about 67 lb. per sq. in. for the material examined and is the same as the modulus of shear for infinitesimal deformations. For such deformations both Equations (2a) and (2b) yield

$$\sigma = \sigma_0 = 3G\lambda$$

On the basis of Equations (2a) and (2b) and taking $2G$ = 134 lb. per sq. in., Table I has been calculated and the values plotted in Fig. 1.

TABLE I

$+\lambda$	$e^{\lambda} + \frac{1}{2}e^{-\lambda/2}$	$\lambda(e^{\lambda} + \frac{1}{2}e^{-\lambda/2})$	σ	σ_0
0.0	1.500	0.0	0	8
0.2	1.674	0.3348	45	37
0.4	1.901	0.7604	102	60
0.6	2.192	1.3152	176	97
0.8	2.561	2.0488	275	124
1.0	3.021	3.021	405	140
1.2	3.594	4.32	580	174
1.4	4.303	6.03	810	200
1.6	5.178	8.29	1110	224

An analogous table for the negative values of λ is given in Table II.

TABLE II

$-\lambda$	$e^{-\lambda}+\frac{1}{2}e^{\lambda/2}$	$\lambda(e^{-\lambda}+\frac{1}{2}e^{\lambda/2})$	σ	σ_0
0.0	1.500	0.0	0	0
0.2	1.371	0.2742	37	45
0.4	1.281	0.5124	69	102
0.6	1.224	0.7344	98	179
0.8	1.195	0.9560	128	285
1.0	1.192	1.192	160	435
1.2	1.212	1.455	195	645
1.4	1.253	1.750	235	951
1.6	1.316	2.100	282	1397

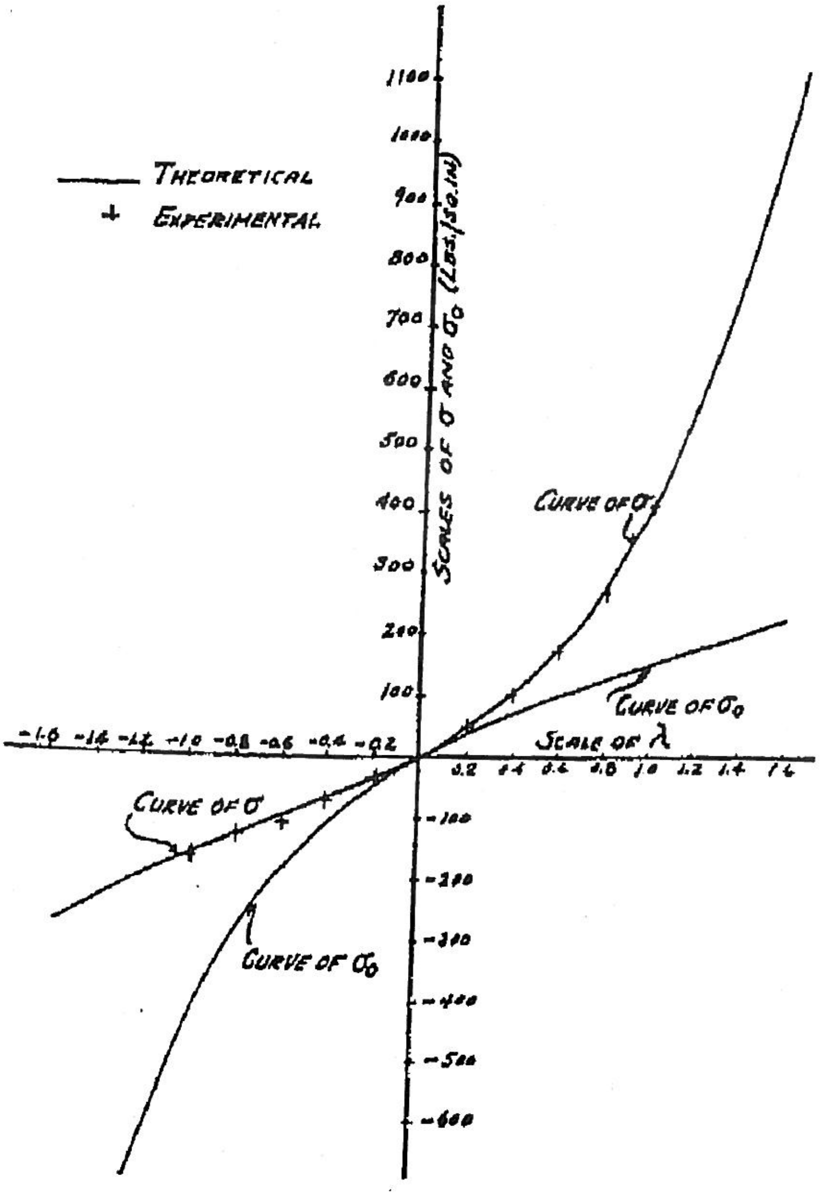

Figure 1

Experiments

The rubber with which the experiments were made was delivered in form of closed belts of 2 in. × 0.32 in. cross section and 38 in. circumference. Two different compounds (Nos. 4183 and 4799) were used and there were 12 belts in all. The values of the elastic constants are about the same for both compounds, but slight variations in the elastic constants occur even at different parts of one and the same specimen.

The Bulk Modulus.—To measure the bulk modulus, rubber cylinders 1 in. in diameter and 1.5 in. long were placed in a compression chamber and the distance between the pressure heads measured with Ames dials. The bulk modulus K was found to be 387,000 lb. per sq. in. or 27,200 kg. per sq. cm. (average of three experiments). This is somewhat larger than the modulus of water, which is 23,000 kg. per sq. cm., but agrees well with the fact that the specific gravity of rubber is about the same as that of water.

Considering the great difference between the shear and bulk moduli (their ratio is $G/K = 0.211 \times 10^{-3}$), it is justifiable to consider rubber as incompressible in the range for which validity is claimed for the elastic law (2). The commonly used modulus of elasticity E is in that case equal to $3G = 200$ lb. per sq. in.

The Tensile Test.—The tensile test under uniaxial stress is technically very easy to carry out. However, by an increase in length of 200 per cent or more, time effects and elastic strain hardening play an important role. The strain hardening is due to the microstructure of rubber and can only be taken into account by employing a statistical theory. This strain hardening has in some respects the character of internal friction; therefore the unloading curve coincides nearly with the author's theoretical curve.

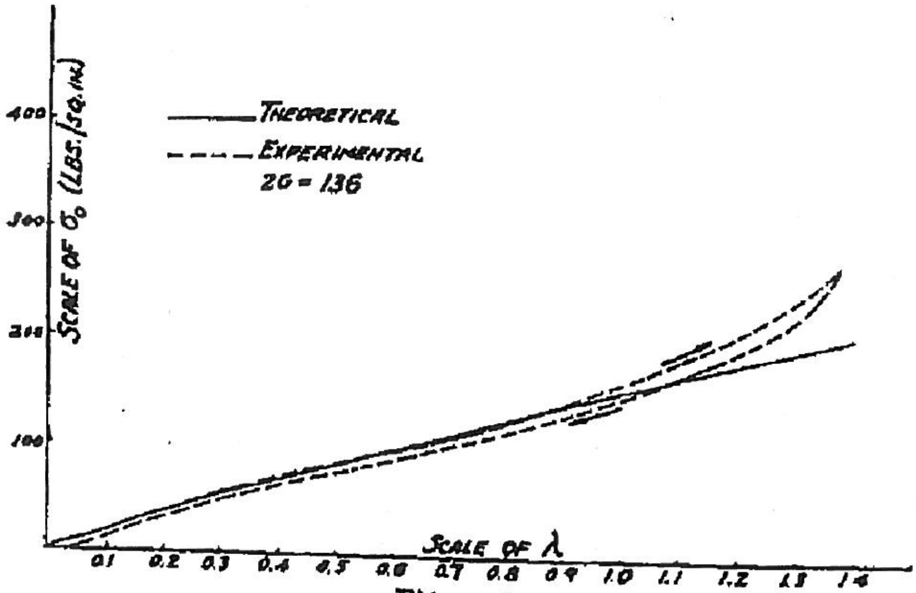

Figure 2

The curves of Fig. 2 show the loading and unloading at a constant rate of applying strain and removing stress (time of loading and unloading, 4 hrs. in all) with the stresses related to the original area of cross section. Table III gives the values of the true stresses and strains for very slow loading and unloading. The observed values of λ are reduced to equal differences by interpolation, because it would not have been possible otherwise to take the average of the experiments with three specimens.

TABLE III

+ λ	Calculated Stress σ (Equation [2a]), Lb. per Sq. In.	Measured Stress σ, Lb. per Sq. In.
0.0	0.0	0.0
0.2	44.8	44.5
0.4	101.8	100.7
0.6	176.2	171.0
0.8	275.0	270.8
1.0	405.0	410.0

The Compression Test.—The compression test is very difficult to perform and almost impossible to carry out for higher pressures if uniaxial stress has to be used. Not only is the state of uniform uniaxial stress unstable even for comparatively short rubber cylinders, but it is even difficult to realize such a state of stress. Despite careful greasing of the planes of contact with the pressure heads of the testing machine, dry friction was detected. Evidently the grease is removed under too much pressure. Table IV gives the calculated and measured stresses.

TABLE IV

$-\lambda$	Calculated Stress σ (Equation [2a]), Lb. per Sq. In.	Measured Stress σ, Lb. per Sq. In.
0.0	0.0	0.0
0.2	36.8	37.3
0.4	68.7	72.7
0.6	98.5	103.7
0.8	128.0	129.8
1.0	160.0	162.4

The change in the elastic behavior in going from positive to negative strains is very marked, but is satisfactorily represented by the theoretical formula. Figure 2 shows the deviations from the theoretical curve at the larger strains.

Conclusions

As a consequence of the work described the author arrives at the conclusion that in the deformation range from 0 to 270 per cent only one elastic constant is needed for a full mathematical description, and that this constant is the modulus of shear defined for infinitely small deformations.

If models could be made of rubber at less expense, this material could be used for the integration of the equations of elasticity. The solutions could be easily corrected for the lack of compressibility and for the change in the common law of elasticity.

APPENDIX

Mathematical Derivations

Taking an element of a body and stressing it in three directions perpendicular to each other, x_1, x_2, x_3, we get the new lengths $\overline{dx_1}$, $\overline{dx_2}$, $\overline{dx_3}$ from the old ones dx_1, dx_2, dx_3. If we denote the principal strains by ϵ_1, ϵ_2, ϵ_3, the corresponding strains are, according to (1) and (1a),

$$\epsilon_1 = \log_e\left(\frac{\overline{dx_1}}{dx_1}\right) \tag{3}$$

It is very convenient to take the average of these strains, namely,

$$\epsilon = \frac{1}{3}(\epsilon_1 + \epsilon_2 + \epsilon_3) \tag{3a}$$

whence

$$3\epsilon = \log_e\left(\frac{\overline{dx_1}\,\overline{dx_2}\,\overline{dx_3}}{dx_1\,dx_2\,dx_3}\right) = \log_e\left(\frac{\overline{dV}}{dV}\right)$$

ϵ arises from that part of the strain which is equal in all directions. Consequently if we subtract ϵ from ϵ_1, ϵ_2, ϵ_3, and denote the result by φ_1, φ_2, φ_3,

$$\varphi = \epsilon_1 - \epsilon \tag{3b}$$

will represent the pure deformation without the change in volume. We can resolve the stresses acting at any point in the same manner. $S = \frac{1}{3}(S_1 + S_2 + S_3)$ is that part of the stress which can exist in an ideal liquid. This hydrostatic part of the stress S is connected with the change of volume, the remainder of the stress, $S_1 - S$, is connected with the change of form.

Now in the case of infinitely small strains it is not necessary to state that the stresses in the deformed states are to be employed when we formulate the elastic law. It does not make any difference for which state we give the stresses. With finite deformations, however, only the true physical stresses after equilibrium is attained can be used. For any homogeneous and isotropic material we can write down easily the simplest form the elastic energy can assume. We introduce into this two moduli, the modulus of shear G and the bulk modulus K, and choose the simplest possible analytical form, which becomes the expression of the classical theory of elasticity when we assume infinitely small displacements:

$$2W = 2G(\varphi_1^2 + \varphi_2^2 + \varphi_3^2) + 9K\epsilon^2 \tag{4}$$

W is the elastic energy per unit volume in the undeformed state. The energy must be related to the undeformed state because it is a physical entity connected with the mass of the body. Equation (4) is naturally an assumption and has to be checked by experiment.

But there is yet another doubt. Must we not relate the formula for the energy to the real bearer of the energy, the molecule? If the substance is built up from such units, should we not assume expression (4) as the energy of the molecule? Our experimental results indeed point in this direction. But before we study this question we shall formulate another expression for the work done which is independent of any law of elasticity.

If dV is the volume before and $\overline{dV}$ the volume after the deformation,

$$\delta W dV = S_1\, \overline{dx_2}\, \overline{dx_3}\, \delta\, \overline{dx_1} + S_2\, \overline{dx_1}\, \overline{dx_3}\, \delta\, \overline{dx_2} + S_3\, \overline{dx_1}\, \overline{dx_2}\, \overline{dx_3}$$

or

$$\delta W = \frac{\overline{dV}}{dV}(S_1\delta\epsilon_1 + S_2\delta\epsilon_2 + S_3\delta\epsilon_3) = e^{3\epsilon}(S_1\delta\epsilon_1 + S_2\delta\epsilon_2 + S_3\delta\epsilon_3) \tag{5}$$

$e^{3\epsilon}$ is the relative volume, and will be denoted by v. Assuming now that Equation (4) is applied to isotropic and continuous matter, we can immediately derive the law of elasticity.

By differentiating [4],

$$\begin{aligned}\delta W &= 2G[\varphi_1(\delta\epsilon_1 - \delta\epsilon) + \varphi_2(\delta\epsilon_2 - \delta\epsilon) + \varphi_3(\delta\epsilon_3 - \delta\epsilon)] + 3K\epsilon(\delta\epsilon_1 + \delta\epsilon_2 + \delta\epsilon_3)\\ &= \delta\epsilon_1(2G\varphi_1 + 3K\epsilon) + \delta\epsilon_2(2G\varphi_2 + 3K\epsilon) + \delta\epsilon_3(2G\varphi_3 + 3K\epsilon)\end{aligned}$$

Comparing this expression with Equation (5),

$$S_1 v = 2G\varphi_1 + 3K\epsilon \tag{6a}$$

or in another form,

$$\left.\begin{aligned} S_1 - S &= \frac{2G}{v}\varphi_1\\ S &= \frac{K}{v}\log_e v\end{aligned}\right\} \tag{6b}$$

The apparent increase of the modulus of shear for high compression measured by Bridgman is accounted for in Equation (6b), because v is in that case less than unity.

For rubber v differs but little from unity so long as S is in the range of 0–4000 lb. per sq. in.

Under these circumstances we can neglect the change in volume entirely, writing

$$S_1 = 2G\varphi_1 + S \tag{7}$$

for an ideal incompressible material.

But having considered an element dx_1, dx_2, dx_3 of continuous matter which led us to Equation (7), we now assume an element containing thousands of molecules. As it does not matter which form we assume them to be in, let us take them as small spheres. The true shape of the molecule of rubber is not known, but it probably is of a prismatic form. We must keep clearly in mind, therefore, that we are neglecting the true microstructure of rubber in assuming spherical molecules.

Here we no longer apply Equation (7) to the stresses, but to the forces with which the molecules act on one another, writing

$$P_1 - P = 2G'\varphi_1 \tag{8}$$

where $P = {}^1/_3\,(P_1 + P_2 + P_3)$.

If N is the number of molecules lying in the unit of area of a certain cross section in the undeformed state, then after deformation there will be in the $\overline{dx_2}$, $\overline{dx_3}$ plane the number Ne^{φ_1} molecules since

$$\frac{\overline{dx_2}}{dx_2}\,\frac{\overline{dx_3}}{dx_3} = \frac{\overline{dx_1}}{dx_1} = e^{-\varphi_1}$$

for $\epsilon = 0$.

Multiplying Equation (8) by Ne^{φ_1} gives the true stresses

$$\begin{aligned} Ne^{\varphi_1}(P_1 - P) &= 2G'N\varphi_1 e^{\varphi_1} \\ Ne^{\varphi_2}(P_2 - P) &= 2G'N\varphi_2 e^{\varphi_2} \\ Ne^{\varphi_3}(P_3 - P) &= 2G'N\varphi_3 e^{\varphi_3} \end{aligned}$$

These stresses now comprise an amount of hydrostatic stress which has to be subtracted. As the hydrostatic stresses in incompressible material are not elastically but statically determined, we can so determine them and, putting $G'N = G$, obtain

$$S_1 - S = 2G\left[\varphi_1 e^{\varphi_1} - \left(\frac{\varphi_1 e^{\varphi_1} + \varphi_2 e^{\varphi_2} + \varphi_3 e^{\varphi_3}}{3}\right)\right] \tag{9}$$

Putting $\varphi_1 = \lambda$, $\varphi_2 = \varphi_3 = -\,{}^1/_2\lambda$, $S_1 = \sigma$, and $S = {}^1/_3\,\sigma$ in (9) we get Equation (2a), and then, after dividing by e^{λ}, Equation (2b).

It is easy to generalize the law (9) so that it can be applied to other substances. We wish, however, to conserve under all circumstances the condition that the stresses can be derived from an elastic potential.

If we put

$$\left.\begin{aligned} S_1 - S &= \frac{2G}{v}\left\{f(\varphi_1) - \frac{1}{3}\,[f(\varphi_1) + f(\varphi_2) + f(\varphi_3)]\right\} \\ S &= \frac{3K}{v}\,\psi(\epsilon) \end{aligned}\right\} \tag{10}$$

the work done by the stresses, with $\epsilon_1 = \epsilon + \varphi_1$, $\epsilon_2 = \epsilon + \varphi_2$, $\epsilon_3 = \epsilon + \varphi_3$, will be, according to (5),

$$\delta W = [(S_1 - S)\delta\varphi_1 + (S_2 - S)\delta\varphi_2 + (S_3 - S)\delta\varphi_3 + 3S\delta\epsilon]v$$

Introducing the value of $S_1 - S$ from (10),

$$W = 2G\left[\int_0^{\varphi_1} f(\varphi_1)\delta\varphi_1 + \int_0^{\varphi_2} f(\varphi_2)\delta\varphi_2 + \int_0^{\varphi_3} f(\varphi_3)\delta\varphi_3\right] + 9K\int_0^{\epsilon} \psi(\epsilon)\delta\epsilon \quad (11)$$

The functions $f(\varphi_1)$ and $\psi(\epsilon)$ must therefore be obtained experimentally.

The use of Equation (11) is more satisfactory than that of expressions which can be considered only as interpolation formulas of the experiments, because, according to (11), the work done is independent of the manner in which the material is loaded.

In the study of the behavior of rubber under deformations greater than 270 per cent we cannot neglect the influences of the time effects comprising relaxation and creeping (see Fig. 2). The shape of the rubber molecules also plays an important part in the behavior under very large deformation.

The study of these deformations is not important for technical purposes because in practice such large strains are never used, but the subject is very interesting from the standpoint of physical chemistry, because it enables definite conclusions to be drawn concerning the microstructure of rubber.

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