

## Stiffness of Molecular Chain of Synthetic Rubber

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### Synopsis

The principal polarizabilities of rubber molecules including copolymers were evaluated by the arithmetic sum of the polarizabilities of the components. The mean polarizabilities thus calculated are in good accordance with those from the refractive indices. The anisotropic polarizabilities of polymeric units for several kinds of rubbers were calculated and compared with those estimated from stress-optical coefficients. Length or molecular weight of a statistical link thus estimated is used as a measure of stiffness of rubber molecules. There is a good accordance in statistical links between anisotropic and viscosity measurements in theta solvent. The molecular weight or length of a statistical link thus obtained is found to be compatible with the value calculated from end-to-end distance in theta solvent. The ultimate elongation of rubber is correlated with the stiffness and can be estimated from the measure of stiffness of the chain.

### Introduction

Several kinds of synthetic rubbers have been industrially developed. They have many uses according to their chemical and physical properties, which are associated with their chemical structures. The stiffness of the molecular chain may be one of the important factors governing its rubber elasticity, ultimate elongation, and dynamic properties.

Attempts to evaluate the stiffness by stress birefringence measurements were made by Kuhn and Grün,<sup>1</sup> Treloar,<sup>2</sup> and other investigators<sup>3,4</sup> for natural rubber and polychloroprene. They measured the stress-optical coefficient and compared it with the calculated anisotropy of the polymer unit to evaluate the length of the equivalent link as stiffness.<sup>5-8</sup> Other polymers such as polyisobutene, polyethylene, and polystyrene were investigated by several authors in a similar way.<sup>9-22</sup> However, data on other kinds of rubbers were left uncollected, except for polybutadiene.

This report deals with the anisotropy of natural rubber, emulsion polybutadiene, *cis*-polybutadiene, lithium polybutadiene, styrene-butadiene rubber (SBR), nitrile rubber, ethylene-propylene rubber (EPR), butyl rubber (IIR), polyisobutene, as well as polyethylene and polystyrene at elevated

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temperatures. Length or molecular weight of statistical links thus obtained is used for a measure of stiffness of the rubber molecule, which is correlated with the physical properties.

### Calculated Polarizabilities of Polymer Units

In conformity with the calculations of Treloar<sup>5</sup> for the polarizabilities of isoprene unit, we carried out the calculations of three polarizabilities of the

TABLE I  
Polarizabilities of Chemical Bonds

Bond	$\alpha_1, 10^{-25} \text{ cm}^3$	$\alpha_2, 10^{-25} \text{ cm}^3$
C—C	18.8	0.2
C—H	7.9	5.8
C=C	28.6	10.6
C <sub>ar</sub> =C <sub>ar</sub>	22.5	4.8
C—Cl	36.7	20.8

TABLE II. Calculated Polarizabilities of Polymeric Units of Rubbers<sup>a</sup>

Polymer	$\alpha_{11}$	$\alpha_{22}$	$\alpha_{11} - \alpha_{22}$		$\bar{\alpha}$
			lit.	this paper	
Natural rubber	105.3	85.3	17.8 <sup>6</sup> 30.8 <sup>6</sup> 21.3 <sup>36</sup>	21.8	94.1
Emulsion polybutadiene	93.0	65.7	—	27.4	74.7
<i>cis</i> -Polybutadiene	95.7	64.2	—	31.5	74.7
Li polybutadiene	97.1	65.6	—	31.5	75.3
1,2-Polybutadiene	70.8	76.8	—	-6.0	74.8
<i>trans</i> -Polybutadiene	98.7	62.9	—	35.8	74.8
Polyisobutene	77.6	78.6	-1.4 <sup>20</sup>	-1.0	78.3
SBR 1502	96.4	80.1	—	16.3	85.5
Polychloroprene	113.7	84.8	—	28.7	94.4
Nitrile rubber (35% AN)	81.0	63.0	—	18.0	69.0
EPR (50% ethylene)	55.7	44.9	—	10.8	48.5
Polypropylene	63.0	55.8	—	7.2	58.2
Polyethylene	48.4	34.0	1.3 <sup>36</sup>	14.4	38.8
Poly(vinyl chloride)	63.4	55.9	5.0 <sup>3</sup>	7.5	58.4
			8.9 <sup>3</sup>		
Polyacrylonitrile					
A	56.8	59.0	—	-2.2	58.3
B	58.6	58.1	—	0.5	58.3
Polystyrene (perpendicular)	118.2	170.7	-62.8 -6.2 <sup>37</sup>	-52.5	153.9
Polystyrene	169.1	146.3	-6.2	22.8	153.9

<sup>a</sup> In units of  $10^{-25} \text{ cm}^3$ .

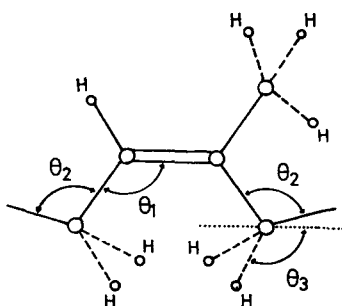


Fig. 1. Structure of polymeric unit of polyisoprene:  $\theta_1 = 125^\circ$ ,  $\theta_2 = 109^\circ 30'$ ,  $\theta_3 = 109^\circ 45'$ .

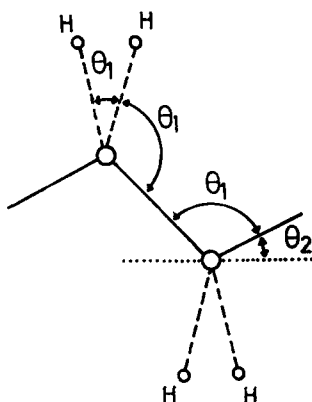


Fig. 2. Structure of polymeric unit of polyethylene:  $\theta_1 = 109^\circ 30'$ ,  $\theta_2 = 35^\circ 15'$ .

polymeric unit,  $\alpha_{11}$ ,  $\alpha_{22}$ , and  $\alpha_{33}$  referring respectively to the direction of the chain length, the transverse direction in the plane of the molecule, and the direction normal to the plane, by using the following equations:

$$\alpha_{11} = \Sigma \alpha_{xx}, \alpha_{22} = \Sigma \alpha_{yy}, \alpha_{33} = \Sigma \alpha_{zz} \quad (1)$$

$$\left. \begin{aligned} \alpha_{xx} &= (\alpha_1 - \alpha_2) \cos^2 \theta + \alpha_2 \\ \alpha_{yy} &= (\alpha_1 - \alpha_2) \sin^2 \theta \cos^2 \phi + \alpha_2 \\ \alpha_{zz} &= (\alpha_1 - \alpha_2) \sin^2 \theta \sin^2 \phi + \alpha_2 \end{aligned} \right\} \quad (2)$$

Here,  $\theta$  and  $\phi$  are pertinent angles in polar coordinate systems, and  $\alpha_1$  and  $\alpha_2$  are the parallel and transverse polarizabilities of a chemical bond, respectively. According to Denbigh<sup>23</sup> the bond polarizabilities were taken to be as in Table I.

The polarizabilities of polymer units are calculated by summing the principal polarizabilities  $\alpha_{xx}$ ,  $\alpha_{yy}$ , and  $\alpha_{zz}$  of the bonds composing the polymer unit, by taking the x-axis to be the direction along polymer chain whose structures are shown in Figures 1 to 9.

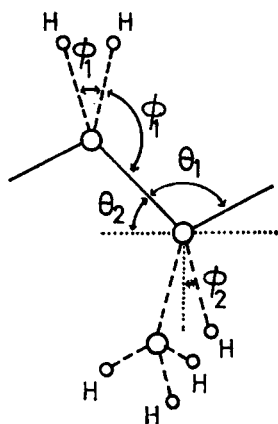


Fig. 3. Structure of polymeric unit of polypropylene:  $\theta_1 = 109^\circ 30'$ ,  $\theta_2 = 35^\circ 15'$ ,  $\phi_1 = 109^\circ 30'$ ,  $\phi_2 = 54^\circ 45'$ .

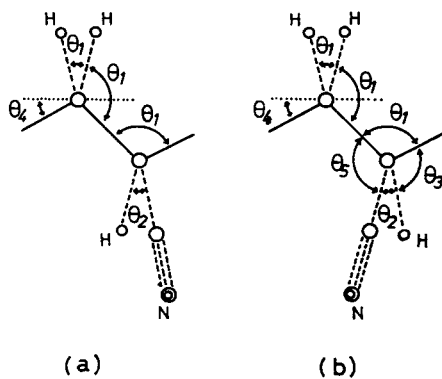


Fig. 4. Structure of polymeric unit of polyacrylonitrile: (a), (b)  $\theta_1 = 109^\circ 30'$ ,  $\theta_2 = 122^\circ$ ,  $\theta_3 = 122^\circ$ ,  $\theta_4 = 35^\circ 15'$ ,  $\theta_5 = 122^\circ 36'$ .

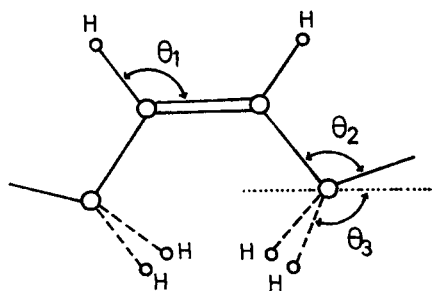


Fig. 5. Structure of polymeric unit of *cis*-polybutadiene:  $\theta_1 = 125^\circ$ ,  $\theta_2 = 109^\circ 30'$ ,  $\theta_3 = 109^\circ 45'$ .

In the copolymers such as SBR, nitrile rubber, EPR, and butyl rubber, the polarizabilities of the polymeric units are calculated as the arithmetic mean of each component according to their compositions, which were determined by chemical analysis. In emulsion polybutadiene, the microstructures were determined by infrared analysis to be 71% *trans*-, 19% vinyl,

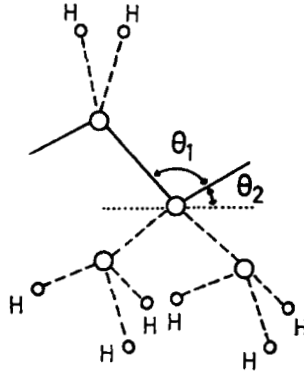


Fig. 6. Structure of polymeric unit of polyisobutene:  $\theta_1 = 109^\circ 30'$ ,  $\theta_2 = 35^\circ 15'$ .

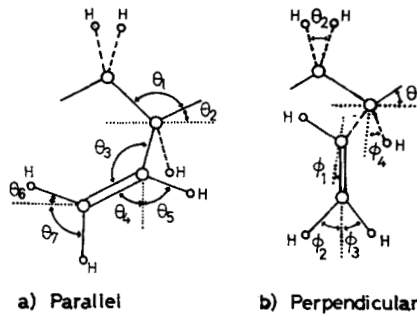


Fig. 7. Structure of polymeric unit of poly-1,2-butadiene: (a) parallel,  $\theta_1 = 109^\circ 30'$ ,  $\theta_2 = 35^\circ 15'$ ,  $\theta_3 = 125^\circ$ ,  $\theta_4 = 55^\circ$ ,  $\theta_5 = 70^\circ$ ,  $\theta_6 = 20^\circ$ ,  $\theta_7 = 90^\circ$ ; (b) perpendicular,  $\theta_1 = 35^\circ 15'$ ,  $\theta_2 = 109^\circ 30'$ ,  $\phi_1 = 15'$ ,  $\phi_2 = 54^\circ 45'$ ,  $\phi_3 = 35^\circ 15'$ ,  $\phi_4 = 54^\circ 45'$ .

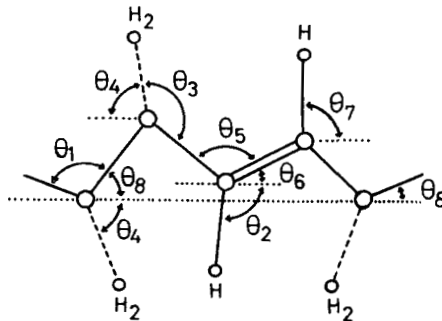


Fig. 8. Structure of polymeric unit of *trans*-polybutadiene:  $\theta_1 = 109^\circ 30'$ ,  $\theta_2 = 83^\circ 45'$ ,  $\theta_3 = 125^\circ 15'$ ,  $\theta_4 = 83^\circ 45'$ ,  $\theta_5 = 122^\circ$ ,  $\theta_6 = 29^\circ$ ,  $\theta_7 = 87^\circ$ ,  $\theta_8 = 41^\circ 30'$ .

and 10% *cis*-configurations. Almost the same structure was estimated for polybutadiene units in SBR and nitrile rubber. The polarizabilities of polymer units thus calculated are summarized in Table II.

The mean polarizabilities,  $\bar{\alpha}$ , are also calculated as follows:

$$\bar{\alpha} = (\alpha_{11} + \alpha_{22} + \alpha_{33})/3 \quad (3)$$

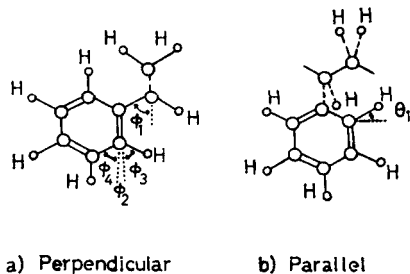


Fig. 9. Structure of polymeric unit of polystyrene: (a) perpendicular,  $\theta_1 = 90^\circ$ ,  $\phi_1 = 54^\circ 45'$ ,  $\phi_2 = 5^\circ 15'$ ,  $\phi_3 = 65^\circ 15'$ ,  $\phi_4 = 54^\circ 45'$ ; (b) perpendicular,  $\theta_1 = 30^\circ$ ,  $\theta_2 = 54^\circ 45'$ .

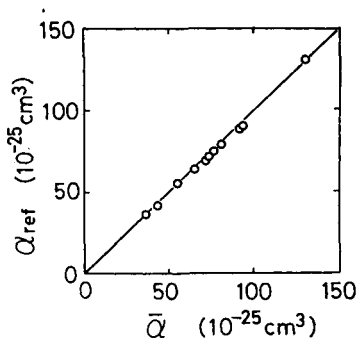


Fig. 10. Comparison of the mean polarizabilities calculated from refractive indices,  $\alpha_{ref}$ , and from bond polarizabilities,  $\bar{\alpha}$ .

As shown in Figure 10, the values listed in Table III were found to be in good accordance with experimental values given by the Lorenz-Lorentz equation concerning the refractive index,  $n_0$ :

$$\frac{n_0^2 - 1}{n_0^2 + 2} \frac{M}{d} = \frac{4\pi}{3} \bar{\alpha} N \quad (4)$$

where  $M$ ,  $d$ , and  $N$  are molecular weight of polymer unit, density of polymer, and Avogadro's number, respectively. This fact may be taken as support for the correctness of the unit polarizabilities thus calculated.

TABLE III  
Mean Polarizabilities  $\bar{\alpha}$  and  $\bar{\alpha}_{\text{ref}}$  of Polymers

Polymer	Refractive index $n_0$	$M$ , g/mole	$d$ , g/cm <sup>3</sup>	$\bar{\alpha}_{\text{ref}}$ , <sup>a</sup> 10 <sup>-25</sup> cm <sup>3</sup>	$\bar{\alpha}$ , <sup>b</sup> 10 <sup>-25</sup> cm <sup>3</sup>
Natural rubber	1.517	68.0	0.912	90.7	94.1
<i>cis</i> -Polybutadiene	1.526	54.0	0.915	72.0	74.7
Li polybutadiene	1.516	54.0	0.913	71.2	75.3
Emulsion polybutadiene	1.518	54.0	0.913	71.4	74.7
Polysobutene	1.528	56.0	0.910	76.4	78.3
SBR 1502	1.531	60.9	0.935	80.5	85.5
Polychloroprene	1.558	88.5	1.15	91.7	94.4
Nitrile rubber (35% AN)	1.527	53.7	0.985	66.4	69.0
EPR (50% ethylene)	1.479	33.6	0.877	43.4	48.5
Polypropylene	1.528	42.0	0.905	57.1	58.2
Polyethylene	1.521	28.0	0.920	36.8	38.8
Poly(vinyl chloride)	1.540	62.6	1.39	52.6	58.4
Polystyrene	1.595	104.0	1.04	135.	153.9
Butyl rubber	1.509	56.5	0.913	73.7	78.3
Polyacrylonitrile	—	53.0	1.17	—	58.3

<sup>a</sup> Calculated from eq. (3).

<sup>b</sup> Calculated from the refractive index.

### Stress Birefringence and Anisotropy of Rubber

According to Kuhn and Gr $\ddot{u}$ n's theory, birefringence,  $\Delta n$ , is expressed as a function of strain ratio,  $\lambda$ , as follows<sup>1,24</sup>:

$$\Delta n = \frac{2\pi}{45} \frac{(n_0^2 + 2)^2}{n_0} V (\alpha_{11} - \alpha_{22}) [\lambda^2 - (1/\lambda)] \quad (5)$$

where  $V$  is the number of chains per unit volume and  $n_0$  is the mean refractive index of the sample. Since stress,  $S$ , is also a function of the strain ratio,  $\lambda$ , so that

$$S = VRT [\lambda^2 - (1/\lambda)], \quad (6)$$

it follows that

$$\frac{\Delta n}{S} = \frac{2\pi}{45kT} \frac{(n_0^2 + 2)^2}{n_0} (\alpha_{11} - \alpha_{22})_{\text{seg}}. \quad (7)$$

Equation (7) was derived for ideal rubber. In actual cases, a more complicated Mooney-Rivlin equation is to be used<sup>25,26</sup> instead of eq. (6):

$$S = 2C_1 [\lambda^2 - (1/\lambda)] + 2C_2 [\lambda - (1/\lambda^2)] \quad (8)$$

which involves terms containing coefficients  $C_1$  and  $C_2$ . By changing the temperature of measurement and the crosslinking density, values ranging from about 0.3 to 0.8 were obtained for  $C_2/(C_1 + C_2)$ . In Figure 11, an experimental result for sulfur-cured polyisoprene is shown together with the ratio of the anisotropic polarizability of a segment to that of the monomeric unit,  $Z$ . To eliminate the effect of the  $C_2$  term,  $\Delta n/S$  was plotted against  $C_2/(C_1 + C_2)$  and its value was extrapolated to  $C_2/(C_1 + C_2) = 0$ .

The stress birefringence equation, eq. (7), does not involve the concentration of crosslinks but an experimental anisotropic polarizability,  $(\alpha_{11} - \alpha_{22})_{\text{seg}}$ , which is concerned with the statistically equivalent link. Comparing this value with the calculated polarizability of the polymer unit, the number of polymeric units to one statistical unit or its length will be obtained. The results thus obtained are shown in Table IV.

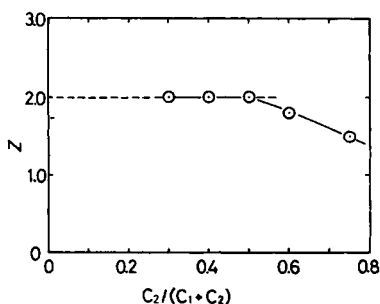


Fig. 11. The ratio of the anisotropic polarizability of a segment to that of the monomeric unit,  $Z$  and  $C_2/(C_1 + C_2)$ .



TABLE IV  
Anisotropic Polarizabilities  $(\alpha_{11} - \alpha_{22})_{\text{seg}}$  and  
 $(\alpha_{11} - \alpha_{22})_{\text{mono}}$  for Monomeric Units, and their ratios  $Z$

Polymer	$\Delta n/S,$ $10^{-10}$ $\text{cm}^2/\text{dyne}$	$(\alpha_{11} - \alpha_{22})_{\text{seg}}^{\text{a}}$ $10^{-25} \text{ cm}^3$	$(\alpha_{11} - \alpha_{22})_{\text{mono}}^{\text{b}}$ $10^{-25} \text{ cm}^3$	$Z$
Natural rubber	2.00	47.7	21.8	2.09
Emulsion polybutadiene	2.22	52.8	27.4	1.92
<i>cis</i> -Polybutadiene	2.88	68.0	31.5	2.16
Li polybutadiene	3.08	73.3	31.5	2.33
SBR 1502	1.86	43.8	16.3	2.69
Nitrile rubber				
(25% AN)	2.63	62.6	20.7	3.03
(35% AN)	2.63	62.6	18.0	3.04
EPR (50% ethylene)	1.84	44.0	10.8	4.07
Polyethylene (120°C)	2.3	74.3	14.4	5.16
Butyl rubber	1.10	26.2	2.1	12.5
Polychloroprene	2.58	58.0	28.9	2.01

<sup>a</sup> Observed.

<sup>b</sup> Calculated.

### Relation Between Length of a Statistical Link and Mechanical Properties of Rubbers

From the theoretical viewpoint, solution viscosity in theta solvent provides a mean square end-to-end distance of the molecular chain,  $\langle R^2 \rangle$ , corresponding to  $nl^2$ . The ratio,  $\sigma^2 = \langle R^2 \rangle / nl^2$ , is given from the viscosity measurements in the literature, as shown in Table V. It was found that

TABLE V  
Measure of Stiffness  $\sigma^2 l$ ,<sup>a</sup> and  $Zl$ <sup>b</sup> (Å)

Polymer	$\sigma^{2c}$	$l$	$\sigma^2 l^{\text{a}}$	$Zl^{\text{b}}$
Natural rubber	3.02	4.8	14.5	10.0
<i>cis</i> -Polybutadiene	2.25	4.8	10.8	10.4
Emulsion polybutadiene	1.85	4.35	8.1	8.4
SBR 1502 <sup>d</sup>	2.65	4.2	11.1	11.3
Polychloroprene	1.96	4.9	9.6	9.8
Polyisobutene	3.24	2.5	8.1	62.5
Nitrile rubber	4.40	4.0	17.6	12.1

<sup>a</sup> Estimated from viscosity.

<sup>b</sup> Estimated from optical data.

<sup>c</sup> These values are adopted from the data summarized in Brandrup and Immergut.<sup>38</sup>

<sup>d</sup> Calculated from Homma and Fujita.<sup>39</sup>

the length of the equivalent statistical link,  $\sigma^2 l$ , obtained from viscosity data in the literature agrees roughly with that obtained from anisotropic data,  $Zl$ , as shown in Table V. Here,  $Z$  is the ratio of the anisotropic polarizability of a statistical segment to that of a monomeric unit and  $l$  is the length of the unit.

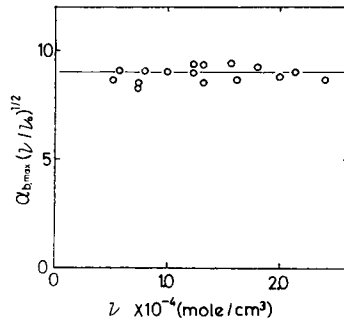


Fig. 12. Reduced plots of maximum extension ratio,  $(\alpha_{b,\max})(V/V_0)^{1/2}$ :  $V_0 = 1.0 \times 10^{-4}$  mole/cm<sup>3</sup>.

According to Flory<sup>27</sup> and others, the ultimate elongation,  $\alpha_{b,\max}$ , is a function of the number of segments between crosslinks,  $n$ :

$$\alpha_{b,\max} = n^{1/2}. \quad (9)$$

Taking the molecular weight of the branch to be negligible, the number of segments will be correlated with the molecular weight between crosslinks,  $Mc$ , as follows:

$$n = Mc/m' = \zeta/Vm' \quad (10)$$

where  $m'$  is the molecular weight of the statistical link and  $\zeta$  is the density of the polymer. The molecular weight of the statistical link obtained physically will be linearly correlated with that from optical measurements.

Ultimate elongation is affected by temperature and elongation rate. At a temperature  $T_{\max}$ , the maximum value was observed, which was found to fit the above eqs. (9) and (10) concerning the degree of vulcanization on natural rubber, polybutadiene rubber, SBR, NBR, EPR, and others. From these plots, the molecular weight of the statistical link,  $m'$ , was evaluated and found to be close to the value  $m$  from anisotropy, with a correction factor:

$$\alpha_{b,\max} = 1.24 (Mc/m)^{1/2} \quad (11)$$

In Figure 12,  $(\alpha_{b,\max})_0$  of nitrile rubber was plotted against the concentration of the crosschain,  $V$ :

$$(\alpha_{b,\max})_0 = (\alpha_{b,\max})(V/V_0)^{1/2} \quad (12)$$

where  $\alpha_{b,\max}$  is the maximum elongation ratio and  $V_0$  was taken to be  $1.0 \times 10^{-4}$  mole/cc in nonfilled rubber. The reduced elongation ratio,  $(\alpha_{b,\max})_0$  was plotted against  $(Mc/m)^{1/2}$  for several kinds of rubbers in Figure 13, where  $Mc$  was determined from equilibrium swelling and  $m$  was estimated from stress birefringence.

These figures indicate that the maximum elongations of rubbers can be estimated from the stress-birefringential data or a measure of stiffness,

$ZM$ , except for butyl rubber. Here  $Z$  is the ratio of anisotropic polarizability of a statistical segment to that of the monomeric unit and  $M$  is the molecular weight of the monomeric unit. In eq. (11),  $m$  was taken to be

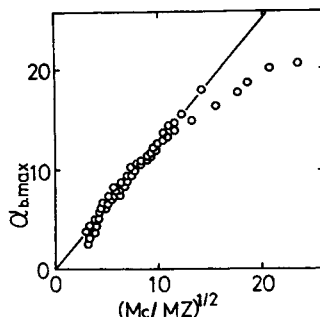


Fig. 13. Maximum extension ratio,  $\alpha_{b,\max}$ , and values estimated from stress birefringence data,  $(M_c/MZ)^{1/2}$ .

$ZM$ . In butyl rubber, the calculated anisotropy of a polymeric unit seems to be somewhat small. The estimated value of  $Z$  from the maximum elongation ratio is 3.5 to 3.7.

## EXPERIMENTAL

### Curing Systems and Conditions

Several kinds of rubber were compounded with the ingredients shown in Table VI on open rolls at temperatures from 50° to 120°C. The transparent samples were used to measure the optical coefficients and two coefficients in the Mooney-Revin equation. The samples cured with sulfur-accelerator systems were subjected to the test of extensibilities at temperatures from -100° to 160°C.

### Concentration of Crosschain

Samples, 0.1 to 0.2 cm in thickness, 1 cm in width, and 2 cm in length, were swelled in solvent at 30°C for 48 hr and weighed, followed by drying under vacuum. The weight and density of the dried samples were measured to calculate the concentration of crosschain according to Flory and Rehner's equation<sup>29,30</sup>:

$$V = \frac{v_r + \mu v_r^2 + \ln(1 - v_r)}{v_r/2 - v_r^{1/3}} \frac{1}{V_s} \quad (13)$$

where  $v_r$  and  $V_s$  are, respectively, volume fraction of rubber in swollen rubber and molar volume of the solvent. Coefficients of polymer-solvent interaction,  $\mu$ , are shown in Table VII.

TABLE VI  
Recipe for the Preparation of Sample

Curing ingredients, <sup>a</sup> phr	Polymer <sup>b</sup>											
	NR	IR	BR	SBR	EPR	IR	BR	SBR	EPR	NBR	IIR	IIR
Zinc oxide	3	3	3	3	1	—	—	—	—	—	5	1-10
Stearic acid	2	2	2	2	1	—	—	—	—	—	1	1
Antioxidant 2246	1	1	1	1	1	—	—	—	—	—	—	—
Sulfur	2	0.5-3	0.5-2.5	0.5-2.5	0.5-2.5	—	—	—	—	—	1.5	1-1.5
Accelerator CZ	—	0.5-3	0.5-2.5	0.5-2.5	0.5-2.5	—	—	—	—	—	1-1.5	1-1.5
Dicumyl peroxide	—	—	—	—	—	0.3-3	0.2-1	0.2-1.5	0.3-3	—	—	—
Accelerator M	0.5	—	—	—	—	—	—	—	—	—	—	1
Accelerator TT	—	—	—	—	—	—	—	—	—	—	—	1
Accelerator H	0.5	—	—	—	—	—	—	—	—	—	—	—

<sup>a</sup> 2246, 2,2'-Methylene bis(4-methyl-6-tert-butyl phenol); CZ, N-cyclohexyl-2-benzothiazyl sulfonamide; M, 2-mercaptobenzothiazole; TT, tetramethylthiuram disulfide; H, hexamethylenetetramine.

<sup>b</sup> NR, RSS #3; IR, Cariflex 305, Natsyn 400; BR, BR #III, Diene 35NF; SBR, JSR 1502; EPR, EPsyn 70A, Royalene 305; NBR, JSR 230S; IIR, Esso butyl 268.

TABLE VII  
Polymer-Solvent Interaction Parameter  $\mu$

Polymer	$\mu$	Solvent	Reference
<i>cis</i> -Polybutadiene	0.39	benzene	31
<i>cis</i> -Polyisoprene	0.44	benzene	32
EPR	$0.49 + 0.33v_r$	benzene	33
SBR	$0.37 + 0.27v_r$	benzene	34
IIR	$0.46 + 0.26v_r$	<i>n</i> -heptane	34
NBR	0.207	chloroform	35

### Stress-Birefringential Measurements

Samples of 0.1 to 0.2 cm in thickness, 5 cm in length, and 1.0 cm in width were extended at a rate of 20 mm/min or 2 mm/min. The revealing birefringence of rubber was measured in the rubbery state according to Kolsky and Crawford's method.<sup>28</sup> Stress-strain curves were observed at the same time by Shimazu's stress-strain recorders.

### Elongation Ratio

Ring samples of 1.00 cm in diameter and 0.10 cm in width were cut out from the sheet of about 0.1 cm in thickness. The ring samples were elongated at a speed of 50 mm/min in order to obtain the maximum elongation at temperatures from  $-100^\circ$  to  $160^\circ\text{C}$ . The maximum elongation at each temperature was adopted as the characteristic value.

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