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Determination of Shift Factors from Dynamic Mechanical Measurements at a Constant Frequency

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ABSTRACT: The theoretical interrelation between linear viscoelastic responses and shift factors has been established for thermorheologically simple materials. The analysis of this interrelation shows that the temperature dependence of shift factors is determined by the temperature dependence of dynamic mechanical responses at a single frequency, and as a consequence the isochronal mechanical relaxation scans at one frequency actually carry as much information as isothermal scans at numerous temperatures. Based on this theory a method has been proposed for obtaining shift factors and other useful information from isochronal dynamic data. Using this method the shift factors for unoriented poly(ethylene terephthalate) have been determined over the temperature range from −131 to 182 °C. The result shows that the temperature dependence of shift factors for this sample is quite different in the range above and below 75 °C. The former could be described by a WLF equation with T_o = 85 °C, C_1 = 24.9, and C_2 = 88.8, except for the data at temperatures above 150 °C. The latter shows linear dependence on temperature. However, as far as the range between −75 and −15 °C is considered, the shift factors could be fitted well to an Arrhenius equation with apparent activation energy of 16.5 kcal/mol. These results are in good agreement with the data available from literature for similar samples.

Introduction

Computer automated dynamic mechanical measurements have become a general method for studing the mechanical relaxation behavior of polymers. The experiments are often carried out in a temperature scan mode, and the data obtained are storage and loss moduli as a function of temperature at one or a few selected frequencies. In past decades this technique has been extensively used in polymer characterization. However, very few theories for interpreting the data quantitatively were available up to now.

One of the most important results from theoretical analysis of isochronal experiments is the theory on areas below loss factor versus $1/T$ plots given by Read and coworkers.[1,2] The theory makes it possible to calculate the average activation energy from the difference of unrelaxed and relaxed modulus and the area below loss modulus versus $1/T$ plots. Boyd et al. proposed a method for fitting the Cole–Cole equation to isochronal mechanical relaxation scans.[3-5] The parameters determined by the curve fitting have been used to interpret the relaxation process of the polymers with satisfaction.[3,4] In 1985 we reported a procedure by which the shift factor and other basic viscoelastic functions in the main transition zone can be worked out from dynamic mechanical temperature spectra, assuming that the temperature dependence of shift factors takes certain forms.[6] The result has been confirmed by independent experiments and the data in the literature.[7,8] Some parameters obtained are useful in studing the fine structure–property relation of the materials. Recently this work has been greatly improved from both theoretical and practical points of view. It could be concluded from our present work that, for a thermorheologically simple material, an isochronal mechanical relaxation scan actually carries as much information as numerous isothermal scans at various temperatures. A detailed description of the theory and method for obtaining the information from isochronal dynamic data is made in this paper.

Theory

1. The Relationship between the Linear Viscoelastic Responses and Shift Factors. As is well-known, the dynamic mechanical response of linear viscoelastic materials is usually represented by the storage and loss moduli which could be expressed as follows

$$E'(\omega,T) = E_0 + \int_{-\infty}^{\infty} H(\tau,T)\omega^2\tau^2/(1+\omega^2\tau^2)\,\mathrm{d}(\ln\tau) \qquad (1)$$

$$E''(\omega,T) = \int_{-\infty}^{\infty} H(\tau,T)\omega\tau/(1+\omega^2\tau^2)\,\mathrm{d}(\ln\tau) \qquad (2)$$

where $E'(\omega,T)$ and $E''(\omega,T)$ are storage and loss moduli at frequency ω and temperature T, E_0 is the relaxed modulus, and $H(\tau,\mathrm{T})$ is the relaxation time spectrum at temperature T. Consider a system obeying the simple time–temperature superposition principle, or thermorheologically simple material. We have

$$E^*(\omega,T_1) = E^*(\omega a_{T_2}(T_1),T_2) \qquad (3)$$

where E* is the complex modulus, T_1 and T_2 represent two arbitrary temperatures which can be constants or variables, $a_{T_2}(T_1)$ represents the shift factor a_T at temperature T_1 with T_2 as a reference temperature. This new expression for the shift factor will be extensively used for convenience in the following discussion. From eq 3 we have

$$a_{T_1}(T_2) = 1/a_{T_2}(T_1) \qquad (4a)$$

$$a_{T_1}(T_2) = a_{T_1}(T_0)\,a_{T_0}(T_2) \qquad (4b)$$

where T_0 represents another arbitrary temperature, and

$$H(\tau,T_1) = H(\tau a_{T_1}(T_2),T_2) \qquad (5)$$

Now the dummy variable in eq 1 is substituted by variable T' as

$$\tau = \tau_0 a_{T'}(T) \qquad (6)$$

where τ_0 is an arbitrary constant. The equation thus

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becomes

$$E'(\omega,T) = E_0 + \int_{-\infty}^{\infty} H(\tau_0 a_{T'}(T),T)\frac{\partial \ln a_{T'}(T)}{\partial T'} \times \frac{\omega^2 \tau_0^{\,2} a_{T'}(T)^2}{1 + \omega^2 \tau_0^{\,2} a_{T'}(T)^2}\,\mathrm{d}T' \quad (7)$$

Note that from eq 5

$$H(\tau_0 a_{T'}(T),T) = \mathrm{H}(\tau_0,T')$$

and that $\partial \ln a_{T'}(T)/\partial T'$ is solely a function of T' (independent of T) as can be seen from eq 4. Put

$$H(\tau_0,T')\,\partial \ln a_{T'}(T)/\partial T' = H^*(\tau_0,T') \quad (8)$$

Thus eq 7 can be rewritten as

$$E'(\omega,T) = E_0 + \int_0^{\infty} H^*(\tau_0,T')\omega^2 \tau_0^{\,2} a_{T'}(T)^2/(1 + \omega^2 \tau_0^{\,2} a_{T'}(T)^2)\,\mathrm{d}T' \quad (9)$$

In exactly the same way, one could derive the alterative expressions for loss modulus from eq 2

$$E''(\omega,T) = \int_0^{\infty} H^*(\tau_0,T')\omega \tau_0 a_{T'}(T)/(1 + \omega^2 \tau_0^{\,2} a_{T'}(T)^2)\,\mathrm{d}T' \quad (10)$$

and the stress relaxation modulus at temperature T

$$\begin{aligned} E(t,T) &= E_0 + \int_{-\infty}^{\infty} H(\tau,T)\exp(-t/\tau)\,\mathrm{d}(\ln \tau) \\ &= E_0 + \int_0^{\infty} H^*(\tau_0,T')\exp(-t/\tau_0 a_{T'}(T))\,\mathrm{d}T' \quad (11) \end{aligned}$$

From eqs 9 to 11 it is evident that $H^*(\tau_0,T')$ plays the role of a relaxation spectrum. If T' is termed relaxation temperature which is related to the relaxation time τ by eq 6, then $H^*(\tau_0,T')\,\mathrm{d}T'$ gives the contributions to the responses associated with relaxation temperatures between T' and $T' + \mathrm{d}T'$. Equations 9–11 show that all of the storage and loss moduli, $E'(\omega,T)$ and $E''(\omega,T)$, as well as the stress relaxation modulus $E(t,T)$ can be directly related to the same relaxation spectrum $H^*(\tau_0,T')$ and shift factor $a_{T'}(T)$.

Similar expressions can be derived by analogous treatment for the storage and loss compliances $D'(\omega,T)$ and $D''(\omega,T)$, and the creep compliance $D(t,T)$. The results are presented here for completion

$$\begin{aligned} D'(\omega,T) &= D_0 + \int_{-\infty}^{\infty} L(\tau,T)/(1 + \omega^2\tau^2)\,\mathrm{d}(\ln \tau) \\ &= D_0 + \int_0^{\infty} L^*(\tau_0,T')/(1 + \omega^2 \tau_0^{\,2} a_{T'}(T)^2)\,\mathrm{d}T' \quad (12) \end{aligned}$$

$$\begin{aligned} D''(\omega,T) &= \int_{-\infty}^{\infty} L(\tau,T)\omega\tau/(1 + \omega^2\tau^2)\,\mathrm{d}(\ln \tau) \\ &= \int_0^{\infty} L^*(\tau_0,T')\omega\tau_0 a_{T'}(T)/(1 + \omega^2 \tau_0^{\,2} a_{T'}(T)^2)\,\mathrm{d}T' \quad (13) \end{aligned}$$

$$\begin{aligned} D(t,T) &= D_0 + \int_{-\infty}^{\infty} L(\tau,T)(1 - \exp(-T/\tau))\,\mathrm{d}(\ln \tau) \\ &= D_0 + \int_0^{\infty} L^*(\tau_0,T')(1 - \exp(-t/\tau_0 a_{T'}(T)))\,\mathrm{d}T' \quad (14) \end{aligned}$$

where $L(\tau,T)$ is the retardation spectrum at temperature T, D_0 is the instantaneous elastic compliance, τ_0 is an arbitrary constant, and $L^*(\tau_0,T')$ is a new retardation spectrum defined by

$$L^*(\tau_0,T') = L(\tau_0,T')\,\partial \ln a_{T'}(T)/\partial T' \quad (15)$$

$L^*(\tau_0,T')\,\mathrm{d}T'$ gives the contribution to the responses associated with retardation temperatures between T' and $T' + \mathrm{d}T'$.

In the present work our interest is mainly focused on the dynamic moduli as a function of temperature at a constant frequency. In this case we have $\omega = \omega_0$. Put $\tau_0 = 1/\omega_0$. Then eqs 9 and 10 become

$$E'(\omega_0,T) = E_0 + \int_0^{\infty} H^*(1/\omega_0,T')\,a_T'(T)^2/(1 + a_{T'}(T)^2)\,\mathrm{d}T' \quad (16)$$

$$E''(\omega_0,T) = \int_0^{\infty} H^*(1/\omega_0,T')\,a_{T'}(T)/(1 + a_{T'}(T)^2)\,\mathrm{d}T' \quad (17)$$

An abbreviation of $E'(\omega_0,T)$, $E''(\omega_0,T)$, and $H^*(1/\omega_0,T')$ to $E'(T)$, $E''(T)$, and $H^*(T')$ allows us to write

$$E'(T) = E_0 + \int_0^{\infty} H^*(T')\,a_{T'}(T)^2/(1 + a_{T'}(T)^2)\,\mathrm{d}T' \quad (18)$$

$$E''(T) = \int_0^{\infty} H^*(T')\,a_{T'}(T)/(1 + a_{T'}(T)^2)\,\mathrm{d}T' \quad (19)$$

The equations show that the isochronal dynamic moduli could be related to shift factors in a simple and explicit version. It will be verified that the equations are very useful in analysis of isochronal mechanical relaxation scans.

2. Determination of Shift Factors from Dynamic Mechanical Measurements at a Constant Frequency. For eqs 18 and 19 derived above, a question of interest arises: given $E'(T)$ and $E''(T)$, can one solve the shift factor $a_{T'}(T)$ and relaxation spectrum $H^*(T')$ from the equations? In this section the problem will be discussed with the help of a numerical treatment. In fact, for a sufficiently large N, we have

$$E'(T) \doteq E_0 + \sum_{i=1}^{N} H^*(T_i)h_i\,a_{T_i}(T)^2/(1 + a_{T_i}(T)^2) \quad (20a)$$

$$E''(T) \doteq \sum_{i=1}^{N} H^*(T_i)h_i\,a_{T_i}(T)/(1 + a_{T_i}(T)^2) \quad (20b)$$

where T_i ($i = 1, ..., N$) are temperatures (quadrature nodes) properly chosen, h_i ($i = 1, ..., N$) are corresponding coefficients of quadrature. Letting the left and right sides of eq 20 be equal at every node T_j ($j = 1, ..., N$), we have the following system of equations

$$E'(T_j) = E_0 + \sum_{i=1}^{N} H^*(T_i)h_i\,a_{T_i}(T_j)^2/(1 + a_{T_i}(T_j)^2) \quad (21a)$$

$$E''(T_j) = \sum_{i=1}^{N} H^*(T_i)h_i\,a_{T_i}(T_j)/(1 + a_{T_i}(T_j)^2) \quad (21b)$$

$$j = 1, ..., N$$

where $H^*(T_i)$ and $a_{T_i}(T_j)$ ($i;j = 1, ..., N$) are approximate values for $H^*(T')$ and $a_{T'}(T)$ at the specified T_i, T_j ($i;j = 1, ..., N$). Using the abbreviation as $E'(T_j) = E'_j$, $E''(T_j) = E''_j$, $a_{T_i}(T_j) = a_{ij}$ and $H^*(T_i)h_i = E_i$, eq 21 can be simplified as

$$E'_j = E_0 + \sum_{i=1}^{N} E_i\,a_{ij}^{\,2}/(1 + a_{ij}^{\,2}) \quad (22a)$$

$$E'' = \sum_{i=1}^{N} E_i\,a_{ij}/(1 + a_{ij}^{\,2}) \quad (22b)$$

$$j = 1, ..., N$$

In eq 22 E'_j and E''_j ($j = 1, ..., N$) can be obtained from

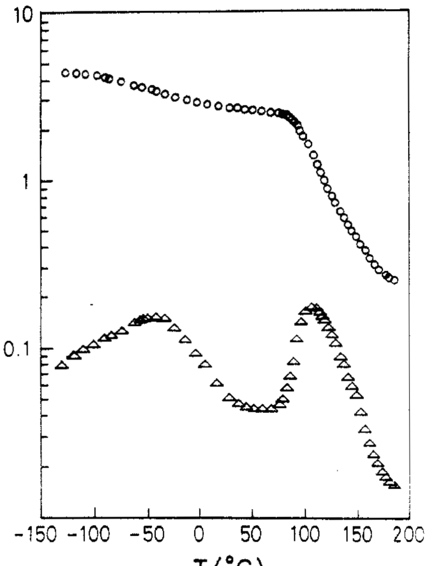

Figure 1. Dynamic mechanical properties of unoriented PET as a function of temperature at 110 Hz.

isochronal dynamic measurements, E_i ($i = 0, 1, ..., N$) and a_{ij} ($i;j = 1, ..., N$) are unknown parameters. Note that from eq 4

$$a_{ij} = a_{ik}a_{kj} = a_{ik}/a_{jk}$$

$$1 \leqslant i;j;k \leqslant N$$

and therefore the total number of independent unknown parameters for a_{ij} ($i;j = 1, ..., N$) is $N - 1$ (note $a_{kk} = 1$). Thus, eq 22 is actually a system of nonlinear equations including $2N$ equations for $2N$ independent unknown parameters, and an unique solution might be found by means of a suitable computational method. Once a_{ij} and E_i are obtained, $a_{T'}(T)$ and $H^*(T')$ could be approximately determined by such mathematical methods as interpolation.

The analysis above has revealed a fact that $a_{T'}(T)$ and $H^*(T')$ are determined by an isochronal mechanism relaxation scan, recalling eqs 9 and 10 which show that the dynamic moduli at any frequency and temperature can be obtained if $a_{T'}(T)$ and $H^*(\tau_0,T')$ are available. Thus it can be concluded that a single isochronal scan actually carries as much information as numerous frequency scans at various temperatures, and the isochronal experiments are not only more convenient but also useful in comparison to isothermal ones.

Experimental Section

The sample used in dynamic mechanical measurements is an unoriented film of poly(ethylene terephthalate) (PET). The polymer with an inherent viscosity of 0.59 was pressed at 300 °C and subsequently quenched by cold water. The film with a thickness of about 0.1 mm was then annealed at 190 °C for 2 h under vacuum. The dynamic mechanical measurements were performed on a Rheovibron Model DDV-II-EA over the range from −150 to 200 °C at a heating rate of 2 °C/min. The frequency used was 110 Hz.

Results and Discussion

Figure 1 illustrates the temperature dependence of dynamic mechanical properties (complex moduli) for the sample from −131.3 to 182.7 °C. The data were analyzed using the method described above assuming that the material is approximately thermorheologically simple in this temperature range. From this interval 54 temperatures have been chosen as T_i ($i = 1, ..., N$; $N = 54$), as shown in Table I. Substituting storage and loss moduli at T_i ($i = 1, ..., N$) into eq 22 gives the shift factor a_T and E_i ($i = 0, 1, ..., N$), the result is listed in Table I. The plots of log a_T against temperature and log a_T against the reciprocal of absolute temperature were shown in Figures 2 and 3, respectively.

Table I. Discrete Relaxation Spectrum and Shift Factors for Unoriented PET Calculated from Dynamic Data of Figure 1[a]

i	T_i, °C	E_i	log a_T	i	T_i, °C	E_i	log a_T
1	−131.3	11.45	33.00	31	90.6	6.14	13.97
2	−119.5	6.33	32.46	32	92.6	10.39	13.52
3	−111.3	8.27	31.86	33	94.6	8.36	13.01
4	−101.3	8.77	31.33	34	96.7	16.18	12.53
5	−93.3	9.71	30.68	35	100.7	19.89	11.68
6	−87.9	10.67	30.15	36	104.7	19.44	10.97
7	−76.5	11.34	29.22	37	108.6	19.79	10.15
8	−71.4	9.92	28.85	38	112.6	13.70	9.45
9	−65.4	10.67	28.26	39	116.6	11.24	8.96
10	−57.2	11.34	27.82	40	120.7	11.83	8.30
11	−52.7	11.77	27.28	41	124.6	7.94	7.75
12	−43.7	13.23	26.76	42	128.7	6.29	7.28
13	−35.2	14.06	26.13	43	132.6	5.49	6.82
14	−25.3	12.67	25.51	44	136.6	5.45	6.28
15	−15.3	12.83	24.80	45	140.6	3.87	5.79
16	−5.2	7.36	24.15	46	144.6	3.33	5.33
17	4.6	6.80	23.57	47	148.7	3.57	4.73
18	14.7	6.78	22.83	48	152.6	3.66	4.11
19	24.7	5.09	22.08	49	156.6	2.98	3.27
20	32.6	5.00	21.40	50	158.6	2.18	2.78
21	40.7	4.47	20.66	51	162.7	2.66	1.95
22	48.6	4.59	20.14	52	166.7	1.89	1.29
23	56.6	4.40	19.17	53	172.6	1.49	0.685
24	64.6	3.81	18.56	54	182.7	2.18	0.00
25	72.6	4.54	17.99				
26	76.6	5.25	17.26	0		24.44	
27	80.6	6.79	16.45				
28	84.6	8.79	15.48				
29	86.6	2.54	14.92				
30	88.6	9.86	14.46				

[a] The original data for E_i and a_T determined by computer contains seven significant digits which are not completely presented here for the sake of brevity. Units of E_i are 10^{-2} GPa; the reference temperature of a_T is 182.7 °C.

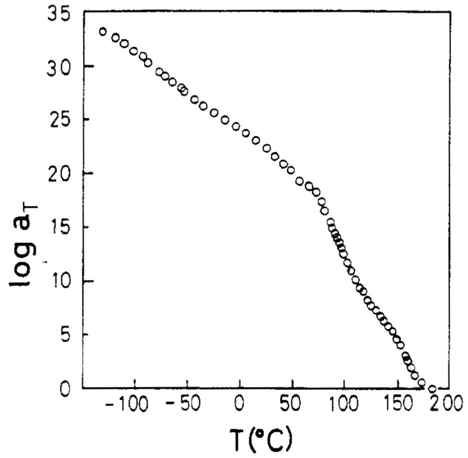

Figure 2. Temperature dependence of shift factors for unoriented PET obtained from dynamic data shown in Figure 1.

The rational for the result obtained is evaluated first by examining the precision of the solution. The precision could be represented by a deviation of back-calculated moduli (denoted as $\hat{E}'_j$ and $\hat{E}''_j$) from observed ones (denoted as E'_j and E''_j), that is

$$\rho_{E'} = \max_{1 \leqslant j \leqslant N} |(E'_j - \hat{E}'_j)/E'_j|$$

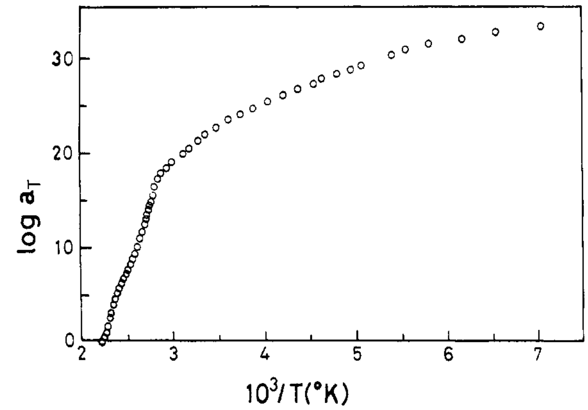

Figure 3. Logarithm of shift factors plotted against reciprocal absolute temperature for unoriented PET.

$$\rho_{E''} = \max_{1 \leqslant j \leqslant N} |(E''_j - \hat{E}''_j)/E''_j|$$

Where $\rho_{E'}$ and $\rho_{E''}$ represent the deviations for storage and loss moduli, respectively. The values of the deviation for our solution as displayed in Table I are $\rho_{E'} = 1.69 \times 10^{-7}$ and $\rho_{E''} = 1.69 \times 10^{-6}$. Obviously, the precision of the solution is excellent, especially considering that the problem dealt with is a system of nonlinear equations including as many as 54 pairs of experimental data covering a very broad temperature range.

Figure 2 shows the temperature dependence of shift factors from −131.3 to 182.7 °C, with 182.7 °C as a reference temperature. As can be seen from the figure, the shift behavior is significantly different in the range above and below 75 °C. In the range below 75 °C the log a_T–T curve is approximately a straight line with a slope of 0.073. From 75 °C the shift factor becomes much more temperature dependent, and the log a_T–T curve turns into a WLF type (T_0 = 85 °C, C_1 = 24.9, C_2 = 88.8), except for the data at temperatures above 150 °C.

The abrupt change in shift behavior at a temperature around 75 °C is obviously due to the glass transition of PET. In the temperature range above 75 °C an α-relaxation process takes place, and the temperature dependence of viscoelastic behavior has been extensively investigated.[2,9-14] Based on the extensional creep measurements, Ward[9] pointed out that the a_T for unoriented PET (heat treated at 180 °C for 1 h) obeys the WLF equation in the range between 75 and 110 °C. The WLF equation was believed to be still valid when the temperature was raised to 140 °C if the dielectric data of Reddish[10] was taken into account. The stress relaxation measurements of Takayanagi[11] and the dielectric data of Saito[12] have also shown that the temperature dependence of a_T for PET obeys the WLF equation, although the coefficients of the equation may depend on the crystallinity of the samples.[12] For an oriented PET sample however, the stress relaxation data reported by Murayama[13] show different behavior. The sample was found to be thermorheologically simple in the range between 22 and 182 °C, but from 64 to 182 °C the log a_T–T plot consists of a straight line. In the viewpoint of Murayama, a straight line with a slop of 0.225 fits the data of Ward and Takayanagi equally well, but the temperature range covered was too small to decide definitely which relationship fits the data best.[13] From our results covering broad temperature ranges, one can note that the WLF equation fitted the shift factor data for unoriented PET. However, if a straight line is drawn to fit our data at temperatures between 70 and 110 °C, the slop of the line will be 0.232, a value which is in reasonable agreement with that of Ward and Takayanagi in the same range of temperatures.

The temperature dependence of the relaxation process for PET in a low temperature range was characterized by the dependence of the loss-peak on temperature.[14] As noted by Illers and Breuer, the shift data were concentrated in the range between −70 and −25 °C and an Arrhenius dependence was observed.[14] Typical values of the activation energy were 17 kcal/mol for dry or nearly dry PET as derived from mechanical measurements and 12.4 kcal/mol as derived from all dielectric results together with the dynamic mechanical results of water-swelled PET. All activation energies reported in the literature are within these limits.[14] As can be seen from Figure 3, our shift data below 75 °C could not be fitted to an Arrhenius equation completely. But if the temperature range between −75 and −15 °C is considered, log a_T–$1/T$ plot could be represented well by a straight line. That is, the temperature dependence of a_T in this range obeys an Arrhenius equation. The activation energy obtained from Figure 3 is 16.5 kcal/mol, which is in good agreement with the results reported in the literature obtained by conventional methods.

Table I also listed the results of E_i ($i = 0, 1, ..., N$) which has been referred to as a discrete relaxation spectrum. No attempt has been made to smooth the data. It should be pointed out that, for some purpose such as the conversion of dynamic data into other viscoelastic functions, it may be more convenient and accurate to use the discrete data directly. For example, the stress relaxation modulus at T_k ($1 \leqslant k \leqslant N$) can be calculated by substituting a_{ij} and E_i into the following equation derived from eq 11

$$E(t,T_k) = E_0 + \sum_{i=1}^{N} E_i \exp(-\omega_0 t/a_{ik}) \qquad (23)$$

A detailed discussion on the relaxation spectrum obtained will be given elsewhere.

Information about our algorithm, which is a key to the present method, is now outlined briefly. The applicability of the algorithm was examined first by computer simulation experiments. That is, the synthesized dynamic data were analyzed to see if the original relaxation spectrum and shift factors can be recovered. Then, all suitable isochronal dynamic data measured in our laboratory have been successfully converted into shift factors and relaxation spectra by the program, with the agreement between dynamic moduli observed and back-calculated being as good as that of the unoriented PET described above. The maturity of the program can be further demonstrated as follows: First, the program can automatically decide the proper number N and the distribution of relaxation temperatures T_j ($j = 1, ..., N$) according to the dynamic data prior to iteration. This was done based on the theory developed by us[15] for estimating discrete relaxation temperature distributions. Second, the shift factors and relaxation spectrum obtained from eq 22 are independent of starting parameters for iteration if T_j ($j = 1, ..., N$) has been decided. Finally, the time required for computation is fairly short. A typical computation time is about 10 min on a personal computer when N is around 50. Thus, this program will make the analysis convenient and useful in practice.

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Author-supplied Registry No. PET, 25038-59-9.

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