# Strain Energy Functions for Filled Elastomers* 

M. SHINOZUKA and A. M. FREUDENTHAL,<br>Department of Civil Engineering, Columbia University, New York, New York

## Synopsis

It is the purpose of this research to investigate possible forms of elastic strain energy functions which reproduce the fundamental behavior of compressible, isotropic filled elastomers as observed in experiments. Three different forms of the strain energy function are considered, although they can be classified into two essentially different classes: in the first case, the function is assumed to be the sum of the distortional and the volumetric strain energy, while in the second it represents a possible modification of Mooney's strain energy function. For these three strain energy functions, the relations between axial and lateral deformations under uniaxial tension are plotted together with the result of experiments performed on dumbbell-shaped specimens of a KCl -filled polyurethane. The relations in uniaxial tension and compression between the axial stress and deformation are also plotted.

## I. INTRODUCTION

The purpose of this research is to investigate possible forms of the elastic strain energy functions for compressible, isotropic filled elastomers which can reproduce the fundamental behavior of the material as observed in experiments.

Two essentially different forms are assumed for the strain energy function $W$, which is a function of the three invariants $I_{1}, I_{2}$, and $I_{3}$ of the deformation tensor and which is measured per unit volume of undeformed body, where

$$
\begin{gather*}
I_{1}=\lambda_{1}{ }^{2}+\lambda_{2}{ }^{2}+\lambda_{3}{ }^{2} \\
I_{2}=\lambda_{1}{ }^{2} \lambda_{2}{ }^{2}+\lambda_{2}{ }^{2} \lambda_{3}{ }^{2}+\lambda_{3}{ }^{2} \lambda_{1}{ }^{2} \\
I_{3}=\lambda_{1}{ }^{2} \lambda_{2}{ }^{2} \lambda_{3}{ }^{2} \tag{1}
\end{gather*}
$$

$\lambda_{1}, \lambda_{2}$, and $\lambda_{3}$ being the three principal stretches of the deformation. ${ }^{1}$
In the first case, $W$ is assumed to be the sum of the distortional energy $W_{1}$ and the volumetric strain energy $W_{2}$.

$$
\begin{equation*}
W\left(I_{1}, I_{2}, I_{3}\right)=W_{1}\left(I_{1}, I_{2}\right)+W_{2}\left(I_{3}\right) \tag{2}
\end{equation*}
$$

where $W_{1}$ vanishes when the principal stretches of the deformation are identical, while $W_{2}$ is zero when no volume change is observed during deformation.

[^0]In the second case, Mooney's strain energy function valid for incompressible material ${ }^{2}$ is modified to include the effect of the dilatational strain so that it becomes a function of all three invariants but reduces to Mooney's form [eq. (3)] under volume constant infinitesimal deformation.

$$
\begin{equation*}
W\left(I_{1}, I_{2}\right)=(\mu / 4)\left[(1+a)\left(I_{1}-3\right)+(1-a)\left(I_{2}-3\right)\right] \tag{3}
\end{equation*}
$$

where $\mu$ is the shear modulus for infinitesimal deformation and $a$ is the coefficient of asymmetry introduced by Mooney.

In both cases, such forms of the strain energy function are chosen that the condition

$$
\begin{equation*}
\left(\partial W / \partial I_{1}\right)+\lambda^{2} \alpha\left(\partial W / \partial I_{2}\right) \geq 0 \tag{4}
\end{equation*}
$$

is satisfied, in addition to the well-known requirement that the strain energy function and the stresses are zero at zero deformation. Equation (4) is a necessary and sufficient condition for the greatest (smallest) tension to coincide with the direction of the greatest (smallest) stretch. ${ }^{1}$ The identity in eq. (4) is satisfied only when $\lambda_{\beta}=\lambda_{\alpha}, \lambda_{\alpha}, \lambda_{\beta}$, and $\lambda_{\gamma}$ being the three principal stretches.

Moreover, the well-known identity ${ }^{3}$

$$
\begin{equation*}
K=\left.v \frac{d^{2} W^{*}}{d v^{2}}\right|_{v=1} \tag{5}
\end{equation*}
$$

should be satisfied; here $K$ is the bulk modulus in infinitesimal deformation,

$$
v=I_{3}^{1 / 2}=d V / d V_{0}
$$

with $d V_{0}$ and $d V$ being the volume element in the reference and the deformed state, respectively, and $W^{*}$ the strain energy under pure dilatational deformation. Hence $W^{*}$ is a function of $v$ only and obtained from $W\left(I_{1}\right.$, $I_{2}, I_{3}$ ), replacing $I_{1}, I_{2}$, and $I_{3}$ by $3 v^{2 / 3}, 3 v^{1 / 3}$ and $v^{2}$, respectively.

Finally, it is important that the selected strain energy function should reproduce the observed relations between the lateral contraction $\lambda_{2}$ and the longitudinal extension $\lambda_{1}$ of a cylindrical or prismatic bar under conditions of simple extension ( $\lambda_{2}=\lambda_{3}$ ) defined by

$$
\begin{align*}
x_{1} & =\lambda_{1} X_{1} \\
x_{2} & =\lambda_{2} X_{2} \\
x_{3} & =\lambda_{3} X_{3} \tag{6}
\end{align*}
$$

where $x_{i}$ and $X_{i}(i=1,2,3)$ denote, respectively, the spatial and material coordinates within the same rectangular Cartesian reference frame. The $\lambda_{1}-\lambda_{2}$ relations usually observed are such that $\lambda_{2}$ is a monotonically decreasing function of $\lambda_{1}$.

It appears that there are several strain energy functions of the form of eq. (2) or of the modified form of eq. (3) which reproduce the $\lambda_{1}-\lambda_{2}$ relations compatible with experimental results. In the following section, three different strain energy functions are derived on imposing certain additional
conditions. Methods of introducing the third invariant $I_{3}$ into the strain energy function are suggested by the derivation of these three particular forms of the strain energy function.

## II. DERIVATION OF STRAIN ENERGY FUNCTIONS

The general relations of the theory of finite deformation used in the subsequent analysis are found, for example, in the work by Eringen. ${ }^{1}$

In curvilinear coordinate systems with metric tensors $g$ for the spatial coordinates $x^{i}(i=1,2,3)$ and $\mathbf{G}$ for the material coordinates $X^{L}(L=$ $1,2,3)$, the covariant components $E_{L M}$ of the material strain tensor are, with the usual convention of summation over repeated indices,

$$
\begin{equation*}
E_{L M}=1 / 2\left(g_{i t} x_{L}^{i} x^{i} x_{, M}^{i}-G_{L M}\right) \tag{7}
\end{equation*}
$$

The contravariant components $T^{\mathrm{LM}}$ and $t^{i j}$ of the material and the spatial stress tensor are, respectively,

$$
\begin{equation*}
T^{L M}=\partial W / \partial E_{L M} \tag{8}
\end{equation*}
$$

and

$$
\begin{equation*}
t^{i j}=I_{3}^{-1 / 2} T^{L M} x^{i},{ }_{L} x^{i},{ }_{M} \tag{9}
\end{equation*}
$$

where $g_{i j}$ and $G_{L M}$ are the covariant components of $\mathbf{g}$ and $\mathbf{G}$, respectively, and $x^{i},{ }_{L}=\partial x^{i} / \partial X^{L}$.

For the deformation described by eq. (6), eqs. (7-9) take the form

$$
\begin{gather*}
{\left[E_{L M}\right]=\left[\begin{array}{ccc}
1 / 2\left(\lambda_{1}{ }^{2}-1\right) & 0 & 0 \\
0 & 1 / 2\left(\lambda_{2}{ }^{2}-1\right) & 0 \\
0 & 0 & 1 / 2\left(\lambda_{3}{ }^{2}-1\right)
\end{array}\right]}  \tag{10}\\
{\left[\mathrm{T}^{L M}\right]=\left[\begin{array}{ccc}
\partial W / \partial E_{11} & 0 & 0 \\
0 & \partial W / \partial E_{22} & 0 \\
0 & 0 & \partial W / \partial E_{33}
\end{array}\right]} \tag{11}
\end{gather*}
$$

and

$$
\left[t^{i j}\right]=\left[\begin{array}{ccc}
I_{3}{ }^{-1 / 2} \lambda_{1}{ }^{2} T^{11} & 0 & 0  \tag{12}\\
0 & I_{3}{ }^{-1 / 2} \lambda_{2}{ }^{2} T^{22} & 0 \\
0 & 0 & I_{3}{ }^{-1 / 2} \lambda_{3}{ }^{2} T^{33}
\end{array}\right]
$$

$$
\begin{aligned}
& \text { Strain Energy Functions of the Form of Eq. (2) with } \\
& \qquad W_{2}\left(I_{3}\right)=A\left(1-I_{3}{ }^{-\alpha}\right)-B\left(1-I_{3}{ }^{-\beta}\right)
\end{aligned}
$$

The first strain energy function $W$ discussed is of the form of eq. (2) in which

$$
\begin{equation*}
W_{1}\left(I_{1}, I_{2}\right)=C I_{1}\left(I_{1}{ }^{2}-3 I_{2}\right) \tag{13}
\end{equation*}
$$

and

$$
\begin{equation*}
W_{2}\left(I_{3}\right)=A\left(1-I_{3}^{-\alpha}\right)-B\left(1-I_{3}^{-\beta}\right) \tag{14}
\end{equation*}
$$



Fig. 1. Potential energy $\phi(d)$ and strain energy $W_{2}{ }^{*}$ due to purely dilatational deformation.
$W_{1}\left(I_{1}, I_{2}\right)$ according to eq. (13) represents the distortional energy which must be zero when the deformation is purely dilatational, for it can be shown that

$$
\begin{equation*}
I_{1}{ }^{2}-3 I_{2}=1 / 2\left[\left(\lambda_{1}{ }^{2}-\lambda_{2}{ }^{2}\right)^{2}+\left(\lambda_{2}{ }^{2}-\lambda_{3}{ }^{2}\right)^{2}+\left(\lambda_{3}{ }^{2}-\lambda_{1}{ }^{2}\right)^{2}\right] \tag{15}
\end{equation*}
$$

Since it can also be shown that

$$
\begin{equation*}
\left(\partial W / \partial I_{1}\right)+\lambda_{\alpha}^{2}\left(\partial W / \partial I_{2}\right)=3\left[\lambda_{\beta}^{4}+\lambda_{\gamma}{ }^{2} \lambda_{\beta}{ }^{2}+\lambda_{\gamma}^{4}\right]>0 \tag{16}
\end{equation*}
$$

eq. (4) is also satisfied.
$W_{2}\left(I_{3}\right)$ in eq. (14) is assumed on the basis of Mie's equation ${ }^{4}$ of the form of eq. (17) representing the potential energy of one pair of interacting particles as a function of their distance $d$ :

$$
\begin{equation*}
\phi(d)=-a d^{-m}+b d^{-n} \tag{17}
\end{equation*}
$$

The first term of the right-hand side of eq. (17) represents the potential of attraction, the second that of repulsion, where the integers $n$ and $m$ are such that $n>m$ for a possible equilibrium distance $d_{0}$. Summation over all molecular pairs gives a total "lattice energy" per mole of

$$
\begin{equation*}
\Phi=-A^{\prime} d^{-m}+B^{\prime} d^{-n} \tag{17a}
\end{equation*}
$$

where $A^{\prime}$ and $B^{\prime}$ are constants proportional to Avogadro's number, but depending also on the molecular structure as well as on the values of $m$ and $n$.

When the deformation is purely dilatational, $d^{3}$ and $d_{0}{ }^{3}$ may be assumed proportional to the deformed volume element $d V$ and the undeformed volume element $d V_{0}$, respectively. Therefore, the strain energy per unit volume of the undeformed body can be written in the following form:

$$
\begin{equation*}
W_{2}^{*}=-A v^{-m / 3}+B v^{-n / 3}=-A I_{3}^{-\alpha}+B I_{3}^{-\beta} \tag{17b}
\end{equation*}
$$

where $\alpha=m / 6$ and $\beta=n / 6$. Hence, one has eq. (14) except for the constants which are added, so that $W_{2}=0$ when $I_{3}=1.0$.

Hence, the strain energy function is

$$
\begin{equation*}
W=C I_{1}\left(I_{1}{ }^{2}-3 I_{2}\right)+A\left(1-I_{3}^{-\alpha}\right)-B\left(1-I_{3}^{-\beta}\right) \tag{18}
\end{equation*}
$$

Equations (17) and (17b) are schematically presented in Figures $1 a$ and $1 b$, respectively. The derivative of $\phi(d)$ with respect to $d$ gives the interacting force between particles. This force is also plotted in Figure 1 (broken line), in which $d^{\prime}$ indicates a critical distance between two neighboring particles beyond which irreversible separation of the particles will occur. This implies that the pure expansion is reversible only when $I_{3}<$ $\left(d^{\prime} / d_{0}\right)^{6}$.

Substituting eq. (18) into eq. (12), one obtains

$$
\begin{equation*}
t^{22}=\left(\lambda_{2} / \lambda_{1} \lambda_{3}\right)\left[6 C\left(\lambda_{2}{ }^{2} I_{1}-I_{2}\right)+2 \lambda_{1}{ }^{2} \lambda_{3}{ }^{2}\left(A \alpha I_{3}^{-(1+\alpha)}-B \beta I_{3}^{-(1+\beta)}\right)\right] \tag{19}
\end{equation*}
$$

When the deformation is infinitesimal,

$$
\begin{align*}
&\left|e_{\alpha}\right|=\left|\lambda_{\alpha}-1\right| \ll 1 \quad \alpha=1,2,3  \tag{20}\\
& I_{1} \simeq 3+2 \vartheta \\
& I_{2} \simeq 3+4 \vartheta \\
& I_{3} \simeq 1+2 \vartheta
\end{align*}
$$

and therefore, eq. (19) can be approximated by

$$
\begin{equation*}
t^{22} \simeq 6 C\left(6 e_{2}-2 \vartheta\right)-4\left(A \alpha^{2}-B \beta^{2}\right) \vartheta \tag{21}
\end{equation*}
$$

where $e_{1}, e_{2}$, and $e_{3}$ are the principal extensions and $\vartheta=e_{1}+e_{2}+e_{3}$. In deriving eq. (21), it is assumed that

$$
\begin{equation*}
A \boldsymbol{\alpha}-B \beta=0 \tag{22}
\end{equation*}
$$

so that $t^{22}=0$ when $e_{1}=e_{2}=e_{3}=\vartheta=0$.
Since $A \alpha$ and $B \beta$ have the dimension of stress, one can set

$$
\begin{equation*}
A \alpha=B \beta=\mu / 2 k \tag{22a}
\end{equation*}
$$

and obtain from eq. (21)

$$
\begin{equation*}
t^{22} \simeq[(2 \mu / k)(\beta-\alpha)-12 C] \vartheta+36 C e_{2} \tag{23}
\end{equation*}
$$

where $\mu$ is the shear modulus of the material for infinitesimal deformation and $k$ is a positive number which will be determined later.

Equation (23) has to be identical with Hooke's generalized law in the linearized theory of elasticity;

$$
\begin{equation*}
t^{22} \simeq \lambda \vartheta+2 \mu e_{2} \tag{24}
\end{equation*}
$$

where

$$
\lambda=2 \mu \nu /(1-2 \nu)
$$

Comparison of eq. (23) with eq. (24) provides the relations

$$
\begin{gather*}
c=\mu / 18 \\
k=3(1-2 \vartheta)(\beta-\alpha) /(1+\vartheta) \tag{25}
\end{gather*}
$$

Equations (25) express $C$ and $k$ in terms of the shear modulus $\mu$, Poisson's ratio $\nu$ for infinitesimal deformations, as well as $\alpha(=m / 6)$ and $\beta(=n / 6)$, which are also material constants.

From eqs. (18, 22a, and 25), one obtains

$$
\begin{equation*}
W=(\mu / 18) I_{1}\left(I_{1}{ }^{2}-3 I_{2}\right)+(\mu / 2 k \alpha)\left(1-I_{3}^{-\alpha}\right)-(\mu / 2 k \beta)\left(1-I_{3}^{-\beta}\right) \tag{26}
\end{equation*}
$$

Making use of the second of eqs. (25), one can easily show that $W^{*}(v)$ associated with eq. (26)

$$
\begin{equation*}
W^{*}(v)=(\mu / 2 k \alpha)\left(1-v^{-2 \alpha}\right)-(\mu / 2 k \beta)\left(1-v^{-2 \beta}\right) \tag{27}
\end{equation*}
$$

satisfies eq. (5).

## Strain Energy Function of the Form of Eq. (2) Producing a Relation $\lambda_{2}=$ $\lambda_{1}{ }^{-\nu}$ under Uniaxial Tension and Compression

A strain energy function which, under uniaxial deformation described by eq. (6) with $\lambda_{2}=\lambda_{3}$, produces a $\lambda_{1}-\lambda_{2}$ relation of the form

$$
\begin{equation*}
\lambda_{2}=\lambda_{1}^{-\nu} \quad(\nu>0) \tag{28}
\end{equation*}
$$

or

$$
\begin{equation*}
\left(1+e_{2}\right)=\left(1+e_{1}\right)^{-\nu} \tag{28a}
\end{equation*}
$$

is derived because of extensive use of eq. (28) in previous work. ${ }^{5,6}$
Equation (28a) tends towards

$$
\begin{equation*}
e_{2}=-\nu e_{1} \tag{28b}
\end{equation*}
$$

for small values of $e_{1}$ and $e_{2}$. Hence $\nu$ denotes Poisson's ratio for small strain. With reference to eq. (28), $\nu$ can thus be considered as a generalized Poisson ratio for logarithmic definition of strain

$$
\begin{equation*}
\nu=-\left(\ln \lambda_{2} / \ln \lambda_{1}\right) \tag{28c}
\end{equation*}
$$

as suggested by Smith. ${ }^{6}$
The strain energy function is assumed to be of the form of eq. (2) with $W_{1}\left(I_{1}, I_{2}\right)$ according to eq. (13) :

$$
\begin{equation*}
W=\mathrm{C} I_{1}\left(I_{1}{ }^{2}-3 I_{2}\right)+W_{2}\left(I_{3}\right) \tag{29}
\end{equation*}
$$

$W_{2}\left(I_{3}\right)$ in the last equation is determined from the condition

$$
\begin{equation*}
T^{22}=\partial W / \partial E_{22}=0 \tag{30}
\end{equation*}
$$

for uniaxial tension or compression.
From eqs. (29) and (30), one obtains

$$
\begin{equation*}
d W_{2} / d I_{3}=-3 C\left[\left(\lambda_{2}{ }^{2} / \lambda_{1}{ }^{2}\right)-1\right] \tag{31}
\end{equation*}
$$

If $\lambda_{1}=\lambda_{2}{ }^{-\nu}$, then

$$
\begin{gathered}
I_{3}=\lambda_{1}{ }^{2} \lambda_{2}{ }^{4}=\lambda_{1}{ }^{2(1-2 \nu)} \\
\lambda_{1}=I_{3}{ }^{1 /[2(1-2 \nu)]}
\end{gathered}
$$

and

$$
\lambda_{2}=I_{3}^{-\nu /[2(1-2 \nu)]}
$$

Hence, eq. (31) has a solution $\lambda_{2}=\lambda_{1}{ }^{-v}$ if $W_{2}$ satisfies the equation

$$
\begin{equation*}
d W_{2} / d I_{3}=-3 C\left[I_{3}^{-(1+\nu) /(1-2 i)}-1\right] \tag{32}
\end{equation*}
$$

Upon integration,

$$
\begin{equation*}
W_{2}\left(I_{3}\right)=3 C\left\{I_{3}+[(1-2 \nu) / 3 \nu] I_{3}^{-3 \nu /(1-2 \nu)}-[(1+\nu) / 3 \nu]\right\} \tag{33}
\end{equation*}
$$

where a constant has been added so that $W_{2}\left(I_{3}\right)=0$ for $I_{3}=1.0$.
This method of determining $W_{2}\left(I_{3}\right)$ is essentially due to Blatz and Ko, ${ }^{5}$ who introduced eq. (28) for the $\lambda_{1}-\lambda_{2}$ relation following Smith's generalization of $\nu$.

Similar to eq. (19),

$$
\begin{equation*}
t^{22}=\left(\lambda_{2} / \lambda_{1} \lambda_{3}\right)\left[6 C\left(\lambda_{2}{ }^{2} I_{1}-I_{2}\right)+6 C \lambda_{1}{ }^{2} \lambda_{3}{ }^{2}\left(1-I_{3}^{-(1+\nu) /(1-2 \nu)}\right)\right] \tag{34}
\end{equation*}
$$

For infinitesimal deformation

$$
\begin{equation*}
t^{22} \simeq 36 C e_{2}+36 C[\nu /(1-2 \nu)] \vartheta \tag{35}
\end{equation*}
$$

which reduces to eq. (24) when

$$
\begin{equation*}
C=\mu / 18 \tag{36}
\end{equation*}
$$

Hence,

$$
\begin{equation*}
W=\frac{\mu}{18} I_{1}\left(I_{1}{ }^{2}-3 I_{2}\right)+\frac{\mu}{6}\left[I_{3}+\frac{1-2 \nu}{3 \nu} I_{3}^{-3 \nu /(1-2 \nu)}-\frac{1+\nu}{3 \nu}\right] \tag{37}
\end{equation*}
$$

It is easy to see that $W$ in eq. (37) satisfies eqs. (4) and (5) and that it does not possess a term representing the volume change in the form of eq. (17b); under pure dilatational deformation, it increases without limit as illustrated by the dashed curve in Figure $1 b$.

## A Possible Modification of the Mooney Strain Energy Function

The Mooney strain energy function [eq. (3)] is modified by writing it in the form

$$
\begin{equation*}
W=-C_{1}\left(I_{1}^{-1} I_{3}^{-\alpha^{\prime}}-3^{-1}\right)+C_{2}\left(I_{2} I_{3}^{-\beta^{\prime}}-3\right) \tag{38}
\end{equation*}
$$

so that it satisfies eq. (4) and takes the form of eq. (17b) for purely volumetric deformation with $I_{1}=3 v^{2 / 3}$ and $I_{2}=3 v^{4 / 3}$.

$$
\begin{equation*}
W^{*}=-\left(C_{1} / 3\right)\left(v^{-2\left(\alpha^{\prime}+1 / 3\right)}-1\right)+3 C_{2}\left(v^{-2\left(\beta^{\prime}-2 / 3\right)}-1\right) \tag{39}
\end{equation*}
$$

In eq. (39) an inequality $\alpha=m / 6=\alpha^{\prime}+1 / 3<\beta=n / 6=\beta^{\prime}-2 / 3$ has to be satisfied in order that $W^{*}$ attains a minimum value for equilibrium conditions.

Similar to eqs. (19) and (34),

$$
\begin{align*}
t^{22}=\left(\lambda_{2} / \lambda_{1} \lambda_{3}\right)\left[2 C_{1} I_{1}^{-2} I_{3}{ }^{\prime} \alpha^{\prime}\right. & +2 C_{2}\left(\lambda_{1}^{2}+\lambda_{3}^{2}\right) I_{3}^{-\beta^{\prime}} \\
& \left.+2 \lambda_{1}{ }^{2} \lambda_{2}^{2}\left(\alpha^{\prime} C_{1} I_{1}{ }^{-1} I_{3} \alpha^{\prime-1}-\beta^{\prime} C_{2} I_{2} I_{3}{ }^{-\beta^{\prime}-1}\right)\right] \tag{40}
\end{align*}
$$

which, for infinitesimal deformations, can be approximated by

$$
\begin{align*}
& t^{22} \simeq^{2} / 9\left(1+3 \alpha^{\prime}\right) C_{1}+2\left(2-3 \beta^{\prime}\right) C_{2} \\
& \quad+\left[-4 / 9 C_{1}\left(3 \alpha^{\prime 2}+2 \alpha^{\prime}+2 / 3\right)+4 C_{2}\left(3 \beta^{\prime 2}-4 \beta^{\prime}+1\right)\right] \vartheta \\
& \quad+\left[-4 / 3 \alpha^{\prime} C_{1}-4 C_{2}\left(1-3 \beta^{\prime}\right)\right] e_{2} \tag{41}
\end{align*}
$$

which has to be identical with eq. (24). Hence

$$
\begin{gather*}
1 / 9\left(1+3 \alpha^{\prime}\right) C_{1}+\left(2-3 \beta^{\prime}\right) C_{2}=0  \tag{42}\\
-2 / 9\left(3 \alpha^{\prime 2}+2 \alpha^{\prime}+2 / 3\right) C_{1}+2\left(3 \beta^{\prime 2}-4 \beta^{\prime}+1\right) C_{2}=\mu \nu /(1-2 \nu) \\
-2 / 3 \alpha^{\prime} C_{1}-2 C_{2}\left(1-3 \beta^{\prime}\right)=\mu \tag{43}
\end{gather*}
$$

From eqs. (42-44), one obtains

$$
\begin{align*}
& C_{1} / \mu=9\left(3 \beta^{\prime}-2\right) /\left(6 \alpha^{\prime}+6 \beta^{\prime}-2\right)  \tag{45}\\
& C_{2} / \mu=\left(3 \alpha^{\prime}+1\right) /\left(6 \alpha^{\prime}+6 \beta^{\prime}-2\right) \tag{46}
\end{align*}
$$

and

$$
\begin{align*}
& 3\left(3 \alpha^{\prime}+1\right) \beta^{\prime 2}-3\left[3 \alpha^{\prime 2}+6 \alpha^{\prime}+2+\nu /(1-2 \nu)\right] \beta^{\prime} \\
& \quad+\left\{6 \alpha^{\prime 2}+[7-3 \nu /(1-2 \nu)] \alpha^{\prime}+7 / 3+\nu /(1-2 \nu)\right\}=0 \tag{47}
\end{align*}
$$

From eqs. (45) and (46), two constants $C_{1}$ and $C_{2}$ can be determined in terms of the material constants $\mu, \alpha^{\prime}$, and $\beta^{\prime}$. Equation (47) gives a relation between $\alpha^{\prime}, \beta^{\prime}$, and Poisson ratio $\nu$.

With the aid of eqs. (42-44), it can be shown that $W^{*}$ in eq. (39) satisfies eq. (5).

The strain energy function of the form of eq. (38) with $C_{1}$ and $C_{2}$ given in eqs. (45) and (46) reduces to the following equation under (volume constant) infinitesimal deformation:

$$
\begin{equation*}
W=(\mu / 4)\left[(1+a) \vartheta_{2}^{\prime}+2(1-a) \vartheta_{2}\right] \tag{48}
\end{equation*}
$$

which can also be obtained from eq. (3), under the same condition where

$$
\begin{gathered}
\vartheta_{2}=e_{1}^{2}+e_{2}^{2}+e_{3}^{2} \\
\vartheta_{2}^{\prime}=\vartheta_{2}-\left(e_{1} e_{2}+e_{2} e_{3}+e_{3} e_{1}\right)
\end{gathered}
$$

and

$$
\begin{equation*}
a=\left(\beta^{\prime}-\alpha^{\prime}-1\right) /\left(\alpha^{\prime}+\beta^{\prime}-1 / 3\right)>0 \tag{49}
\end{equation*}
$$

If the material is intrinsically incompressible, there is no physical meaning for $\alpha^{\prime}$ and $\beta^{\prime}$ as discussed before.

## III. SIMPLE DEFORMATION AND EXPERIMENTAL RESULTS

For the three strain energy functions derived in the foregoing section, the relations between the axial stress $t^{11}$ and $\lambda_{1}$ as well as $\lambda_{1}$ and $\lambda_{2}$ are con-
sidered. The $\lambda_{1}-\lambda_{2}$ relations obtained from the energy functions are compared with the result of uniaxial tension tests performed on a dumbbellshaped specimen of a KCl-filled polyurethane (see Appendix).

## The Strain Energy Function, Eq. (26)

For uniaxial tension and compression, $\lambda_{2}=\lambda_{3}$ and the lateral stress $t^{22}=0$. Hence, from eq. (19) with $C, k, A \alpha$, and $B \beta$ given by eqs. (25) and (22a), one obtains the $\lambda_{1}-\lambda_{2}$ relation:

$$
\begin{align*}
& {[(1-2 \nu)(\beta-\alpha) /(1+\nu)] \lambda_{1}^{2 \beta} \lambda_{2}^{4+4 \beta}\left(\lambda_{2}{ }^{2}-\lambda_{1}{ }^{2}\right)} \\
&  \tag{50}\\
& +\lambda_{1}^{2(\beta-\alpha)} \lambda_{2}{ }^{4(\beta-\alpha)}-1=0
\end{align*}
$$



Fig. 2. Relations between axial and lateral stretches, $\lambda_{1}$ and $\lambda_{2}$, under uniaxial tension and compression for the strain energy functions given by eqs. (26), (37), and (38) with Poisson ratio $\nu=1 / 3$.

For the following numerical examples, it is assumed that $\alpha=1$ and $\beta=2$ ( $m=6$ and $n=12$ ); these are typical values for molecular bonds.

In Figure 2 , the $\lambda_{1}-\lambda_{2}$ relation is plotted as curve $I$ in the region $\lambda_{1}=0$ to 2.0 for $\nu=1 / 3$; in Figure 3, where the experimental results performed on dumbbell-shaped specimens are also shown, the $\lambda_{1}-\lambda_{2}$ relation is plotted in the region $\lambda_{1}=1.0-1.2$ for various values of $\nu$.

For simple tension and compression, the physical component of stress $\sigma_{11}$ is identical with $t^{11}=\lambda_{1} \lambda_{2}{ }^{-2} \partial W / \partial E_{11}$, and for the strain energy function eq. (26),


Fig. 3. Relations between axial and lateral stretches, $\lambda_{1}$ and $\lambda_{2}$, under uniaxial tension for the strain energy functions given by eqs. (26) and (37) with various values of Poisson ratio, and the result of experiment.

$$
\begin{align*}
& \frac{\sigma_{11}}{\mu}=\frac{\lambda_{1} \lambda_{2}-2}{3} \\
& \times\left\{\lambda_{1}{ }^{4}-\lambda_{2}{ }^{4}+\lambda_{2}{ }^{4}\left[\left(\lambda_{1}{ }^{2} \lambda_{2}{ }^{4}\right)^{-\alpha-1}-\left(\lambda_{1}{ }^{2} \lambda_{2}{ }^{4}\right)^{-\beta-1}\right] \frac{(1+\nu)}{(1-2 \nu)(\beta-\alpha)}\right\} \tag{51}
\end{align*}
$$

Using $\lambda_{1}$ and $\lambda_{2}$ which satisfy eq. (50), one can obtain $\sigma_{11}$ as a function of $\lambda_{1}$ from eq. (51). In particular, $\sigma_{11}$ for $\nu=1 / 3$ is shown by curve I in Figure 4.

The sharp drop in $\sigma_{11}$ in Figure 4 as $\lambda_{1}$ reaches approximately 1.4 may be due to the fact that the strain energy function cannot be valid when the critical separation distance $d^{\prime}$ between neighboring particles is attained; the irreversible separation occurs when $\lambda_{1}$ reaches approximately 1.4 under simple tension with $\nu=1 / 3, \alpha=1$, and $\beta=2$. Hence, in Figure 2, curve I is also not valid beyond $\lambda_{1}=1.4$.

When the material is incompressible, $\sigma_{11}$ is given ${ }^{7}$ by

$$
\begin{equation*}
\sigma_{11}=2\left(\lambda_{1}^{2}-\lambda_{1}^{-1}\right)\left(\partial W / \partial I_{1}+\lambda_{1}^{-1} \partial W / \partial I_{2}\right) \tag{52}
\end{equation*}
$$

When the first term of eq. (26) is considered as the strain energy function for incompressible media,

$$
\begin{equation*}
\sigma_{11} / \mu=1 / 3\left(\lambda_{1}^{2}-\lambda_{1}^{-1}\right)\left(\lambda_{1}^{4}+\lambda_{1}+\lambda_{1}^{-2}\right) \tag{53}
\end{equation*}
$$

and this is plotted as curve II in Figure 4.
For comparison, the $\sigma_{11}-\lambda_{1}$ relation for the Mooney strain energy function [eq. (3)]

$$
\begin{equation*}
\sigma_{11} / \mu=1 / 2\left(\lambda_{1}{ }^{2}-\lambda_{1}{ }^{-1}\right)\left[1+a+(1-a) \lambda_{1}{ }^{-1}\right] \tag{54}
\end{equation*}
$$



Fig. 4. Relations between axial stress and stretch, $\sigma_{11}$ and $\lambda_{1}$, under uniaxial tension and compression for the strain energy functions given by eqs. (26) and (37).
with the coefficient of asymmetry $a=0.233$, a value for a tread stock ${ }^{2}$ is also shown by curve III in Figure 4. It should be noted, however, that the Mooney function is more flexible than the first term of eq. (26) in the sense that it has two parameters $\mu$ and $a$ to adjust to experimental results.

## The Strain Energy Function, Eq. (37)

In this case, the $\lambda_{1}-\lambda_{2}$ relation under simple tension and compression is given by eq. (28), which is plotted for $\nu=1 / 3$ in Figure 2 (curve II). The same relation is also plotted in Figure 3 for $\nu=0.25,0.333,0.4$, and 0.5 .

With the aid of eq. (28), the uniaxial stress $\sigma_{11}=t^{11}=\lambda_{1} \lambda_{2}{ }^{-2} \partial W / \partial E_{11}$ for this strain energy function becomes

$$
\begin{equation*}
\sigma_{11} / \mu=1 / 3 \lambda_{1}^{5+2 \nu}\left(1-\lambda_{1}{ }^{-6(1+\nu)}\right) \tag{55}
\end{equation*}
$$

and the relation for $\nu=1 / 3$ is shown by curve IV in Figure 4 .

It is interesting to note that the strain energy functions given by eqs. (26) and (37) produce practically identical values $\sigma_{11}$ in the range of $\lambda_{1}$ where the strain energy eq. (26) is valid.

This strain energy function evidently reduces to the same form as eq. (26) does for incompressible media.

The Strain Energy Function Eq. (38), with $C_{1}$ and $C_{2}$ given by Eqs. (45) and (46)

In the same way as eq. (50) is derived, it can be shown that the $\lambda_{1}-\lambda_{2}$ relation under simple tension or compression for the strain energy function eq. (38) is

$$
\begin{align*}
\left(C_{1} / \mu\right)\left(\lambda_{1}{ }^{2} \lambda_{2}\right)^{\beta^{\prime}-\alpha^{\prime}}\left[\lambda_{2}{ }^{2}+\alpha^{\prime}\left(\lambda_{1}{ }^{2}\right.\right. & \left.\left.+2 \lambda_{2}{ }^{2}\right)\right]+\left(C_{2} / \mu\right) \lambda_{2}{ }^{2}\left(\lambda_{1}{ }^{2}+2 \lambda_{2}{ }^{2}\right)^{2} \\
& \times\left[\lambda_{1}{ }^{2}+\lambda_{2}{ }^{2}-\beta^{\prime}\left(2 \lambda_{1}{ }^{2}+\lambda_{2}{ }^{2}\right)\right]=0 \tag{56}
\end{align*}
$$

and the uniaxial stress $\sigma_{11}$ is

$$
\begin{gather*}
\sigma_{11} / \mu=\left(2 \lambda_{1} / \lambda_{2}{ }^{2}\right)\left\{\left(C_{1} / \mu\right)\left(\lambda_{1}{ }^{2}+2 \lambda_{2}{ }^{2}\right)^{-2}\left(\lambda_{1}{ }^{2} \lambda_{2}{ }^{4}\right)^{-\alpha^{\prime}}+2\left(C_{2} / \mu\right) \lambda_{2}{ }^{2}\left(\lambda_{1}{ }^{2} \lambda_{2}{ }^{4}\right)^{-\beta^{\prime}}\right. \\
+\lambda_{2}{ }^{4}\left[\alpha^{\prime}\left(C_{1} / \mu\right)\left(\lambda_{1}{ }^{2}+2 \lambda_{2}{ }^{2}\right)^{-1}\left(\lambda_{1}{ }^{2} \lambda_{2}{ }^{4}\right)^{-\alpha^{\prime}-1}\right. \\
 \tag{57}\\
\left.\left.\quad-\beta^{\prime}\left(C^{2} / \mu\right)\left(2 \lambda_{1}{ }^{2} \lambda_{2}{ }^{2}+\lambda_{2}{ }^{4}\right)\left(\lambda_{1}{ }^{2} \lambda_{2}{ }^{4}\right)^{-\beta^{\prime}-1}\right]\right\}
\end{gather*}
$$

where $C_{1} / \mu$ and $C_{2} / \mu$ are given in eqs. (45) and (46).
In the following examples, it is assumed that $\alpha^{\prime}=1 / 3$; hence $\alpha=1$ and $m=6$.

The $\lambda_{1}-\lambda_{2}$ relation for $\alpha^{\prime}=2 / 3, \beta^{\prime}=0.237$, and $\nu=1 / 3$ is plotted as curve III in Figure 2, while the same relation is plotted in Figure 5 for various values of $\nu$ together with the experimental results.


Fig. 5. Relations between axial and lateral stretches, $\lambda_{1}$ and $\lambda_{2}$, under uniaxial tension for the strain energy function given by eq. (38) with various values of Poisson ratio, and the result of experiment.


Fig. 6. Relations between axial stress and stretch, $\sigma_{11}$ and $\lambda_{1}$, under uniaxial tension and compression for the strain energy function given by eq. (38).

The $\sigma_{11}-\lambda_{1}$ relation for the same set of parameter values is obtained from eq. (57) by using $\lambda_{1}$ and $\lambda_{2}$ satisfying eq. (56) and shown as curve I in Figure 6.

Figure 6 seems to indicate that, under simple tension the critical distance $d^{\prime}$ between two neighboring particles is attained when $\lambda_{1}$ reaches approximately 1.4 while under simple compression the similar phenomenon is expected between two neighboring particles probably in the lateral direction when $\lambda_{1}$ reaches approximately 0.7 , although this is not observed for the material with the strain energy function eq. (26).

For incompressible media, eq. (38) reduces to

$$
\begin{equation*}
W=-C_{1}\left(I_{1}^{-1}-3^{-1}\right)+C_{2}\left(I_{2}-3\right) \tag{58}
\end{equation*}
$$

From eqs. (52) and (58),

$$
\begin{equation*}
\sigma_{11} / \mu=1 / 2\left(\lambda_{1}^{2}-\lambda_{1}^{-1}\right)\left[9(1+a)\left(\lambda_{1}^{2}+2 \lambda_{1}^{-1}\right)^{-2}+(1-a) \lambda_{1}^{-1}\right] \tag{59}
\end{equation*}
$$

Equation (59) produces the $\sigma_{11}-\lambda_{1}$ relation under the condition of incompressibility which is plotted for $a=0.233$ as curve II in Figure 6,
where the same relation based on the Mooney function is also shown as curve III.

## APPENDIX

## Tests on Dumbbell-Shaped Specimens

Uniaxial tension tests to failure were performed on the specimen type shown in Figure 7 with an Instron universal testing machine working at a crosshead rate of $0.05 \mathrm{in} . / \mathrm{min}$. Longitudinal extension ratio and midspecimen diameter were measured by means of the illustrated transducers. The results of five tests are presented in Figures 3 and 5.


Fig. 7. Test specimen.
The data show considerable scatter. However, taking the average response as a reasonable trend, a ratio of $\nu=0.33-0.40$ seems justified for the "instantaneous" values of the generalized Poisson ratio. The scatter of the data is due to the diameter reading being taken at one point only.

The authors are greally obliged to Dr. E. Lenoe, who performed the experiments reported in the Appendix.

## References

1. Eringen, A. C., Nonlinear Theory of Continuous Media, McGraw-Hill, New York, 1962.
2. Mooney, M., J. Appl. Phys., 11, 582 (1940).
3. Kittel, C., Introduction to Solid State Physics, Wiley, New York, 1953, p. 36.
4. Freudenthal, A. M., The Inelastic Behavior of Engineering Materials and Structures, Wiley, New York, 1950, p. 74.
5. Blatz, P. J., and W. L. Ko, Trans. Soc. Rheol., 6, 223 (1962).
6. Smith, T. L., Trans. Soc. Rheol., 3, 120 (1959).
7. Rivlin, R. S., Proceedings of the First Symposium on Naval Structural Mechanics, Pergamon Press, New York, 1960, p. 188.

## Résumé

Le but de cette recherche est d'étudier les formes possibles des fonctions d'énergie de la déformation élastique, qui reproduisent le comportement général des élastomères compressibles, isotopiques chargés tel que le montre l'expérience. On considère trois formes différentes de fonction d'énergie élastique de déformation, bien qu'elles puissent être classifiées en deux catégories essentiellement différentes: dans le premier cas, la fonction est supposé être la somme de l'énergie de déformation et de volume, tandis que dans le second cas elle représente une modification possible de la fonction d'énergie de déformation de Mooney. Pour ces trois fonctions d'énergie de déformation, on compare les relations entre les déformations axiales et latérales en extension uniaxiale avec le résultat des essais réalisés sur des échantillons de polyuréthann chargés avec du KCl expérimental. On rapporte également des courbes expérimentales donnant la relation entre la tension et la déformation pour l'extension uniaxiale et la compression.

## Zusammenfassung

In der vorliegenden Arbeit sollen die möglichen Formen der elastischen Verformungsenergiefunktionen untersucht werden, welche das grundsätzliche, experimentell beobachtbare Verhalten kompressibler, isotrop gefüllter Elastomerer wiedergeben. Es werden drei verschiedene Formen der Verformungsenergiefunktion in Betracht gezogen; die jedoch in zwei, wesentlich verschiedene Klassen eingeteilt werden können: im ersten Fall wird angenommen, dass die Funktion die Summe von Distortions- und Volumsverformungsenergie ist, während sie im zweiten eine mögliche Modifizierung der Verformungsenergiefunktion von Mooney darstellt. Für diese drei Verformungsenergiefunktionen wird die Beziehung zwischen axialer und seitlicher Verformung bei uniaxialer Spannung zusammen mit dem Ergebnis von Versuchen an hantelförmigen Proben eines KCl-gefüllten Polyurethans graphisch dargestellt. Ebenso wird die Beziehung zwischen axialer Spannung und Deformation bei uniaxialer Dehnung und Kompression graphisch dargestellt.
Received February 1, 1965


[^0]:    * This work was supported by the Office of Naval Research under Contract No. Nonr266(78).

