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THE INFLUENCE OF CHEMICAL STRUCTURE ON THE
STRENGTH OF RUBBER

by

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The detailed chemical structure of the polymer molecule has surprisingly little effect upon many of the important physical properties of crosslinked elastomers. For example, the elastic modulus, extensibility, tensile strength and tear strength are all much the same for many common elastomers. However, the strength of elastomers under some conditions is strikingly different. Two particular modes of fracture are considered here: tearing, and abrasive wear. Certain elastomers crystallize rapidly...
on stretching and become self-reinforcing, so that their tear strength is greatly enhanced. Factors that govern the speed of strain-induced crystallization are reviewed. In abrasive wear, the macroradicals generated by molecular rupture are highly reactive and their reactions affect both the nature and the extent of wear. The wear processes that occur in various reinforced elastomers are described and compared.
1. Introduction

Many of the physical properties of crosslinked elastomers do not depend directly upon the local chemical structure of the molecule at all. Instead, they depend upon other quantities, for example, the number \( V \) of molecular strands per unit volume, upon their contour length \( L \), and mass \( M_c \), and upon the local rate \( \phi \) of Brownian motion of molecular sub-units, consisting of small portions, about 5 main-chain atoms in length, of a molecular strand. (The actual number of main-chain atoms per molecular sub-unit is denoted \( q \) below; it is a measure of chain stiffness and hence it is somewhat smaller for more flexible molecules and larger for stiffer ones.) Some of these quantities are only indirectly related to the local molecular structure. As a result, many physical properties are found to be quite similar for elastomers that have markedly different chemical structure. For example, the tensile modulus of elasticity \( E \) for a network made up of flexible molecular strands is predicted by the statistical theory of rubber elasticity to be given by (1):

\[
E = 3\gamma kT
\]

where \( k \) is Boltzmann's constant, and \( T \) is the absolute temperature. Although the analysis leading to equation 1 takes into account the limited flexibility of elastomer molecules, this feature of their chemical structure does not appear in the final result. Thus, whether the individual molecules are relatively stiff or relatively flexible is unimportant in so far as the elastic modulus of the network is concerned.

Even the maximum extensibility of the network is only slightly dependent upon the molecular flexibility, over the range that might be expected for simple elastomeric polymers. It is principally determined by the molecular length \( L \), and hence molecular weight \( M_c \) between points of molecular interlinking (crosslinking) (1). It can be characterized by the ratio \( \lambda_m \) of the fully-stretched-out molecular length \( L \) to the average distance \( L_o \) between the ends of molecular strands in the unstretched state. The former quantity is given by

\[
L = n \ell
\]

where \( n \) is the number of molecular sub-units in a molecular strand and \( \ell \) is the length of a sub-unit. The latter quantity is given by

\[
L_o = n \ell
\]

if it is assumed that the sub-units are connected together by freely-rotating joints. Thus,

\[
\lambda_m = n \ell
\]

where \( n \) is related to the number \( n_e \) of main-chain atoms per molecular strand by

\[
n = n_e/q,
\]
to the molecular weight $M$ of a network strand by

$$n = \frac{M}{qM_0},$$

where $M_0$ is the molecular weight per main-chain atom, and to the number $\nu$ of network strands per unit volume by

$$n = \frac{A}{\nu qM_0},$$

where $A$ denotes Avogadro's number. Equations 4-7 show that $\lambda$ depends upon $\lambda^2$.

2. **Tear Strength of Non-Crystallizing Elastomers**

When elastomeric networks are torn apart under conditions of minimum strength, i.e., when no additional energy is expended in various dissipative processes (for example, viscous motion of molecular strands or detachment from filler particles), then the work $G_{c,o}$ of fracture per unit area torn through is given by (2)

$$G_{c,o} = \nu' n_0 U$$

where $\nu'$ is the number of strands crossing a randomly chosen plane of unit area (the fracture plane, for example) and $U$ is the dissociation energy of a main-chain bond. The value of $\nu'$ is directly related to the number $\nu$ of strands per unit volume and the average distance $L$ between their ends (2):

$$\nu' = \left(\frac{3}{8}\right)^{1/2} \nu L$$

Thus, from equations 3, 7, 8 and 9:

$$G_{c,o} = (3/8)^{1/2} \nu L qM_0^{1/2} / M_0$$

In terms of Young's modulus $E$, from equations 1, 6 and 7:

$$G_{c,o} = (9/8)^{1/2} (OA)^{1/2} (qkT)^{1/2} \nu L M_0^{-1/2} E^{3/2}$$

Equation 1 indicates, and experiments confirm, that the modulus of elasticity $E$ depends primarily upon the number of network strands and not upon their detailed structure. On the other hand, equations 10 and 11 show that the tear strength depends significantly upon the mass $M_0$ per main-chain atom, as well as upon the number of strands and hence $E$. Experimental measurements of tear strength under threshold conditions, i.e., at high temperatures and low rates of tearing, are in good agreement with these theoretical predictions, as shown in Figures 1 and 2 (3). Values of the work of fracture $G_c$ are found to increase in proportion to $M_c^{1/2}$, and to decrease in proportion to $E^{-1/2}$, for networks prepared by crosslinking to different degrees. And, for the same values of $M_c$ or of $E$, substantial differences are found between different polymers, those with larger values of mass $M_0$ per main-chain atom having lower tear strengths, as low as $1/5$ of the tear strength of the simple hydrocarbon elastomers.
Fig. 1. Threshold tear strength $G_{c,0}$ vs. molecular weight $M_c$ of network strands (3). 1, polybutadiene, $M_c=13.5$ a.m.u., ($\Delta$); 2, cis-polyisoprene, $M_c=17$ a.m.u., (0); 3, trans-polyisoprene, $M_c=17$ a.m.u., (●); 4, polydimethyl siloxane, $M_c=37$ a.m.u., (□); 5, phosphonitrilic fluoroelastomer, $M_c=185$ a.m.u., (■).

Fig. 2. Threshold tear strength $G_{c,0}$ vs. Young's modulus $E$ (3). Symbols as in Figure 1.
Under normal conditions the tear strength is many times larger than the small threshold value $G_{c,0}$, about 50-100 J/m$^2$, because of energy expended in various dissipative processes. For simple viscoelastic materials the tear strength is governed by the local viscosity, i.e., by the rate $\dot{\phi}$ of Brownian motion of molecular sub-units. In turn, $\dot{\phi}$ is directly related to the temperature difference $T-T_g$, where $T$ is the test temperature and $T_g$ is the glass transition temperature of the elastomer (4):

$$\log_{10}(\dot{\phi}_T/\dot{\phi}_g) = 17.6(T-T_g)/(52+T-T_g)$$

(12)

where $\dot{\phi}_T$ denotes the rate of sub-unit motion at $T$, about 0.1 jumps/sec. Using a scaling factor $\alpha_T = \dot{\phi}_T/\dot{\phi}_g$ for the rate of tearing, measurements of tear strength for several elastomers at many temperatures can be superimposed to give a master curve for tear strength as a function of the effective rate of tearing at $T_g$, Figure 3. This demonstrates that the tear strength depends only upon $T-T_g$, and not upon the local chemical structure of the elastomer except insofar as that determines the value of $T_g$ (5).

Fig. 3. Tear strength $G_c$ plotted against the effective rate of tearing at $T_g$ (5). Results are shown for six butadiene-styrene (SBR) and butadiene-acrylonitrile (NBR) elastomers, with $T_g$ ranging from -30°C to -80°C.
Far above $T_a$, under threshold conditions, the tear strength depends significantly upon the molecular structure as discussed earlier, and there is some evidence that the same relative differences are maintained under non-equilibrium conditions. But the primary variable for determining the tear strength is $T - T_a$.

The question now arises; which fracture processes, if any, are strongly affected by the local chemical structure? Two examples are considered below: tearing and crack growth, and abrasive wear. Under certain conditions these failure processes are found to depend upon particular features of the elastomer molecule and they are therefore distinctly different, even for closely-related chemical structures. Natural rubber can usefully be compared with cis 1, 4-polybutadiene in this respect, because, although their chemical structures are superficially similar, large differences are observed in their resistance to tearing and in the mechanism of wear.

3: Tearing and Crack Growth in Strain-Crystallizing Elastomers

Certain elastomers, notably natural rubber, crystallize on being stretched by several hundred per cent. They become much stiffer, and rather inelastic due to delays in crystallization and in melting on release. At a crack tip, rubber is highly stressed even when the overall strain is relatively small. The loss of energy associated with crystallization and, later, melting in this region leads to enhanced tear strength at low rates and high temperatures (6), and a much improved resistance to crack growth under repeated stressing (7), as shown in Figures 4, 5 and 6, in comparison with a non-crystallizing elastomer. Strain-induced crystallization is thus a specific, and highly desirable, feature of elastomers. The physical and chemical factors responsible for it are discussed below.

![Graph](image)

**Fig. 4.** Tear strength $G_c$ of natural rubber as a function of test temperature $T$ and rate $R$ of tearing (6).
Fig. 5. Fatigue life $N$ of natural rubber (NR) and a butadiene-styrene rubber (SBR) plotted against the test temperature $T$ (7).

Fig. 6. Fatigue life $N$ of natural rubber (NR) and a butadiene-styrene rubber (SBR) plotted against the depth $l_0$ of an initial edge crack (7).
4. Strain-Induced Crystallization

The phenomenon of rapid crystallization in the strained state and rapid melting on release can be attributed to three main causes:

(i) In the unstrained state the crystal melting temperature $T_m$ lies below ambient temperature and the material is therefore non-crystalline.

(ii) On stretching, the melting temperature is raised markedly, to values well above ambient, so that crystallization is thermodynamically favored and the free energy change on fusion is large.

(iii) The glass transition temperature $T_g$ is quite low, well below ambient. Molecular sub-units are consequently highly mobile at ambient temperature and are able to enter the crystalline state rapidly when the free energy change is favorable.

Many polymers have low values of $T_g$ and therefore satisfy condition (iii). However, many of them have either relatively large latent heats of fusion $h$ or, more commonly (8), relatively small latent entropies of fusion $s$, so that their melting temperatures $T_m$, given by

$$T_m = h/s,$$

lie above ambient temperature. They are therefore normally crystalline in the unstrained state. Some common examples are: polyethylene, polyethylene oxide, trans 1, 4-polybutadiene and trans 1, 4-polyisoprene (see Table 1). These materials do not satisfy condition (i).

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<th>Polymers</th>
<th>$T_m$ (°C)</th>
<th>$h$ (kJ/kg)</th>
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<td>141</td>
<td>280</td>
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<tr>
<td>Polyethylene oxide</td>
<td>66</td>
<td>200</td>
</tr>
<tr>
<td>Trans 1, 4-polyisoprene</td>
<td>74</td>
<td>190</td>
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<td>Trans 1, 4-polybutadiene</td>
<td>148</td>
<td>187</td>
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<tr>
<td>Cis 1, 4-polybutadiene</td>
<td>6</td>
<td>163</td>
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<tr>
<td>Trans 1, 4-polychloroprene</td>
<td>80</td>
<td>95</td>
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<tr>
<td>Cis 1, 4-polyisoprene</td>
<td>30</td>
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Table 1: Melting temperatures $T_m$ and latent heats of fusion $h$ for some representative crystallizing polymers.

a Taken from reference 8 and "Physical Constants of Linear Homopolymers", by O. G. Lewis, Springer-Verlag, New York 1968.
Of the remaining elastomeric materials, some will meet condition (ii) more successfully than others. The reasons for this can be readily deduced from Flory's approximate theoretical treatment for the melting temperature \( T_{m,\lambda} \) of crystallites in a molecular network held at a stretch ratio \( \lambda \) (9). A molecular sub-unit entering a crystallite from a strand in a stretched molecular network undergoes a smaller loss of configurational entropy than from the unstretched state because its configurational entropy has already been lowered somewhat by stretching. The reduction \( \Delta s \) in the entropy of fusion can be evaluated from the statistics of deformed and undeformed molecular networks. The result leads, by means of equation 13, to a predicted increase in the melting temperature on stretching (9):

\[
\left( \frac{h Q_0}{R} \right) \left( T_{m}^{-1} - T_{m,\lambda}^{-1} \right) = \frac{6}{n} \frac{1}{\lambda} - \frac{1}{2n} \left( \lambda^2 + 2\lambda^{-1} \right)
\]

where \( h \) is the latent heat of fusion per gram, \( R \) is the gas constant, and \( \lambda \) is the tensile stretch ratio applied to the network. This relation is found to give reasonably satisfactory predictions of the melting temperatures \( T_{m,\lambda} \) at moderate extensions, in the range 100 to 400 per cent (\( \lambda = 2 \) to 5) and for different degrees of crosslinking, represented by different values of the molecular strand length \( n \). Some typical results are shown in Figures 7 and 8 (10,11).

Both calculated and observed increases in melting temperature on stretching are found to be larger for some elastomers than for others, and for natural rubber the effect is largest of all. The reason for this lies in the unusually small value of the latent heat of fusion \( h \) for cis-polyisoprene, Table 1. As equation 14 indicates, the increase in \( T_{m} \) on stretching is inversely related to \( h \). Thus, the smaller the value of \( h \) the greater will be the tendency to exhibit strain-induced crystallization. An abnormally low value of \( h \) for natural rubber appears to be associated in part with the relatively small change in density that accompanies crystallization and in part with the absence of strong interatomic binding in the unit cell. Whatever the exact cause, the low value of \( h \) is clearly responsible for the facility with which natural rubber crystallizes on stretching.

5. Abrasive Wear

Wear of rubber under sliding conditions resembles small-scale tearing (12). Indeed, it has been treated as cumulative tearing – a mechanical fatigue process – taking place under the repeated action of frictional forces. A quantitative relationship has been derived in this way for the rate of wear in terms of the rate of crack growth under repeated stressing (13,14). When the rubber is rather tough and wear resistant, however, there is evidence of chemical deterioration during sliding, in addition
Fig. 7. Melting temperatures for crosslinked trans 1, 4-polychloroprene, held at various stretch ratios $\lambda$ and crystallized at various temperatures $T_c$ (10). Theoretical relation from equation 14 for increase in $T_m$ with $\lambda$.

Fig. 8. Melting temperatures for crosslinked trans 1, 4-polyisoprene, held at various stretch ratios $\lambda$ (11). Theoretical relation from equation 14 for increase in $T_m$ with $\lambda$. 
to tearing (15,16). When this deterioration is extensive the rubber and abrasion surface become covered with an oily decomposition product and the tearing process is altered, if not stopped altogether.

An example of a particle of wear debris torn from a rather weak material, an unfilled butadiene-styrene (SBR) vulcanizate, is shown in Figure 9. It has characteristically rough, torn surfaces. In contrast, the particle shown in Figure 10, obtained from a carbon-black-filled SBR vulcanizate, has a smooth, shiny appearance and the surface is sticky, as if covered with an oily or tarry film. The debris from carbon-black-filled natural rubber vulcanizates is even more highly degraded, so that the individual particles can hardly be distinguished in this case. On the other hand, the debris from carbon-black-filled cis-1,4-polybutadiene materials is finely-divided and particulate showing no signs of decomposition and every indication of having been mechanically torn away from the rubber surface. Thus, the wear process for reinforced elastomeric materials of roughly equal hardness and friction coefficient, and of comparable tear strength and tensile strength, differs strikingly in character from one polymer to another. These differences must be ascribed to different chemical features of the elastomers.

It should be pointed out at this stage that the formation of an oily degraded surface layer is not necessarily a beneficial feature. If the layer is readily removed from the rubber, then further deterioration can proceed rapidly. Indeed, if in the early stages of decomposition, the rubber is rendered softer and weaker, it will be torn away more easily and the rate of wear will be correspondingly greater than in the absence of general molecular scission. On the other hand, if the liquidlike film is viscous, tarry, and adhesive, it appears to be retained on the rubber surface to act as a protective layer. The rate of wear is then much reduced.
Fig. 9. Photograph of wear debris from an unfilled SBR vulcanizate.
Fig. 10. Photograph of wear debris from a carbon-black-filled SBR vulcanizate.
In order to account for the formation of a degraded surface film in some instances, a number of possible chemical processes can be hypothesized:

(i) Thermal decomposition, as a result of frictional heating.

(ii) Oxidative scission of the molecular network, possibly accelerated by frictional heating.

(iii) Mechanical rupture of the molecular network, followed by internal and external reactions of the polymer radicals generated in this way.

The first process need not be considered further here, because all of these elastomers are more or less equally susceptible to thermal decomposition whereas they do not all degrade during sliding. The second process is also probably not the main mechanism of decomposition because some elastomers show frictional decomposition even in inert atmospheres (15). The third process, however, does appear to be the basic mechanism of molecular decomposition during sliding. A rather good correlation is found to hold between the degree of decomposition observed during frictional sliding, both in air and in an inert atmosphere, and corresponding changes in molecular weight when the original elastomer (before crosslinking) is subjected to continuous mechanical shearing (15,16). For example, polybutadiene forms rather reactive macroradicals by molecular scission, and then undergoes crosslinking reactions, so that both in the shearing of the uncrosslinked polymer and the frictional sliding of the reinforced and crosslinked polymer, the product of mechanochemical processes tends to become a crosslinked solid. In contrast, natural rubber forms a resonance-stabilized macroradical by molecular fracture, which, in the presence of oxygen, forms a peroxy radical and then a hydroperoxide by H abstraction so that the original chain fracture is rendered permanent. Indeed, subsequent oxidation steps may cause scission of other chains as well. Thus, the product of mechanical rupture of the molecular network in this case tends to become liquidlike rather than solid.

These considerations account for the formation of a viscous liquid film on certain materials, and not on others, during frictional sliding. Moreover, the properties of the film, its viscosity and adhesiveness, will clearly depend upon the detailed reactions initiated by mechanical rupture of the elastomer molecules. They will therefore differ from one elastomer to another and they will also depend upon the particular ingredients used in the rubber mix formulation, especially when these substances are themselves able to participate in free-radical reactions.

Many of the differences encountered in the wear behavior of practical rubber compounds can thus be accounted for in a qualitative way when the specific chemical process involved in wear is recognized.
Acknowledgements

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