Technical Report No. 113

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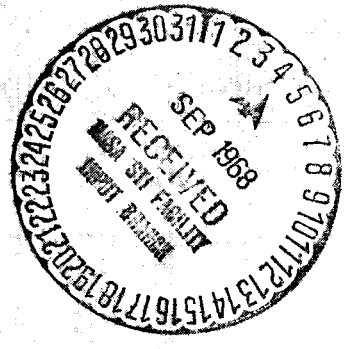

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Behavior of Elastic Networks of Various Degrees of Orientation in the Kinetic Theory of Fracture*

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This paper describes a kinetic theory of fracture using a network of linear elastic elements as a model to calculate the strength and elastic properties of oriented materials. Emphasis has been placed on the questions as to whether the assumptions of small strains as well as the invariant molecular orientation distribution are valid for the total fracture process. During the fracture process the breakage of molecular elements reduces the modulus of elasticity. However, it was found to be less than one per cent for most of the lifetime of a sample.

For any network systems of different degrees of orientation the calculated curves of the logarithm of time to break versus moderate applied stress are linear, but of different slopes. The slopes are inversely proportional to the modulus of elasticity of the network at zero time. Therefore, if the calculated curves of the logarithm of time are plotted versus the initial strain (the applied stress divided by the initial modulus of elasticity), the linear portions of all curves reduce to one. For very small or very large stresses the curves deviate from linearity.

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**Present address: California Institute of Technology

INTRODUCTION

The important problem of brittle fracture of solids has been investigated by many authors experimentally as well as analytically. For a large group of materials the time and temperature dependency of the fracture strength indicated the presence of thermal activation processes during fracture development[1-4]. Especially for high polymer solids it was found that the energy of activation had a value comparable to the binding energy of primary chemical bonds[4]. It was thus suggested that the breakage of primary bonds in the chain molecules actually takes place. Furthermore, this phenomenon is considered to be the dominant cause of the eventual failure of these solids.

With this knowledge in mind it seems desirable to obtain some information as to how the continued breakage of individual molecular chains influence the mechanical behavior of the remaining chain network. As a result of the chain breaking processes, the state of stress in a microscopic surrounding of unbroken chains, the macroscopic strains and the orientation distribution of the molecules in space will change. This change in turn will affect the rate of rupture of other molecular chains. In any high polymer solid the thermal motion of segments or side groups leads to microbrownian motion and contributes toward changing strains and molecular orientation distribution. Since the conditions of small strains and invariant orientation distribution are necessary or desirable for a number of theoretical

considerations, it is of interest to show whether these conditions are fulfilled for a network with no slip of chain segments, as creep due to slip is not taken into account in the present calculations. The time dependent mechanical behavior of such a model network for the case of uniaxial stressing is studied. Instructive results such as the time-to-break and the value of strain as a function of time are obtained. Changes in the relative orientation distribution of the network chains are also found.

THE MODEL

A model very suitable for calculations of the behavior of chain networks has already been developed earlier[5]. It was considered that on a macroscopic scale polymer solids are homogeneous so that continuum theory of elasticity may be applied to relate prescribed forces and deformations to the state of strain of a small subvolume. Within the subvolume the presence of molecular chains and their orientation is taken into account. In this model the load carrying chain molecules are represented by linear elastic elements. A condition of homogeneous strain* is assumed for any sufficiently small subvolume. Consequently the force F, acting upon each of the individual elements within this subvolume is determined by the orientation and the elastic constant κ_i of the element together with the state of strain ε_{mn} of the subvolume as follows:

*A discussion of the consequences following from this and alternative assumptions is given in reference 6.

$$F(\theta,t) = \kappa \ell_o \varepsilon_{mn} s_m s_n \tag{1}$$

where ℓ_o is the length of any of the individual elements and s_m, s_n are components of the unit vector in the direction of orientation identified by θ and ϕ ($s_1 = \sin\theta \cos\phi$, $s_2 = \sin\theta \sin\phi$, $s_3 = \cos\theta$ in spherical coordinates). If for convenience we replace forces by stresses, we obtain:

$$\psi(\theta,\phi,t) = E\varepsilon_{mn} s_m s_n \tag{2}$$

where $E = \kappa \ell_o^2 \lambda$, if λ is the number of elements per unit volume. This general scheme is roughly represented in Fig. 1 with various possible orientations of the molecular network in a material body under a prescribed load, P. We may repeat that the state of stress of an individual element is given by (2). The stress at any particular point within the macroscopic body shall be given as an average of all of the stress contributions of the molecular elements within the subvolume around this point. As shown previously[5] the macroscopic stresses, σ_{ij}, can be calculated from (2) once the orientation distribution function, ρ, is known. Thus a basic constitutive equation describing the mechanical behavior of a molecular solid is obtained. For a brittle solid ε_{mn} is regarded to be small.

It might be mentioned that the kinetic nature of fracture was introduced into this theory by assuming that for any element the given probability of breakage is dependent upon the local stress $\psi(\theta,\phi,t)$. This assumption has led to a system of integro-differential

equations which have been solved successfully for only special cases of prescribed macroscopic stresses and molecular orientation[7,8]. In these cases the time-to-break of the solid had been calculated as a function of the prescribed load. Now, these calculations will be extended to samples of random molecular orientation using a numerical iteration method. This method also yields information on the variation of the modulus of elasticity, ratio of transverse and normal strains, as well as the molecular orientation distribution.

NUMERICAL ITERATIVE SOLUTION OF CONSTITUTIVE EQUATIONS

As can be shown[5] the macroscopic stress tensor σ_{ij} can be calculated as functions of $\psi(\theta,\phi,t)$, ρ, ε_{mn} and the relative number of unbroken elements, $f(\theta,\phi,t)$ to give:

$$\sigma_{ij} = \oint \rho f s_i s_j \psi d\Omega \tag{3}$$

where $d\Omega$ is the infinitesimal solid angle. For the case of uniaxial tension with hexagonal or transverse symmetry with respect to the 33-axis as the direction of orientation of the elements we obtain:

$$\sigma_{11} = \sigma_{22} = 0 = \int_0^{\pi/2} E\rho(\theta)\, f(\theta,t)[\varepsilon_{11}\sin^2\theta + \varepsilon_{33}\cos^2\theta]\sin^3\theta d\theta \tag{4}$$

$$\sigma_{33} = 2\pi\int_0^{\pi/2} E\rho(\theta)\, f(\theta,t)[\varepsilon_{11}\sin^2\theta + \varepsilon_{33}\cos^2\theta]\cos^2\theta\, \sin\theta d\theta \tag{5}$$

Here the strain ratio $\nu = \nu_{3311} = - \varepsilon_{11}/\varepsilon_{33}$ may be used to eliminate ε_{11}.

For all elements with "orientation θ" the rate of rupturing of the unbroken elements is given by the modified activation energy

$$df/dt = - fK_b \tag{6}$$

where

$$K_b = \omega_b \exp[-U/RT + \beta\psi(\theta,t)] \ ^* \tag{7}$$

ω_b and β are constants, R is the gas constant, T absolute temperature and U the activation energy.

The time-dependent quantities $\varepsilon_{33}(t)$, $\nu(t)$ and $f(\theta,t)$ must be found so as to satisfy equations (4) - (7). Numerical results are obtained using an iteration method.

The principle of this method is to determine $\psi(\theta,t_1)$ at time t_1 from $\nu(t_1)$ and $\varepsilon_{33}(t_1)$. With $\psi(\theta,t_1)$ and $f(\theta,t_1)$ known, $(df/dt)_{t_1}$ can be determined. The iteration process then makes use of the approximation:

$$f(\theta,t_2) = f(\theta,t_1) + (df/dt)_{t_1} \Delta t \tag{8}$$

where Δt is an arbitrarily small time, and the calculation of ψ, ν, and ε_{33} is repeated for $t = t_2$ with $f = f(\theta,t_2)$.

In the case of uniaxial stress applied to a randomly

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*The formulation of equation (7) implies that all the load carrying elements have the same potential energy barrier U, i.e., any initial stresses of the unloaded specimen are neglected. The formulation is valid if the variation of U/RT is small compared with $\beta\psi$.

oriented specimen ($\rho = 1/2\pi$) the successive steps of the iteration process are as follows.

At first the change of f is determined.

$$\Delta f_1 = \Delta f(\theta,t_1) = - \bar{\omega}_b \Delta t \cdot f(\theta,t_1) \exp[6\beta\sigma_o \frac{\varepsilon_{33}(t_1)}{\varepsilon_{33}(t_o)} (\cos^2\theta - \nu(t_1)\sin^2\theta)] \quad (9)$$

where $\bar{\omega}_b \equiv \omega_b \exp(-U/RT)$ and $\sigma_o = \sigma_{33} = \text{const.}$

As a second step $f_2 = f(\theta,t_2)$ is calculated from (8) and (9) where Δt may be chosen arbitrarily but small. In order to obtain sufficient accuracy with reasonable computing times $\bar{\omega}_b \Delta t$ was chosen to be $D\exp(-6\beta\sigma_o)$ where D is a number which varies between 0.002 and 0.04.

The third step is the calculation of $\nu(t_2)$ from (4):

$$\nu(t_2) = \int_0^{\pi/2} f_2 \cos^2\theta \sin^3\theta d\theta \Big/ \int_0^{\pi/2} f_2 \sin^5\theta d\theta \quad (10)$$

Finally $\varepsilon_{33}(t_2)/\varepsilon_{33}(t_o)$ is calculated from (5):

$$\varepsilon_{33}(t_2)/\varepsilon_{33}(0) = 1/6 \int_0^{\pi/2} f_2[\cos^2\theta - \nu \sin^2\theta]\cos^2\theta \sin\theta d\theta \quad (11)$$

Then a new cycle may be started with the values obtained from equation (9) - (11). In order to keep the error inflicted by the approximation small (< 5%) D must be decreased every few cycles. The iteration process is terminated once the total number of unbroken elements is reduced and thus the local stress $\psi(0,t)$ increased so

much that catastrophic failure must occur (arbitrarily a value of $6\beta\sigma_o\varepsilon_{33}(t)/\varepsilon_{33}(t_o) - 6\beta\sigma_o = 40$ with a rate of $\Delta f/f = -0.002\, e^{40}$ was chosen to mark the point of catastrophic failure).

RESULTS AND DISCUSSION

Stress ψ as a function of $\cos\theta$

The presence of kinetic fracture processes in a stressed network of elastic elements leads to a continuous decrease of the number of unbroken elements and consequently to an increase in local stress ψ. In Fig. 2, ψ in a uniaxially stressed specimen is shown as a function of $\cos\theta$ (orientation of elements) for a value of $\beta\sigma_o = 0.5$*. It is seen that the shape of the stress distribution is not changed much during most of the "life-time" of the specimen. The angle $\theta_o(t)$ for which $\psi(t) = 0$ changes only very slowly with time. In this figure as well as in the next four figures time is measured in units of $\text{TIME} = \bar{\omega}_b t \exp(6\beta\sigma_o)$. In this way all lifetimes are in the neighborhood of unity for ease of comparison. As can be seen from Fig. 2 the elements oriented in the direction of uniaxial stress have to carry most of the load and will break with the highest rate. The dashed line in the figure indicates that at TIME = 0.808 there are no unbroken elements of such orientation.

Orientation distribution

The high rate of fracture of elements in the direction identified by $\cos\theta = 1$ leads to a pronounced decrease of

*We will discuss the results and draw the figures in terms of $\beta\psi$ or $\beta\sigma$ rather than ψ or σ_o alone because the rate of fracture depends upon $\beta\sigma_o$. This way the results are valid for different materials and different temperatures.

the number of unbroken elements in that direction while the number in other directions decreases slowly. The change in the distribution of elements thus resulted is shown for those values of TIME at which f begins to reach 0.5 and 0. It is seen in Fig. 3 that in the case of a high stress level ($\beta\sigma_o = 25/6$) fracture within a small solid angle of orientation proceeds so much faster than elsewhere that even at catastrophic failure only a small number of elements is seen to be affected. It is also seen that once f comes close to zero the catastrophic failure follows immediately. At a reduced level of stress ($\beta\sigma_o = 3/6$) the proceedings in the central section are more moderate and a larger region is noticably affected; but again, there is only a small difference in TIME (~ 0.043) from where the number of unbroken elements reaches zero to produce the catastrophic failure.

Figure 4 shows the changes in the number of unbroken elements for a very small stress ($\beta\sigma_o = 0.1/6$). In this case the rate of breakage of unbroken elements is practically independent of stress and consequently of orientation. This rate is solely determined by $\overline{\omega}_b = \omega_b \exp(-U/RT)$. For polymeric solids in their glassy state it is known to be extremely small. For various reasons the predicted values as shown in Fig. 4 may not be very significant. As indicated earlier the scatter in the potential barrier U can be neglected only if the term $\beta\sigma_o$ is much larger than the average deviation of U/RT from the mean value. This will not be the case if $\beta\sigma_o$ is too small. Another reason may be the occurrence of reformation processes which will have the most effect at small stresses and even lead to infinite lifetimes at finite stresses[8].

Strain as a function of TIME

To maintain a constant stress σ_o with a decreasing

number of elements the strains in the unbroken elements have to increase. In other words: an elastic network with the above described properties will show additional deformation. In Fig. 5 the change of $\varepsilon_{33}(t)$ is plotted against TIME. It is seen that ε_{33} increases almost linearly at first. After a period of accelerated increments the point of catastrophic failure is well defined where the rate of change in ε_{33} approaches infinity. It may be noted that in terms of TIME even infinitely large stresses lead to a finite time for catastrophic failure. Numerical calculations of breaking TIME with $\beta\sigma_o$ up to 100/6 have shown that this limit is $TIME_b^\infty = 0.622$. An analytical verification of this value is difficult and not yet available. In the appendix it is shown, however, that a limit exists which is larger than 5/12 or $TIME_b^\infty > 5/12$.

Strain ratio as a function of TIME

As a consequence of the different decay rates for elements of different orientations the relative distribution of the elements in the elastic network changes and so does the strain ratio. The variation of ν as used in (10) with TIME is shown in Fig. 6. For very small stresses there is almost no change of ν due to the fact that the rate of breakage of unbroken elements is practically independent of direction. For very high stresses the breaking of elements is concentrated near the direction of applied uniaxial stress, affecting only very few elements and again ν does not change much. If, however, $\beta\sigma_o$ is near unity then the relative distribution of broken and unbroken elements undergoes the greatest change and so does ν. In all cases during catastrophic failure ν tends toward zero.

Lifetime as a function of stress

For each level of stress the point of catastrophic failure determines the lifetime of the specimen. In Fig. 7 for different specimens and different assumptions the calculated time to break is shown as a function of uniaxial stress $\beta\sigma_o$. Curve I refers to a completely oriented specimen (all elements in the direction of stress). This curve has already been discussed earlier[8]. At higher stress values this curve is almost a straight line with a slope of $-1 - 1/\beta\sigma_o$. For small values of $\beta\sigma_o$ Curve I approaches infinity. Curve III refers to a randomly oriented specimen ($\rho = 1/2\pi$). For small values of $\beta\sigma_o$ both curves approach each other. For large values Curve III forms a straight line and the inclination can be calculated in the same way as for Curve I only in this case $\sigma_o/\varepsilon_{33}(t_o) = E/6$ and we obtain $d\ln\bar{\omega}_b t_b/d(\beta\sigma_o) = -6 - 1/\beta\sigma_o$. Accordingly for any intermediate orientation the slope of the lifetime curve would be given by $d\ln\bar{\omega}_b t_b/d(\beta\sigma_o) = -E/E(\varepsilon) - 1/\beta\sigma_o$ where $E(\varepsilon)$ is the initial modulus of elasticity of the oriented sample (orientational strain ε). If $(\beta\sigma_o)^{-1}$ is neglected, then this expression can be written as $d\ln\bar{\omega}_b t_b/[d(\beta\sigma_o)/E(\varepsilon)]$. This means that by suitably reducing the abscissa with $1/E(\varepsilon)$ the lifetimes to fracture for specimen of different degrees of orientation would fall on one line.

Fracture data for oriented material (PAN, Polyacrylonitrile)were recently published by Regel and Leksovsky[9]. These data are shown in Fig. 8. If the assumptions made in deriving (12) are correct, then a simple reduction of the abscissa with $E/E(\varepsilon)$ would make the 4 curves in Fig. 8 fall together to one. This is indeed the case if $E/E(\varepsilon)$ values are suitably chosen. The values used have not yet been compared with direct measurements of

the initial modulus of elasticity of PAN, but they are compared with measurements made on Nylon 66 and with theoretical values (Fig. 9). The change of modulus which has to be assumed for PAN turns out to be very close to the theoretical prediction[6]. To obtain Curves I and III it was assumed that the rate of fracture is correctly given by (7).

Since it is difficult to imagine how linear elements may break under a compressive load without shear, the calculations were repeated using the assumption that $df/dt = 0$ for $\psi \leq 0$. It was found, however, that even for small values of $\beta\sigma_o (\sim 1/12)$ the change in lifetime as a result of using this assumption was less than 3%. For moderate and high stresses no difference was found. If an assumption that the rate of breakage is determinable by:

$$df/dt = - \bar{\omega}_b f \sinh (\beta\psi) \qquad (12)$$

rather than by an exponential law, then the assumption of $df/dt = 0$ for $\psi \leq 0$ does not lead to any unsteadiness of df/dt for $\psi = 0$. The time to break calculated with a rate proportional to $\sinh (\beta\psi)$ is also shown as Curve II in Fig. 7 for comparison.

Partially oriented specimen and time-dependent loading

In previous calculations only a completely oriented specimen or a totally unoriented specimen ($\rho = 1/2\pi$) has been treated. Since a numerical method is employed there will be no difficulty whatsoever to substitute ρ by any orientation distribution function and repeat the calculations. For example, in the case of uniaxial

orientation through a homogeneous plastic deformation the function

$$\rho(\theta) = \frac{1}{2\pi} \frac{(1+\varepsilon)^3}{[\cos^2\theta + (1+\varepsilon)^3 \sin^2\theta]^{3/2}}$$

may be used, where ε is the plastic orientational strain. The results will lie between the values obtained for a randomly oriented system and a completely oriented system.

For a time dependent rather than a constant stress, an analytical solution of equations (6) and (7) is available for several special loading conditions. In most cases a twofold approximation will become necessary. At first the time dependent stress will have to be approximated by a constant state of stress within a certain interval of time. Secondly, the continuous change of the number of unbroken elements will be approximated by a stepwise change as in this report.

CONCLUSIONS

The following conclusions may be drawn which also answer the questions raised at the beginning.

a) In uniaxial stressing, only elements within a relatively small solid angle are affected and the size of this angle changes little during 90% of the lifetime (Figs. 2 and 3).

b) The additional strain developed during stressing and thus leading into new orientation distributions of the elements can be calculated from Fig. 5. The general coordinate $\varepsilon_{33}(t)\beta E$ can be transformed into the strain coordinate of one experiment by

specifying β and E. For organic glass β is 2.27 x 10^{-2} cm^2/kp^8 and the modulus of elasticity of a completely oriented material can be estimated to be six times that of the unoriented material of 12,000 kp/cm^2. The general ordinate 10 would correspond in the particular experiment to a strain of $10/(2.27 \times 10^{-2} \times 6 \times 12 \times 10^3) = 0.6\%$. Thus, it seems that the additional strain for most of the lifetime is smaller than 1%. Once the time approaches the point of catastrophic failure the strain increases rapidly so that the orientation distribution of the unbroken elements will change. By that time, however, the fracture process has entered the macroscopic stage and an entirely new formulation of the problem may be needed for further study.

c) The stress-lifetime curves of uniaxially stressed materials of different degree of orientation have linear slopes which are approximately inversely proportional to $E(\varepsilon)$. Thus, if the abscissa is reduced by a factor of $E(\varepsilon)$ all curves will fall together at values of $\beta\sigma_o > 0.4$. Recently published data of the fracture of PAN seem not to disagree with this prediction.

APPENDIX

Lower limit of TIME-to-break for infinitely high stresses

If $TIME = \bar{\omega}_b t \exp(6\beta\sigma_o)$ is used as time scale, then the calculated TIME-to-break for high stresses tends towards a limit of $TIME_b^{\infty} = 0.622$. It may be shown in this Appendix that a limit exists which is larger than 5/12. Let us abbreviate $\varepsilon_{33}(t_n)/\varepsilon_{33}(0)$ by A_n. Thus (11) can be given as:

$$A_n^{-1} = 6\int_0^{\pi/2} f_n(\cos^2\theta - \nu\sin^2\theta)\cos^2\theta \, \sin\theta d\theta \qquad (A1)$$

For very high stresses it has been shown that $\nu(t) = \nu = 0.25$ is constant throughout most of the lifetime. Using this and also (8) and (9) we may rewrite (A1) as:

$$A_n^{-1} = A_{n-1}^{-1} \left\| 6\int_0^{1} f_{n-1} D(\cos^2\theta - 0.25\sin^2\theta)\cos^2\theta \right. \exp[\beta(\psi_{n-1} - 6\sigma_o)]d(\cos\theta) \qquad (A2)$$

If $\cos\theta$ is replaced by $y = 1 - \cos\theta$ and if for small y the term y^2 is neglected we get:

$$A_n^{-1} = A_{n-1}^{-1} - 6\exp[B(A_{n-1}^{-1} - 1)]\int_0^{1} f_{n-1} D(1 - \tfrac{9}{2} y)\exp(-\tfrac{5}{2} yBA_{n-1})dy \qquad (A3)$$

where $B \equiv 6\beta\sigma_o$

If we replace $f_{n-1} < f_o = 1$ by 1 and solve the integral, we obtain:

$$A_n^{-1} > A_{n-1}^{-1} - D\exp[6\beta\sigma_o(A_{n-1}-1)]/2.5\beta\sigma_o A_{n-1} \qquad (A4)$$

and

$$\frac{A_n - A_{n-1}}{A_{n-1}} < \frac{D\exp[6\beta\sigma_o(A_{n-1}-1)]}{2.5\beta\sigma_o} \qquad (A5)$$

Since $D = d(TIME)$ and $(A_n - A_{n-1})/A_{n-1} = dA/A$ we have

$$\int d(TIME) > \int_1^\infty 2.5\frac{\beta\sigma_o dA}{A}\exp[-6\beta\sigma_o(A - 1)]$$

$$= \frac{2.5}{6}6\beta\sigma_o\exp(6\beta\sigma_o)\int_1^\infty \frac{\exp(-6\beta\sigma_o A)}{A}dA \qquad (A6)$$

The integral term is well known and may be approximated by $\exp(-6\beta\sigma_o)/6\beta\sigma_o$. Thus

$$\int d(TIME) = TIME_b^\infty > \frac{2.5}{6} = 5/12 \qquad (A7)$$

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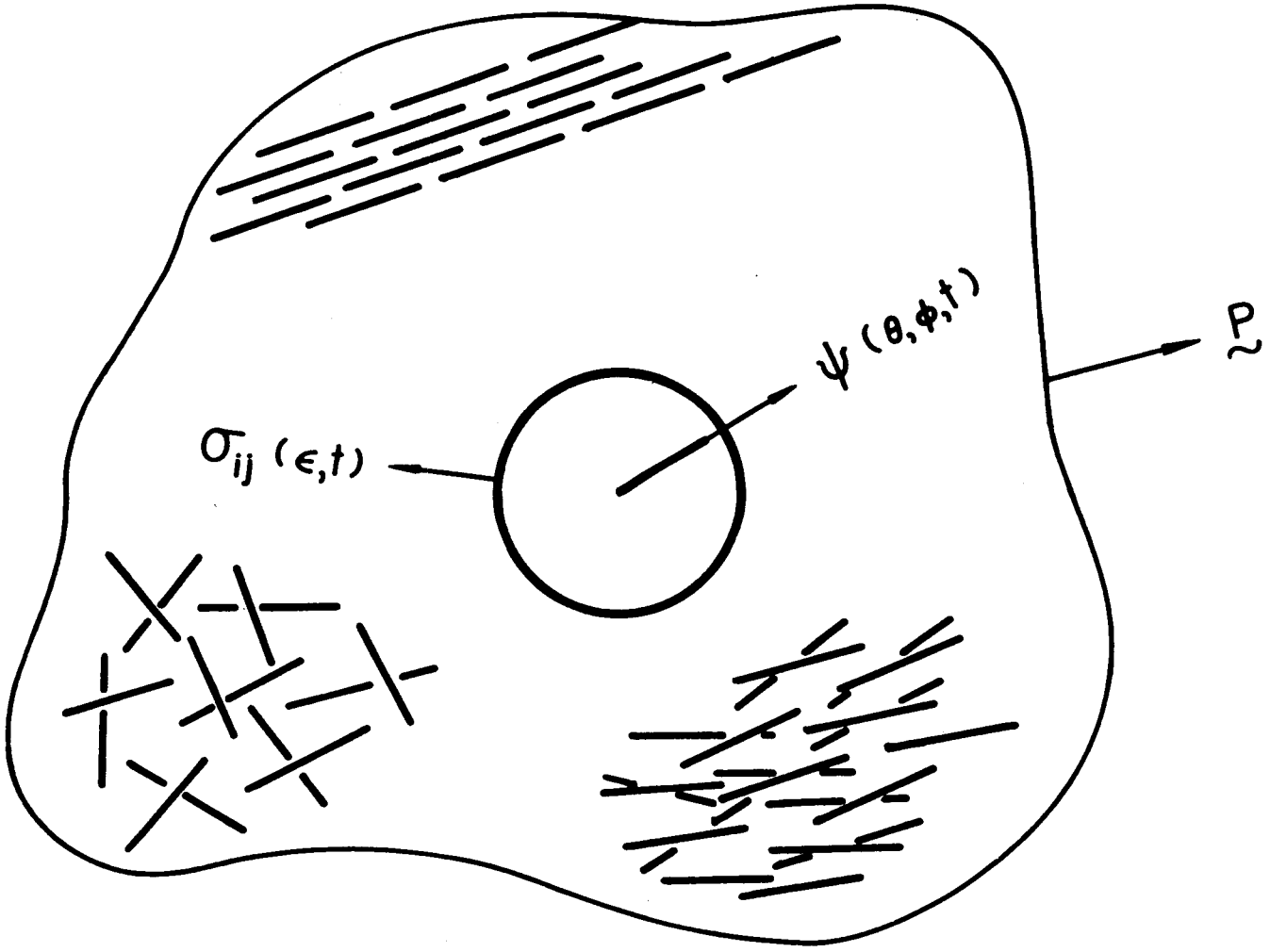
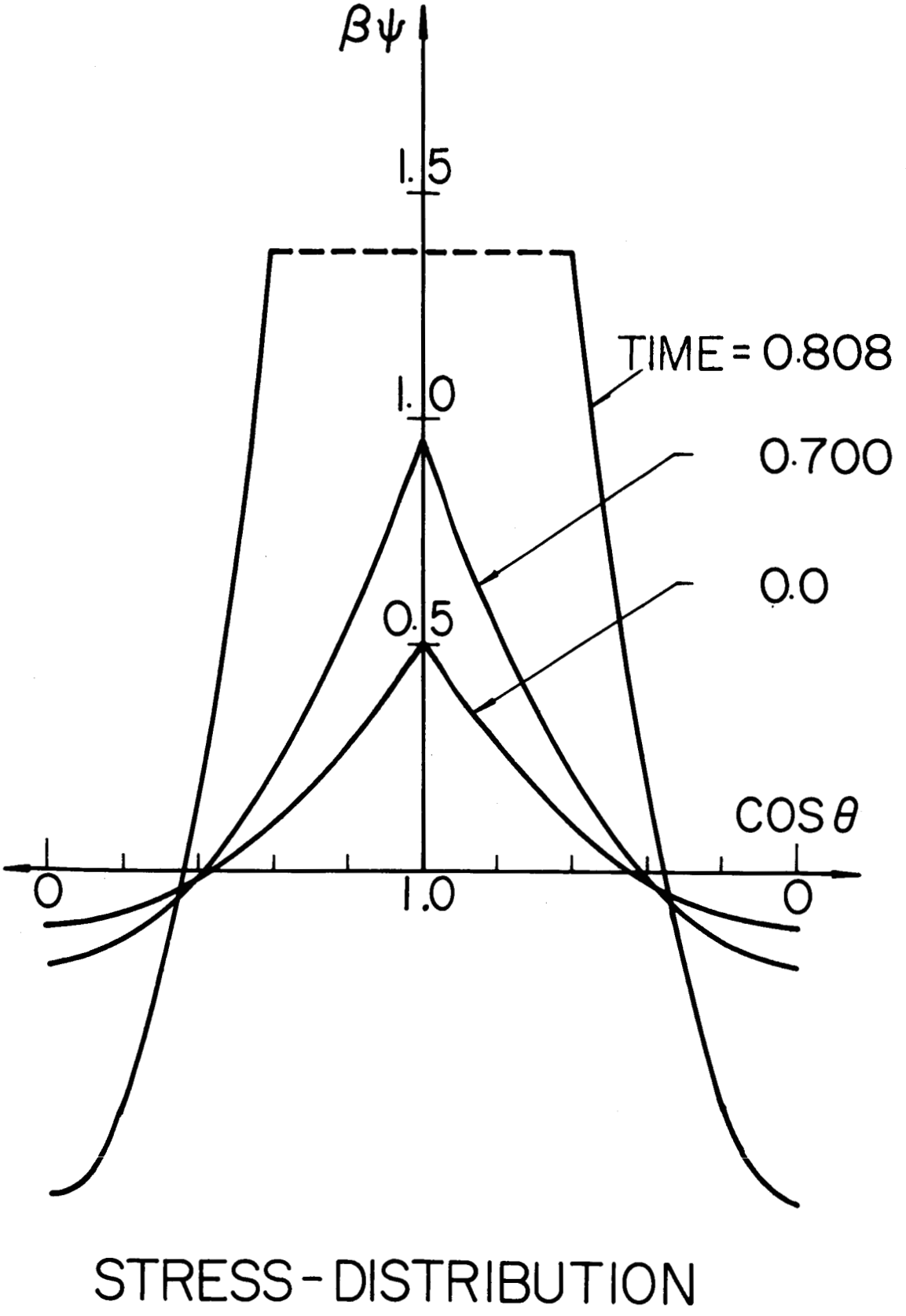


Fig. 1 Illustration of the Mathematical Model, Showing "Elastic Elements" at Different Degrees of Orientation



STRESS-DISTRIBUTION

Fig. 2 Local Stress ψ as a Function of Element Orientation

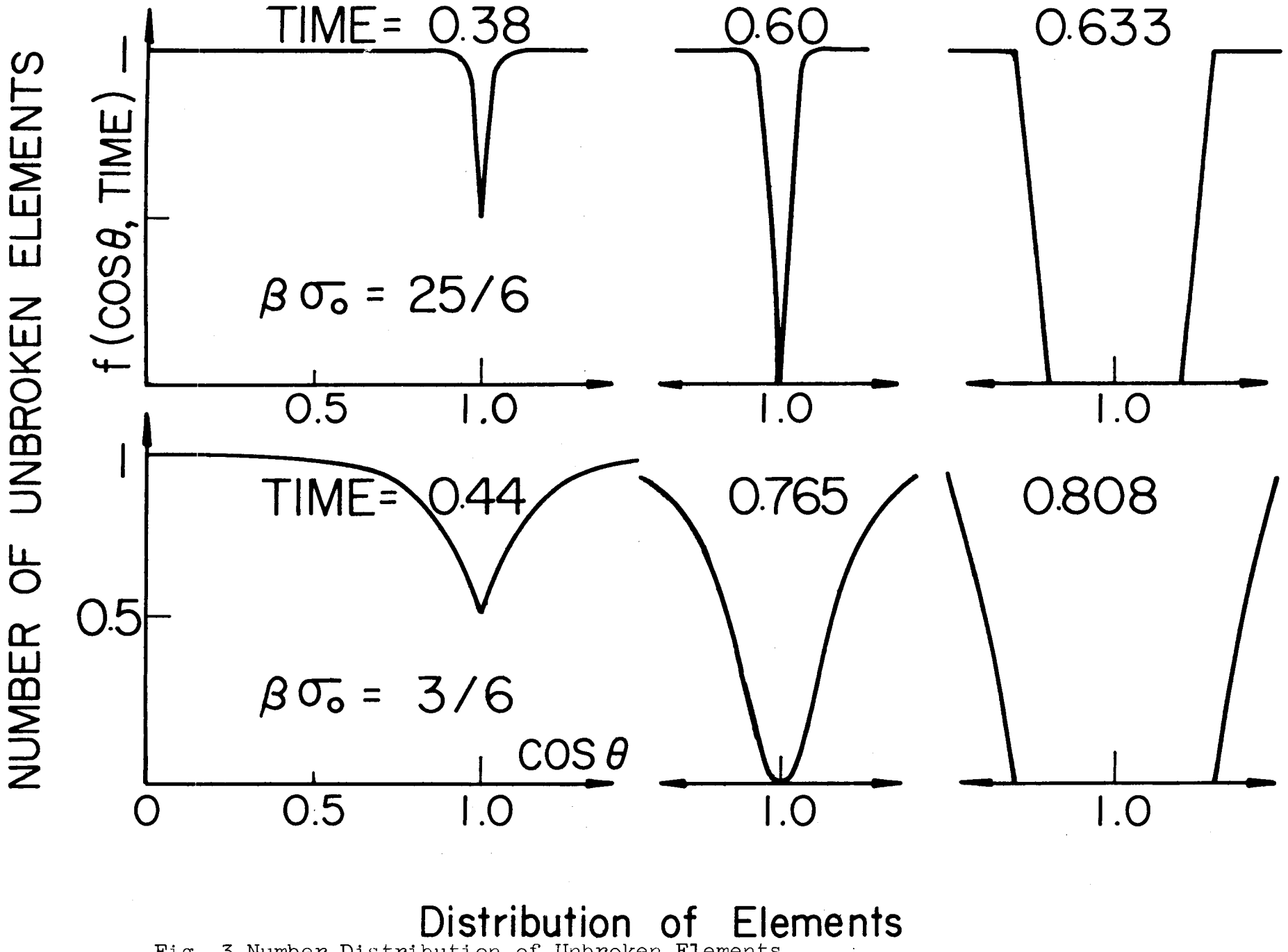


Fig. 3 Number-Distribution of Unbroken Elements

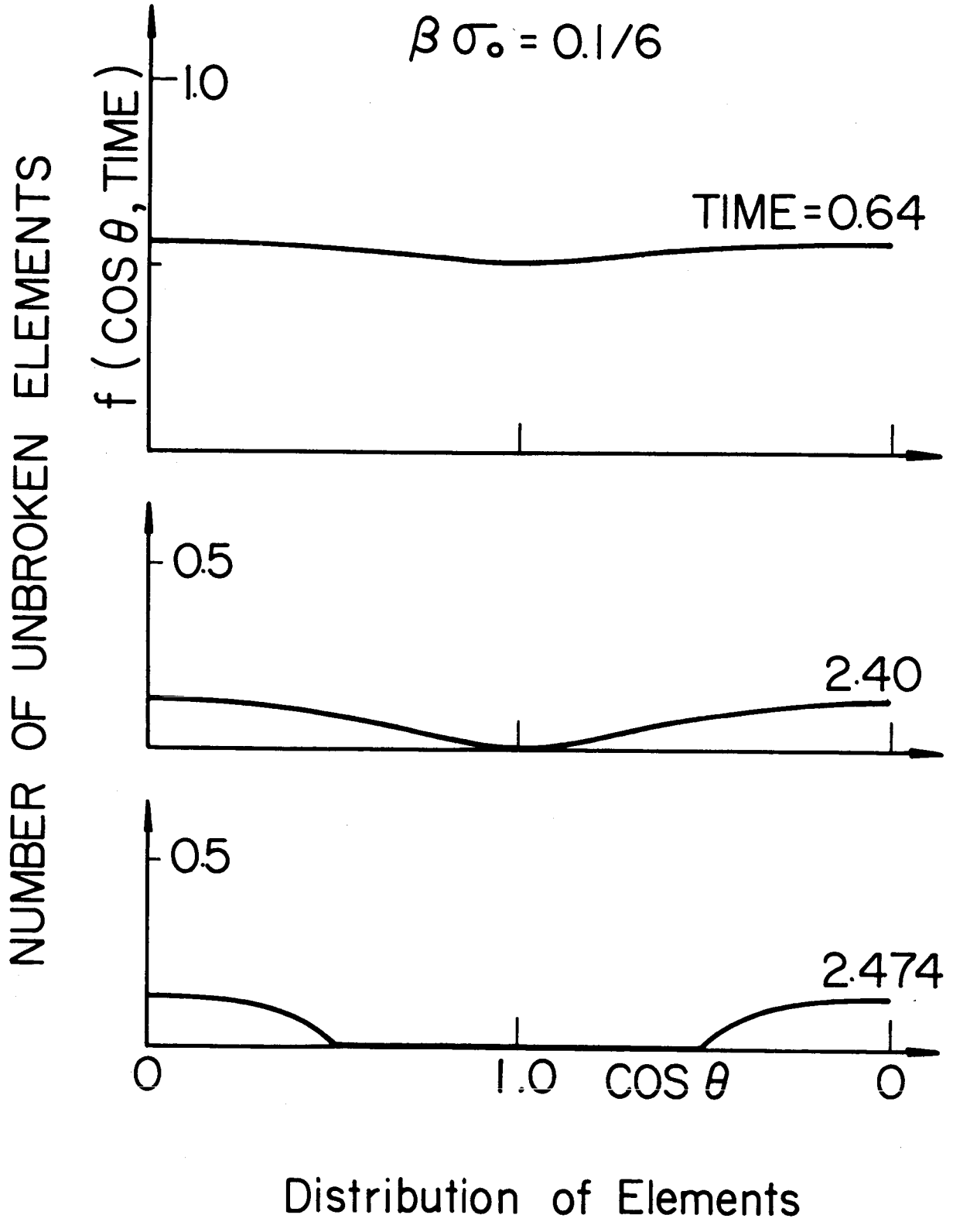


Fig. 4 Number-Distribution of Unbroken Elements

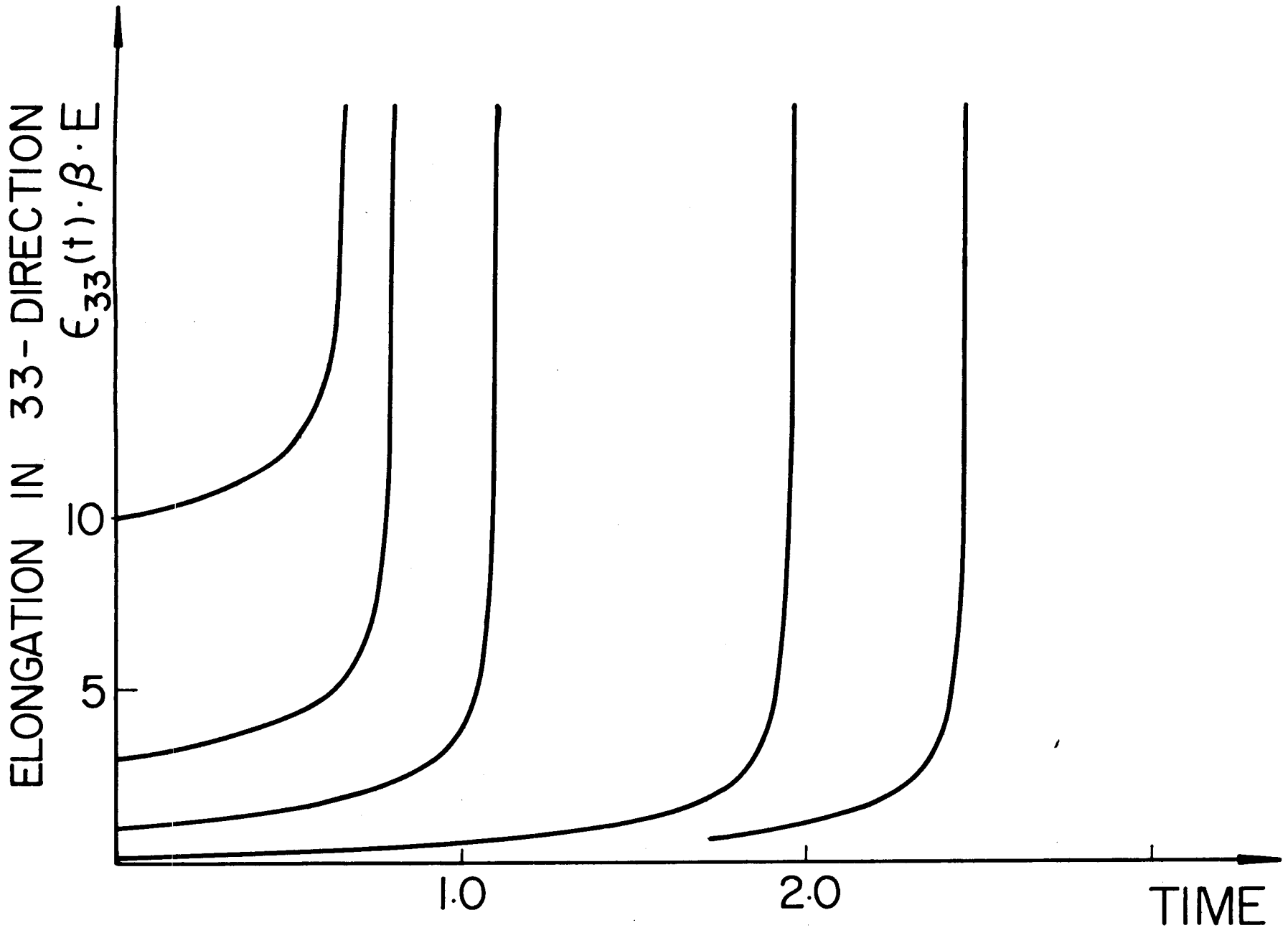
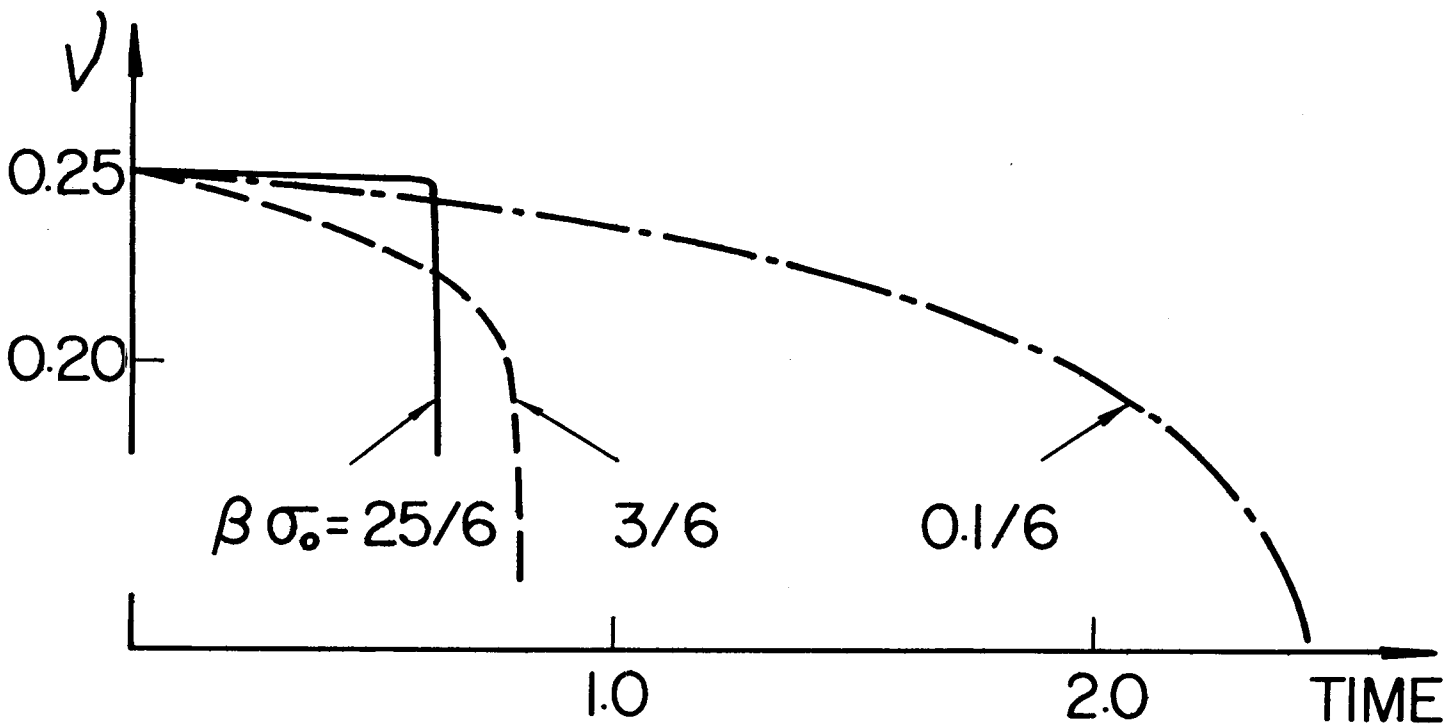


Fig. 5 Elastic Compliance or Elongation in 33-Direction of Uniaxially Stressed Elastic Networks ($\beta\sigma_o$ as parameter)



Change of Strain Ratio with Time

Fig. 6 Change of Strain Ratio with TIME

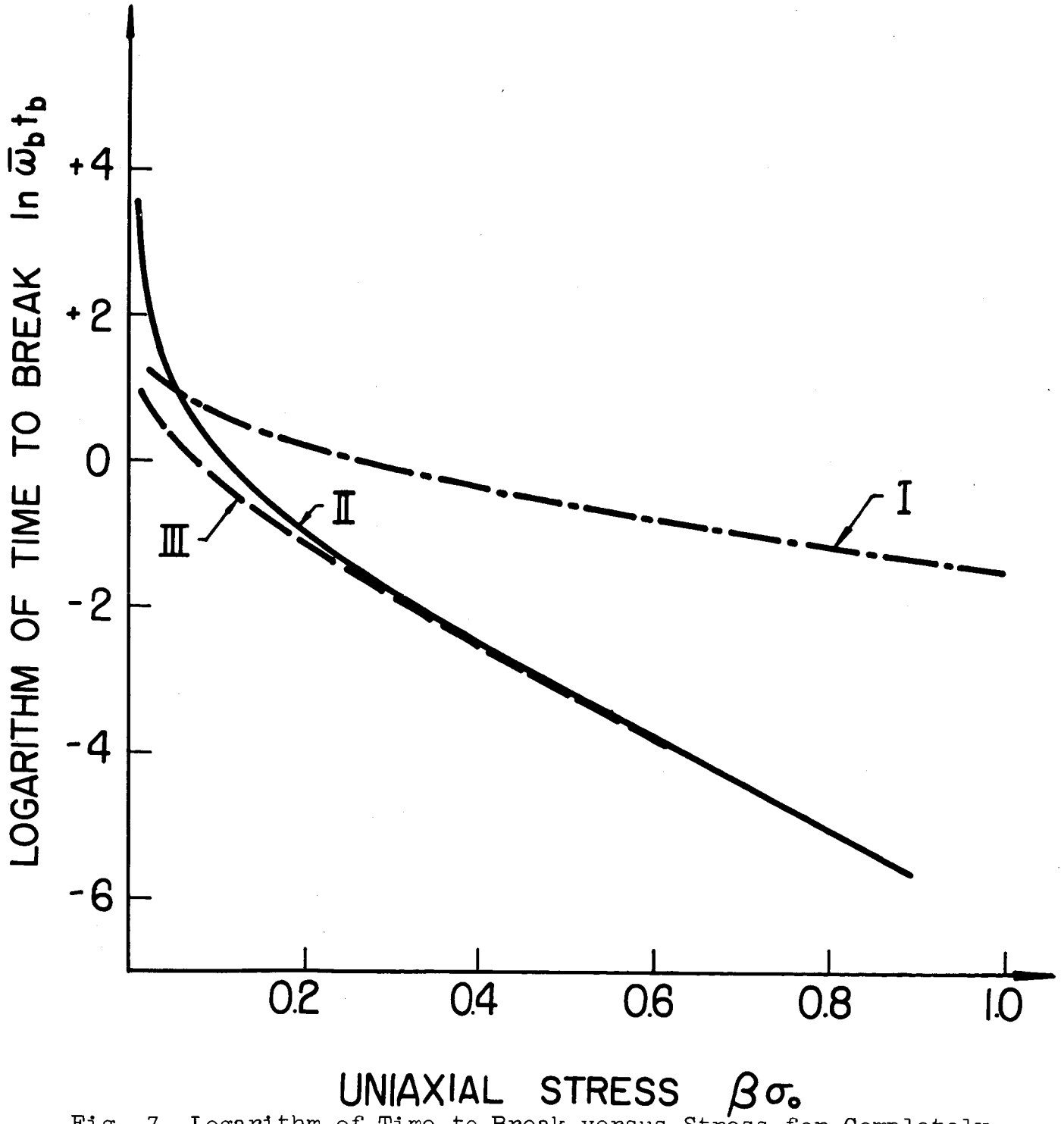


Fig. 7 Logarithm of Time-to-Break versus Stress for Completely Oriented and Randomly Oriented Networks (cf. text for difference of II and III)

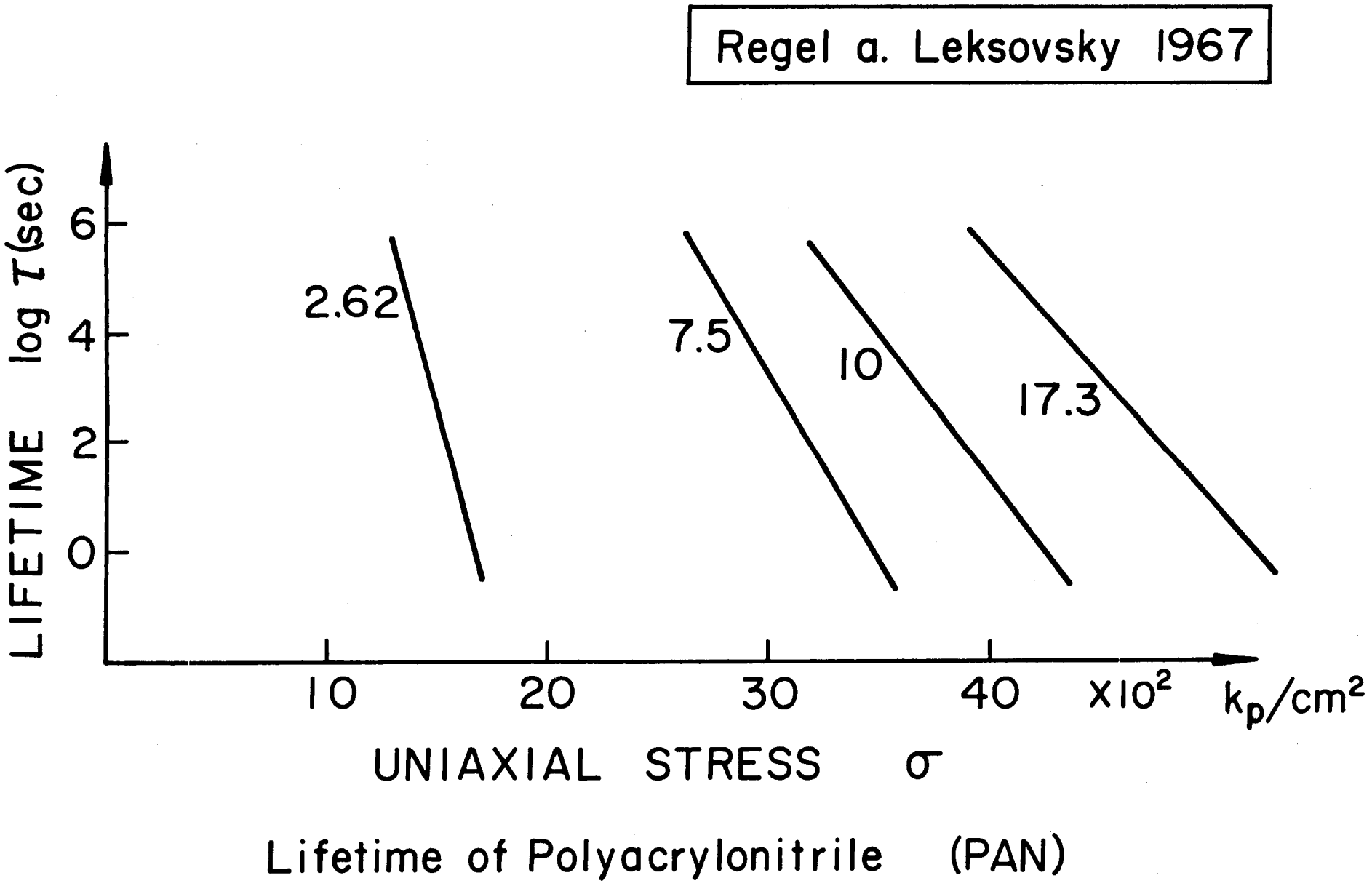
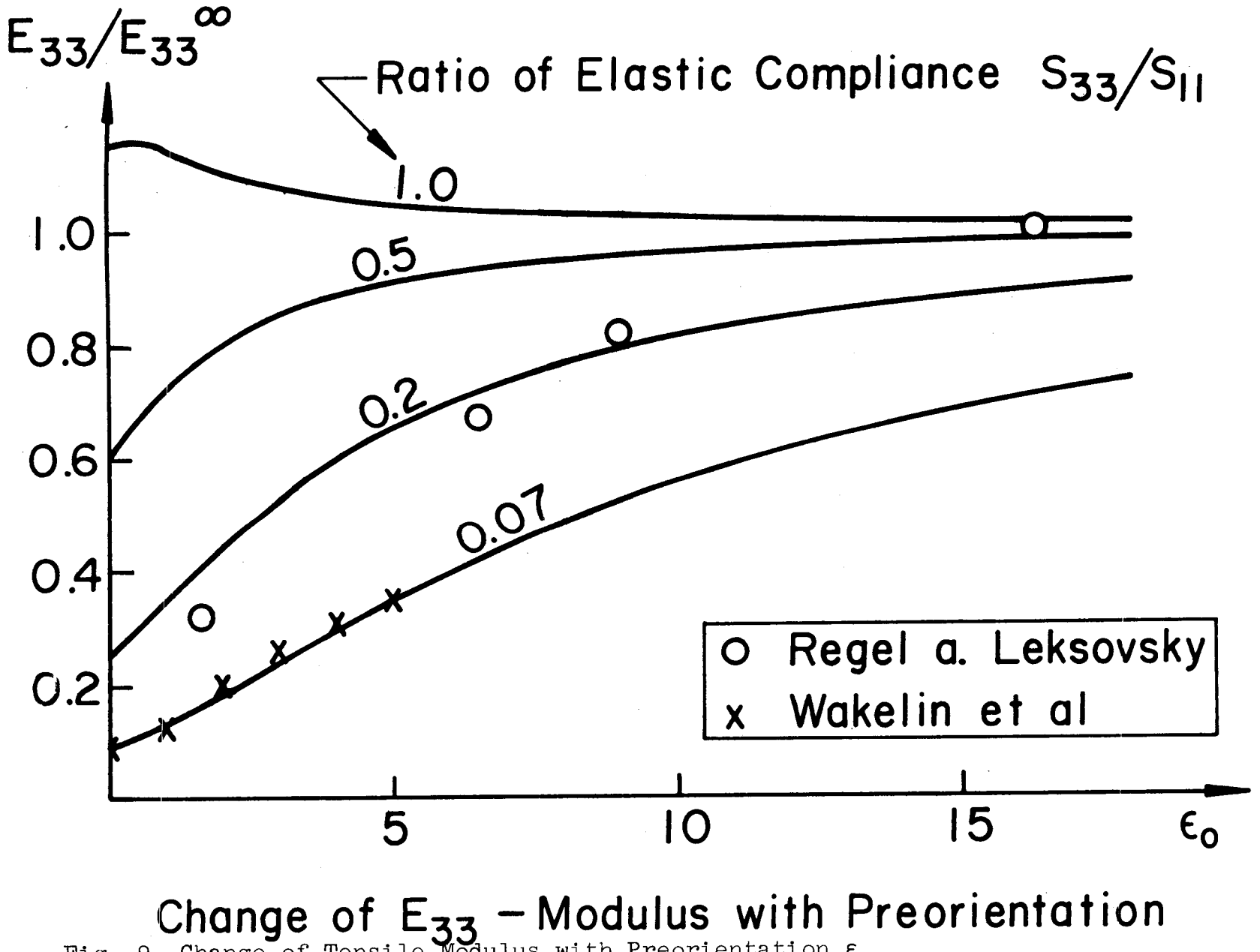


Fig. 8 Logarithm of Time-to-Break versus Stress for Intermediate Orientations of Polyacrylonitrile (PAN-) Samples (Experimental Data of Regel and Leksovsky[9]) (Draw-Ratio of Samples as Parameter)



Change of E_{33} – Modulus with Preorientation

Fig. 9 Change of Tensile Modulus with Preorientation ε

xxxx directly measured by Wakelin et al.[10]

ooo $E(\varepsilon)/E(16.3)$ from data of Fig. 8

—— Calculated Curves for Heterogeneous Materials, using the Assumption of Homogeneous Stress[6]