Analysis of the Relaxation and Retardation Spectra Obtained from the Double-Log Representation of the Viscoelastic Functions

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ABSTRACT: Relaxation or retardation distribution functions are sometimes used to determine the molecular models of the polymeric structure of a linear viscoelastic material. These spectra are usually calculated from the double-log representation of any mechanical property, using approximation methods. In this paper it is shown that the double-log plots distort the viscoelastic behavior, leading to spectra that do not describe the real mechanical response. To solve this problem, a normalized representation of the measurements is proposed and its improvements are discussed in detail. Particularly, the retardation spectrum determined from the normalized dynamic compliance of polyisobutylene is taken as an example, showing some inconsistencies in the model determined from the double-log representation. Finally, even with the normalized representation, the spectra derived from the master curves do not always provide a good fitting of these curves. Hence, it is pointed out that the analysis of individual curves measured over several decades of time or frequency leads to a better determination of the real distribution function of characteristic times.
KEY WORDS Viscoelastic Functions / Double-Log Representation / Relaxation and Retardation Spectra / Master Curve /

The mechanical properties of a polymeric material are used to characterize the micromechanisms that control its linear viscoelastic behavior. In fact, the spectra related to the linear viscoelastic functions by integral transformations, give a quantitative distribution of the relaxation or retardation mechanisms in different regions of the time scale. As these integral transformations are usually complex, the spectra are calculated by approximation methods from the measured moduli or compliances.[1-4] These methods use the first derivative or even derivatives of higher order of the viscoelastic functions measured over several decades of time or frequency. As it is difficult to measure them over large intervals of time or frequency, in the transition region from the glassy to the rubbery state, a master curve constructed using the time-temperature superposition[5] (ref 5, Chapter 11) is generally used. In this transition region any viscoelastic property varies by approximately over three orders of magnitude thus, the data are usually plotted as a function of time or frequency in a double-logarithmic plot, though other representations may be considered as well. Moreover, though the relaxation and retardation distribution functions should be calculated, the linear viscoelastic relationships indicate that both spectra provide the same information of the structure. Generally, the tests can be divided into two groups: the one related to measurements made under a given stress and the other related to tests done at a

fixed strain, but the link between creep and relaxation processes leads to an interrelation of the spectra.[6] In a classical literature, however, it is established that: "Generally the short time processes are revealed in more detail in the relaxation spectrum and the long time processes in the retardation spectrum" (ref 5, Chapter 3). Therefore, an apparent contradiction between the interrelation of the distribution functions and the prevalence of long or short time processes in the spectra appears. It will be shown in this paper that the contradiction is originated from the double-log representation of any transient or dynamic property. Furthermore, an alternative representation using normalized viscoelastic functions will be discussed in detail.

THEORY

In the following analysis the mechanical properties used to determine the approximate distribution functions will be divided into two groups: the one that corresponds to a viscoelastic property V that evolves from a least value V_1 to a maximum value V_m such as the creep compliance, the relaxation modulus or the real part of the dynamic functions, and the other associated with the imaginary component of the dynamic functions characterized by a peak.

As was pointed out in the Introduction, though the mechanical properties are usually represented as a function of time or frequency in a double-log plot, the normalized viscoelastic functions can also be considered. For the transient properties or the real component of the dynamic modulus or compliance, this function is given by

$$f = \frac{V - V_1}{V_m - V_1} \qquad (1)$$

while for the imaginary component of the dynamic modulus or compliance it is defined as

$$g = \frac{V}{V_m - V_1} \qquad (2)$$

Now, according to eq 1, the normalized function f varies from zero when $V = V_1$ to one for $V = V_m$, being 0.5 for the mean value $V^* = (V_1 + V_m)/2$. Hence, the mean mechanical property differs from the least value by the same amount as it differs from the highest value. This homogeneous distribution of the data, however, it not observed in the double-log plot. In fact, considering that the ratio V_m/V_1 is of the order of 10^3, the difference between the maximum ordinate $\log(V_m)$ and $\log(V^*)$ reduces practically to $\log 2$. The difference between $\log(V^*)$ and the minimum ordinate $\log(V_1)$, however, is equal to $\log[(V_1 + V_m)/2V_1]$ which is generally equal to or greater than 2.7. Then in the double-log representation the values of V greater than V^* are clustered in a small region whose width is equal to $\log 2$, while the lower values extend over nearly three orders of magnitude. This distribution of the data leads to a distorted interpretation because in the compressed region where $V^* \leq V \leq V_m$ there is not enough sensitivity to determine accurate derivatives and, accordingly, to calculate the approximated spectrum properly. On the other hand, in the normalized plot the data are distributed proportionally to the values of the mechanical properties.

To illustrate these concepts, the real part of the dynamic compliance, J', and the lognormal retardation spectrum are considered. The lognormal distribution function is defined as (ref 7, p 95)

$$L(\ln(\tau/\tau_m)) = \frac{J_e - J_g}{\beta \sqrt{\pi}} \exp\{-[\ln(\tau/\tau_m)/\beta]^2\} \qquad (3)$$

where τ is the retardation time, τ_m the mean retardation time, β the half-width of the distribution, J_g the compliance in the glassy state and J_e the equilibrium compliance for a viscoelastic solid or the steady-state compliance for a viscoelastic liquid. This function is symmetrical and can be expressed as

$$L(y) = L_l(y) + L_r(y) \qquad (4)$$

being

$$y = \ln(\tau/\tau_m) \qquad (5)$$

$$L_l = \begin{cases} L & \text{for} \quad y \leq 0 \\ 0 & \text{for} \quad y > 0 \end{cases} \qquad (6)$$

and

$$L_r = \begin{cases} 0 & \text{for} \quad y \leq 0 \\ L & \text{for} \quad y > 0 \end{cases} \qquad (7)$$

This separation into the specular halves of the lognormal spectrum will be used to analyze how the short-time and long-time processes contribute to the mechanical response.

It is known that the relationship between the spectrum L and J' can be written as (ref 5, Chapter 3)

$$J'(x) = J_g + \int_{-\infty}^{\infty} \frac{L(y)\mathrm{d}y}{1 + \exp[2(x + y)]} \qquad (8)$$

where $x = \log(\omega\tau_m)$, ω being the angular frequency. Likewise, the contributions of L_l and L_r lead to the partial compliances

$$J'_{(l,r)}(x) = J_g + \int_{-\infty}^{\infty} \frac{L_{(l,r)}(y)\mathrm{d}y}{1 + \exp[2(x + y)]} \qquad (9)$$

It is pointed out that J'_l and J'_r are assumed to have the same lower limit as J' in order to remark only the influence of L_l and L_r. Then,

$$J'_l + J'_r = J' + J_g \qquad (10)$$

The double-log representations of the dynamic compliances J', J'_l and J'_r are shown in Figure 1. From these curves it results that when $x \gg 0$, J' and J'_l are nearly parallel to the horizontal axis with a vertical difference of about log 2. Thus, the approximated spectra associated with both curves will be zero for retardation times longer than τ_m. Consequently, the double-log representation is not sensitive to differenciate between L and L_l because it expands the region where only the short-time mechanisms are involved and compresses the portion of the curve where these
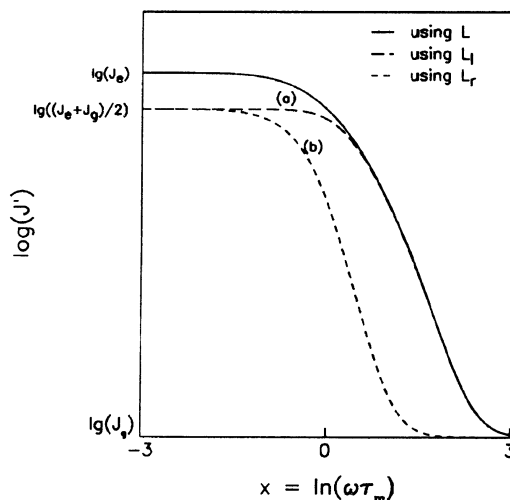

Figure 1. Double-log representation of the real component of the dynamic compliance: the full line corresponds to J', the dashed curves (a) and (b) refer to J'_l and J'_r, respectively.

two functions are different. In effect, a different response is found if the normalized component of the dynamic compliance is considered. Combining eq 1 and 8, the normalized function results

$$f(x) = \frac{1}{J_e - J_g} \int_{-\infty}^{\infty} \frac{L(y)\mathrm{d}y}{1 + \exp[2(x + y)]} \qquad (11)$$

Furthermore, the partial normalized functions f_l and f_r, defined replacing L by L_l and L_r respectively, verify that

$$f(x) = f_l(x) + f_r(x) \qquad (12)$$

for all values of x. These two functions f_l and f_r and also f are represented in Figure 2 showing that when $x \ll 0$,

$$f_l \approx 0.5 \qquad (13)$$

and accordingly to eq 12

$$f_r(x) \approx f(x) - 0.5 \qquad (14)$$

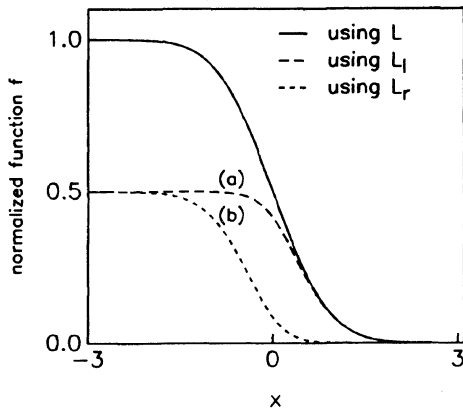while for $x \gg 0$

$$f_r(x) \approx 0 \qquad (15)$$

so that

Figure 2. Representation of the normalized real component of the dynamic compliance: the full line corresponds to f and the dashed lines (a) and (b) are the curves of f_l and f_r, respectively.
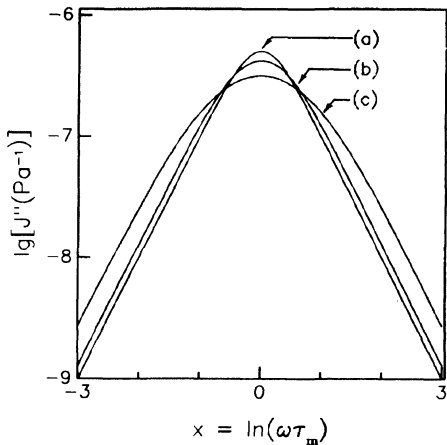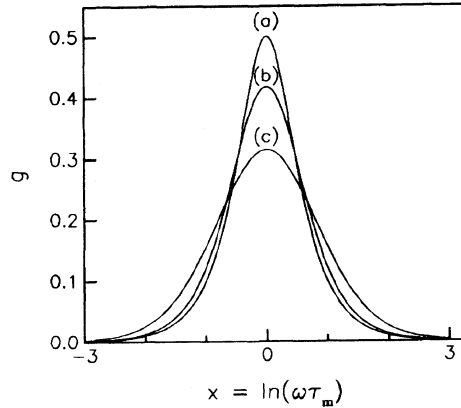

Figure 4. Nomalized representation of the imaginary component of the dynamic compliance. (a) a single mechanism, (b) and (c) two lognormal retardation spectra characterized by the mean time τ_m and half-widths $\beta = 1$ and $\beta = 2$, respectively.

Figure 3. Plot of log J'' against x, where $x = \ln(\omega\tau_m)$. (a) a single mechanism with retardation time τ_m, (b) and (c) two lognormal retardation spectra characterized by the mean time τ_m and half-widths $\beta = 1$ and $\beta = 2$, respectively.

$$f_l(x) \approx f(x) \qquad (16)$$

Then, on applying the first order approximation of L^1

$$L_1(y) = -(J_e - J_g)\frac{\mathrm{d}f(x)}{\mathrm{d}x}\bigg|_{x = -y} \qquad (17)$$

to eq 13 to 16, the approximated partial spectra L_{1r} and L_{11} result

$$L_{11} = \begin{cases} 0 & \text{for} \quad y \gg 0 \\ L_1 & \text{for} \quad y \ll 0 \end{cases} \qquad (18)$$

and

$$L_{1r} = \begin{cases} L_1 & \text{for} \quad y \gg 0 \\ 0 & \text{for} \quad y \ll 0 \end{cases} \qquad (19)$$

which are in accordance with the definitions of L_l and L_r.

Hence, through the approximation methods, f provides genuine information about the distribution of the times that characterize the viscoelastic mechanisms.

Analogously, the double-log plot of the imaginary component of any dynamic property does not provide the whole information about the spectrum. Figure 3 shows the curves log J'' vs. x corresponding to: (a) a single process characterized by a retardation time τ_m and (b), (c) two lognormal distribution functions with half-widths $\beta = 1$ and $\beta = 2$, respectively. It is clear that the approximated spectra calculated from these three curves will not be much different, particularly those determined from curves (b) and (c). However, the molecular mechanisms and the viscoelastic properties characterized by these three spectra are completely different. This can be shown in

Figure 4 where the normalized imaginary components of the dynamic compliances are plotted. The differences in the height and width at the half-maximum of the peaks are more evident because this plot suffers no distortion. As was indicated before, the log-log plot compresses the high values, that is, all the peaks are in a small fringe while the rest of the curve extends over several decades as J'' goes to zero.

Consequently, the normalized functions f and g are more suitable to determine the distribution functions through the approximate methods. Sometimes, however, the spectra can be determined analytically from the viscoelastic functions without making approximations. Also in this case, the normalized functions provide more information than the double-log plot. In fact, in previous papers,[8-11] procedures to determine if the spectrum associated with any mechanical property is lognormal and to find its parameters, were given. These procedures are based on recurrency relationships and special values of the normalized real component of a dynamic function. For instance, if the normalized real component of the dynamic function verifies that

$$f(x) = 1 - f(-x) \qquad (20)$$

then, it can be affirmed that the spectrum is symmetrical. Furthermore, since its inflection point is characterized by an ordinate value of 0.5 at $\omega = \tau_m^{-1}$, the value of τ_m can be determined. Finally, from the recurrency relationships of f, it can be established whether the distribution is lognormal and, in that case, what is the value of the parameter β.[8] Applying the same procedure to the double-log representation, however, no accurate results can be obtained. In effect, even if the spectrum is symmetrical this plot cannot be characterized by a recurrency relationship analogous to eq 20. Furthermore, the inflection point, identified only by its ordinate $\log V^*$, would fall in the compressed region of the curve. Thus, the mean time of the distribution would be determined

with much more error. Moreover, though the recurrency equations valid for f^{11} can be expressed in terms of the logarithm of the viscoelastic function, the distortion of the curve leads to a less accurate value of β.

The analytical determination of a spectrum can also be obtained from the imaginary component of the dynamic modulus or compliance. In this case, a symmetrical peak is related to a symmetrical spectrum. This spectrum is characterized by a mean time that corresponds to the inverse of the frequency at the maximum of the peak. Furthermore, from the recurrency relationships it can be established whether the spectrum is lognormal. If that is the case, as the parameter β is calculated from the width of the peak at its half-maximum,[8] the normalized representation is better to use. In fact, in this representation the value of β results from the width of the peak at 50% of its height while in the double-log plot this width corresponds to a peak decrease not greater than 5%. This introduces much more error in the determination of the spectrum as was pointed out before by a qualitative analysis of Figure 3.

APPLICATIONS

The procedure based on the normalized viscoelastic functions, developed to determine the spectrum associated with a certain mechanical property, will be applied to measurements of the dynamic compliance of polyisobutylene. Generally the measurements are represented in a double-log plot without giving a table of data. In this case, the distortion of the double-log representation discussed before makes it difficult to recalculate the viscoelastic functions including only the experimental error. Fortunately, Fitzgerald et al.[12] measured the frequency dependence of J' and J'', and tabulated the data. The measurements are normalized using eq 1 and 2 with $V = J'$ and J'', respectively, $V_1 = 1.1 \times 10^{-9} \, \text{Pa}^{-1}$, $V_m = 3.1 \times 10^{-6} \, \text{Pa}^{-1}$, and the corresponding fre-
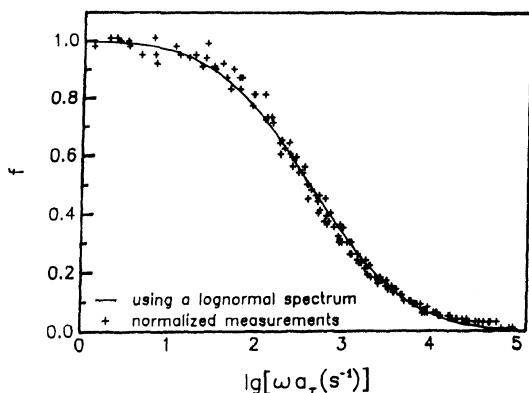
Figure 5. Master curve of the real component of the dynamic compliance of polyisobutylene. The crosses (+) are data measured by Fitzgerald et al.[12] reduced to 298 K using the translation paths given in ref 13. The full line corresponds to a lognormal retardation spectrum characterized by $\log \tau_m = -2.6$ and $\beta = 2.7$.
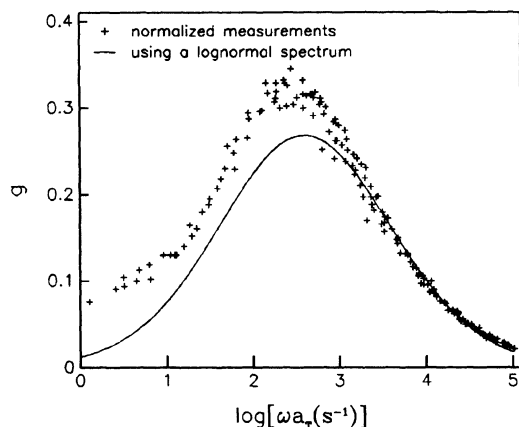

Figure 6. Master curve of the normalized imaginary component of the dynamic compliance of polyisobutylene. The crosses (+) are the measurements done by Fitzgerald et al.[12] shifted according to ref 13. The full line is the function g calculated using a lognormal retardation spectrum with $\log \tau_m = -2.6$ and $\beta = 2.7$.
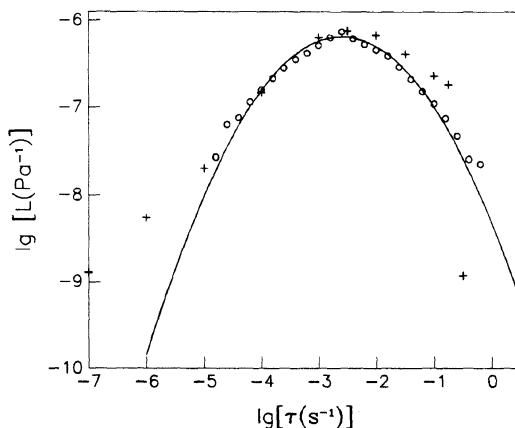

Figure 7. Retardation distribution function of polyisobutylene: the full line is a lognormal function characterized by $\log \tau_m = -2.6$ and $\beta = 2.7$; the first order approximated spectra determined from the normalized curve and the double-log plot are represented by the open circles (○) and the crosses (+), respectively.

quencies are reduced using the translation paths given ref 13. In this way, the master curves of f and g are determined from the normalized data up to $\log \omega = 5$ and represented with crosses in Figures 5 and 6, respectively. It should be noticed that the measured values of f and g exhibit a great dispersion at low angular frequencies. Even in

this case, from the special values of f and its recurrency relationships, a description of the retardation spectrum is possible. At first, the angular frequency at the inflection point of f, ω^*, is determined. Then, on defining the variable $x = \log(\omega/\omega^*)$, it can be shown that f verifies eq 20. Hence, the distribution function associated with f is symmetrical, being characterized by a mean time $\tau_m = (\omega^*)^{-1}$. Furthermore, f satisfies the recurrency equations of a lognormal distribution function and, from these equations the value of β can be obtained. This procedure leads to a lognormal retardation spectrum with $\log \tau_m = -2.6$ and $\beta = 2.7$. The function

$$f(x) = \int_{-\infty}^{\infty} \frac{\exp(-w^2)dw}{1 + \exp[2(x + \beta w)]} \qquad (21)$$

calculated using the parameters given before provides a good fitting to the normalized measurements as shown in Figure 5. Therefore, the lognormal spectrum gives a symmetrical contribution to the mechanical behavior of any process with retardation time τ, that is, not only when $\tau < \tau_m$ but also when $\tau > \tau_m$. The spectrum determined from the double-log plot,

however, emphasises the influence of the short-time mechanisms, falling sharply to zero at times longer than τ_m. This comparison is shown in Figure 7. The spectrum can also be calculated using the first order approximation of the normalized curve of f. Applying eq 17 to the reduced curve shown in Figure 5, the approximated spectrum is obtained, being represented in Figure 7 by the open circles. This spectrum also is a symmetrical function centered at the same mean time τ_m though it is larger and, consequently, lower than the real distribution. Therefore, the normalized real component of the dynamic compliance leads not only analytically but also through an approximation method, to a symmetrical spectrum. This is another proof of the distortion not only of the viscoelastic functions but also of the distribution functions determined from the double-log representation of any mechanical property.

DISCUSSION

Even when a spectrum gives information of the viscoelastic behavior of a polymeric system, it does not provide a direct description of the molecular mechanisms. In some cases, however, according to the shape of the spectrum, it can be established a certain mechanical model which molecular parameters are adjusted according to the distribution function of the characteristic times. For instance, a wedge retardation spectrum characterized by a slope of 0.5 in a double-log plot may be associated with the molecular model of Rouse.[14] This model, originally applied to diluted polymer solutions, was subsequently extended to undiluted polymers just by finding log–log plots of relaxation or retardation spectra characterized by a straight line with a slope of -0.5 or 0.5, respectively. Particularly, Catsiff and Tobolsky[16] determined the approximate relaxation spectrum of polyisobutylene (PIB) from the double-log plot of the relaxation modulus and assumed that it could be

represented by a wedge-shaped function with a slope of 0.5. Furthermore, on considering the approximated expression for the interconversion formula (ref 15, p 96), they determined that the retardation spectrum was also wedge-shaped, but with a slope -0.5. Therefore, they concluded that the viscoelastic behavior of PIB might be explained by the molecular mechanisms of Rouse's theory. However, in this paper it has been demonstrated that the distribution function of the characteristic times of this material cuts sharply just because of the distortion of the double-log representation of the mechanical properties. The normalized representation of the real component of the dynamic compliance, however, leads to a retardation spectrum of PIB that can be described by a lognormal distribution function. This symmetrical distribution is defined for all the retardation times and exhibits no terminal time as Rouse's theory do. Furthermore, in a double-log plot the lognormal distribution function is represented by a parabola which can be approximated by a straight line of slope 0.5 only in a very short interval of retardation times. Hence, using a distorted representation of any viscoelastic function leads to interpret the mechanical behavior of a polymeric structure in terms of an incorrect model.

It should be pointed out that the normalized master curves must be considered with care because they can also lead to inconsistencies originated from the time-temperature superposition. Particularly these inconsistencies appear more clearly in the plots of the normalized imaginary component of the dynamic functions. For instance, going back to the example treated in the Applications, the normalized measurements of J'' are calculated according to eq 2 and shifted using the translation paths proposed in the original reference,[13] leading to the master curve shown in Figure 6. This figure shows that the fitting to the normalized measurements of J'' is not good neither in the left portion of the peak nor

at its maximum and only the right side is fairly well approximated. This difference between the data and the predicted shape of g is originated from the construction of the master curve. In fact, in a previous publication,[17] it has been demonstrated that the time-temperature super-position is not strictly obeyed and that the matching of two segments of curves measured at different temperatures is not characterized by an unique translation path but by a mean path determined subjectively. This means that two curves can be superposed fairly well if their central regions match properly, even when their extremes do not superpose at all. Particularly this is noticed in the upper limit of the master curve of f, shown in Figure 5, where the dispersion of normalized data reveals that all the curves measured at different temperatures have not the same equilibrium compliance J_e. In the plot of g, the difference between the individual curves increases because of their sharper shapes. Particularly the spreading of points near the maximum of the peak, hidden by the double-log plot, is due to the different limiting values of the individual curves. However, to construct the master curve it is assumed that all the curves have the same limits, hence the normalized representation of J'' points out another discrepancy of the time-temperature superposition. Now, even if the limits are assumed to be independent of temperature, the different widths of the in-dividual peaks makes difficult their matching through horizontal displacements. Conse-quently, a criterion to construct the master curve of g may be to shift the curves parallel to the horizontal axis until the right branches match. This seems to be the way used by Ferry et al.[13] to determine the shift factors corresponding to the individual curves of J''. Therefore, analogously to the matching of the individual segments of the f function, this "master peak" provides information about an average behavior of the system where it is assumed that the limiting moduli or compli-ances do not depend on temperature. Hence,

the master curve of g might be described by the same values of τ_m and β as the ones determined from the master curve of f. The expression used to calculate g considering a lognormal spectrum is[11]

$$g(x) = \frac{1}{2\sqrt{\pi}} \int_{-\infty}^{\infty} \exp(-w^2) \cosh^{-1}(x + \beta w) dw$$

(22)

leading to the curve represented in Figure 6. From this figure it is clear that the curve do not approximate the experimental points neither in height nor in width, that is, the parameters of the lognormal distribution do not characterize the master curve of all the dynamical properties.

Nevertheless, τ_m and β can also be calculated from some individual curves. In fact, in a previous paper[11] it has been shown that if the function g is symmetrical with respect to its maximum, then the spectrum is also symme-trical. Furthermore, if the inflection point of f and the maximum of g occurs at the same frequency then, the mean time of the spectrum is the reciprocal of that frequency. Finally, considering the mathematical properties of the functions f and g corresponding to a lognormal distribution, the value of β can be determined from the intersection point of curves f and g. Therefore, this procedure cannot be applied to the individual curves measured at any tempera-ture but only to those that exhibit the inflection point of f and simultaneously the maximum of g. In the set of curves of PIB, the individual curves of J' and J'' measured at 313 K and 323 K show the inflection point and the maximum of the peak, respectively. Then, con-sidering the recurrency relationships, a log-normal spectrum characterized by the same average characteristic time $\log \tau_m = -2.6$ is obtained for both curves. Meanwhile the half-widths of both distributions reduce to $\beta = 2.1$ because the dispersion of the right branch of the master curve leads to a peak wider than any of the individual peaks whose
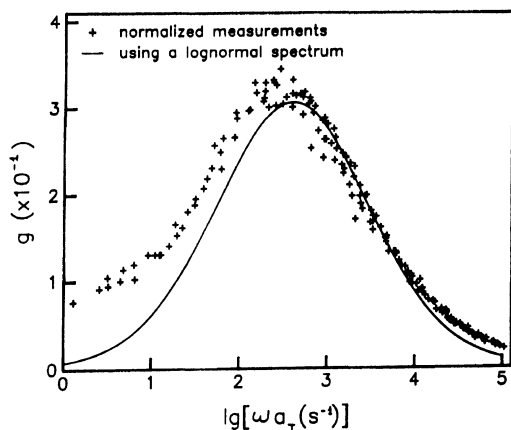
Figure 8. Reduced data of the imaginary component of the dynamic compliance of polyisobutylene[12,13] approximated by the normalized g function associated with a lognormal spectrum characterized by $\log \tau_m = -2.6$ and $\beta = 2.1$.

Figure 9. Reduced data of the real part of the dynamic compliance of polyisobutylene[12,13] fitted by a lognormal spectrum with parameters: $\log \tau_m = -2.6$ and $\beta = 2.1$.

widths can be measured. Then, on considering the new value of β, eq 22 gives the normalized function shown in Figure 8, providing a better fitting to the data even at the maximum where the points are largely spread out. Moreover, if the new value of β is used in eq 21 to calculate the normalized function f, the curve represented in Figure 9 is obtained. This function approximates the master curve of f fairly well in the medium region, that is, near the inflection points of the individual curves used to determine the parameters of the distribution function. Hence, the new value of β can be considered a good parameter to characterize the lognormal distribution. However, this parameter implicitly includes the assumption that the instantaneous and equilibrium moduli or compliances do not depend on temperature. Consequently, though the double-log plot considerably distorts the information about the viscoelastic mechanisms, the normalized master curves might also lead to inaccurate information about the spectra.

CONCLUSIONS

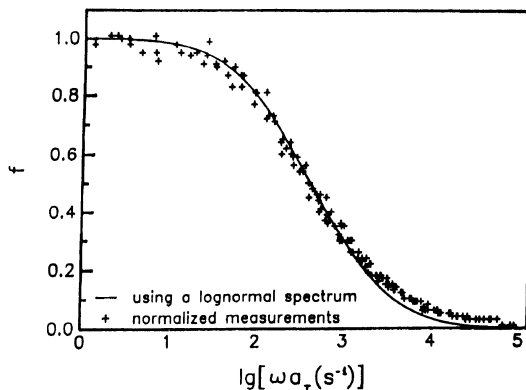Because of the importance of distribution functions, the concepts related to the determi-

nation and interpretation of the spectra must be critically reviewed, particularly the incompletely information provided by the log–log plots of viscoelastic functions. The distortion in these plots, produced by the logarithm leads to molecular models that do not describe the polymeric structure properly. The normalized functions, however, provide a better approach to the viscoelastic behavior. In fact, the distribution functions calculated using approximation methods, or from the recurrency relationships of the normalized functions, lead. to a fairly good fitting to the normalized measurements. This fitting to the data also depends on the translation of the individual curves, that is, on the time-temperature superposition. In effect, the normalized mechanical property calculated from the approximated spectrum does not adjust directly to the measured values but to values shifted parallel to the horizontal axis, according to certain translation paths. Then, when the time-temperature superposition is not strictly obeyed, because of the smooth shape of the transient properties and the real component of the dynamical properties, an apparent matching of the individual segments is found. On the contrary, the imaginary components lead to less defined master curves

due to their sharper shapes. Consequently, the procedure developed in this paper to calculate the distribution functions provides better results when it is applied to the normalized representation of the quasi-static properties on the real components of the dynamic functions. Anyway, the spectrum derived by the fitting to the master curves establishes implicitly that the limits of each individual curve do not depend on temperature. Therefore, a more accurate spectrum can be obtained if the normalization procedure is applied directly to an individual curve of any mechanical property measured over several orders of magnitude of time or frequency. In this way, the mechanical response will lead to a molecular model that interprets the viscoelastic structure adequately.

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