Influence of plasticizer on Poisson's ratio of epoxy polymers

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A series of simultaneous isothermal tensile creep measurements of the longitudinal extension and the lateral contraction in standard thin tension specimens yielded individual curves for the variation of the lateral contraction ratio (Poisson's ratio) at different loading and temperature steps throughout the viscoelastic spectrum for a series of pure and plasticized cold-setting epoxy polymers. By applying the principle of reduced variables and shifting the isothermal creep curves along the log(time) scale, composite curves were obtained for Poisson's ratio which have the shape of sigmoid curves, thus proving the dependence of this viscoelastic function on time and temperature. It was shown that the corresponding transition regions were shifted towards the origin ($t = 0$) as the amount of plasticizer was increased, while the shape of the corresponding composite curves did not change and the variation of the logarithmic time factor remained essentially the same but with steeper gradients for increasing amounts of plasticizer. Comparison of the viscoelastic spectra for this function with the corresponding spectra for creep extension, shear and bulk compliances found previously indicated that the spectrum of this function is broader than the others and while it starts to appear earlier than the shear compliance spectrum it disappears after the corresponding bulk compliance spectrum. Finally, glass transition temperatures for the various mixtures were found to be lower by 5° to 10°C than those obtained from measurements using a thermal analyser.

INTRODUCTION

Since the ratio of the negative lateral contraction to the longitudinal extension in a simple tension test is defined as Poisson's ratio (constant) it is preferable to call the corresponding viscoelastic function the lateral contraction ratio (*LCR*). Unlike the other viscoelastic functions (moduli and compliances) it cannot be rigorously defined on thermodynamic grounds that the *LCR* is a monotonically varying function with time, temperature and frequency.

Schwarzl[1] has introduced an expression for the relaxation *LCR* function in the form of an integrodifferential equation given by:

$$\epsilon_y(t) = -\int_0^t \nu_r(t-\tau)\dot{\epsilon}_x(\tau)\mathrm{d}\tau \qquad (1)$$

where $\nu_r(t)$ expresses the *LCR* function in relaxation and $\epsilon_y(t)$ is the lateral contraction, which is a function of time for constant longitudinal extension ϵ_{x0}, suddenly applied at time $t = t_0$. Schwarzl argued that since the lateral deformation in tension must be a contraction and the volume dilatation must be positive the *LCR* function must vary between $0 < \nu(t) \leqslant 0.5$.

Freudenthal and Henry[2] in a theoretical study of typical viscoelastic models found that for Maxwell and four-parameter bodies *LCR* functions must vary between 0.33 and 0.50 which is in agreement with experimental values for Poisson's ratio, while a Kelvin body and the standard solid present unrealistic limits for Poisson's ratio which are between $-\infty$ and 0.33 for the former and 0.20 and 0.33 for the latter. All these limits were defined for a solid on the assumption that $3K = 8G$. Gottenberg and Christensen[3] studied the variation of the complex Poisson ratio for the special cases where either one or both of G' or G'' assume finite non-zero values, while K' or K'' tend to zero. They found that the real part, ν', of Poisson's ratio varies between -1.0 and 0.5 and the imaginary part ν'' takes positive or negative values. The positive values for ν'' are objectionable, since they assume that $K''/K' > G''/G'$, which is in contradiction to extensive experimental evidence in crosslinked polymers[4]. A similar study by Rigbi[5] examined the cases where either $\nu'' = 0$ or $K'' = 0$, which both are extreme cases of no practical value.

Waterman in a series of two papers[6,7] measured the complex moduli and the *LCR* functions for different polymers. It can be derived from this study that the real parts of Poisson's ratio are always positive, while the imaginary parts are negative so that there is always a lag in the phase diagram between the applied longitudinal strain and the complex Poisson ratio. Furthermore, it was shown that the complex Poisson ratio is a monotonically increasing function with temperature. These results are in complete agreement with theoretical considerations of mechanical behaviour[1].

Experimental evaluation of the *LCR* in the glass transition region for PMMA and PVC by Koppelmann[8] who measured the shear and bulk moduli in this region gave a negative minimum at this area. This paradoxical result was explained by Heydemann[9] who attributed it to the non-linear variation of the free volume in the vicinity of the glass transition. Many other experimental evaluations of Poisson's ratio were executed over restricted parts of the viscoelastic spectrum; constant values (mainly at the glassy region) or values increasing with time, temperature and the inverse of frequency were

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obtained. A brief review of these papers may be found in ref 4.

Theocaris and coworkers[10–15] explicitly defined the transient *LCR* in creep and relaxation and gave their viscoelastic operators in integral form. Furthermore, the transient *LCR* function was measured for a series of polymers over the whole viscoelastic range by applying the principle of reduced variables. The main results of this experimental research showed that *LCR* functions in creep and relaxation are monotonic with time and temperature and the creep *LCR* advances in time to the corresponding relaxation function, the inverse of which convolutes with the respective creep function. However, the difference between the two functions in the transition zone is much less than the differences between the other viscoelastic functions.

In this paper a series of creep experiments in pure tension were undertaken for a series of unplasticized and plasticized epoxy polymers in order to reveal the influence of the amount of plasticizer on the viscoelastic properties of the polymer.

DEFINITION OF POISSON'S RATIO FUNCTIONS

While the Poisson's ratio function in relaxation was given by Schwarzl[1] and expressed by equation (1), the corresponding function in creep may be expressed by:

$$\epsilon_y(t) = -\int_0^t \nu_c(t-\tau)D(t-\tau)\dot{\sigma}_x(\tau)\mathrm{d}\tau \qquad (2)$$

where $\nu_c(t)$ expresses the Poisson's ratio function in creep and $D(t)$ the corresponding creep compliance in pure tension.

Between $\nu_c(t)$ and $\nu_r(t)$ functions the convolution integral holds:

$$\int_0^t \nu_r(t-\tau)\nu_c^{-1}(\tau)\mathrm{d}\tau = 1 \qquad (3)$$

which indicates that in the physical plane the following inequality holds:

$$\nu_r(t)/\nu_c(t) \leqslant 1 \qquad (4)$$

However, in the Laplace transformed plane with parameter p, the inequality (4) becomes an equality, i.e.:

$$\bar{\nu}_r(t)/\bar{\nu}_c(t) = 1$$

In all these relations the applied stresses or strains are considered as step functions, which are zero for negative arguments and positive for non-negative arguments. Thus, the lower limit in all integrals must be understood as (0_+).

The *LCR* functions in creep and relaxation are interrelated with any of the creep compliances or relaxation moduli functions which are given in refs 15 and 16.

Besides the transient *LCR* functions in creep and relaxation the complex *LCR* function derived under the influence of a dynamic loading is expressed by:

$$\nu^* = \nu_c^* = \nu_r^* = \nu_c' - i\nu_c'' = \nu_r' - i\nu_r'' \qquad (5)$$

where the quantities with asterisks correspond to the complex values for ν, while the primes express the storage parts and the double primes the loss parts. It has been shown[4,6,7] that the loss quantities $\nu_c'' = \nu_r''$ are always positive and therefore the Poisson's ratio function always lags the applied stress or strain vector in a phase diagram in creep or relaxation.

EXPERIMENTAL

The family of high polymers studied in this paper was produced from a pure epoxy prepolymer to which different amounts of plasticizer were added. The unplasticized samples were prepared from Shell Epon 828 cold-setting epoxy polymer to which an 8% triethylenetetramine hardener was added. The plasticized samples were prepared from the same epoxy polymer, mixed with different amounts of plasticizer; finally the same amount of hardener was added to the mixture. Thiokol LP3 polysulphide copolymer was used as plasticizer. The quantities of plasticizer added were increased in 10% steps, from 10 up to 40% by wt of the epoxy copolymer.

The epoxy–plasticizer compositions were thoroughly mixed in an open cup; the hardener was subsequently added and thoroughly mixed. The preparation was then deaerated in a vacuum chamber for 20 to 30 min to avoid formation of air bubbles and subsequently cast into moulds in the form of square sheets of thickness varying between 4.90 and 5.10 mm. The setting and curing of the plates has been described elsewhere[17], using maximum temperatures depending on the rubbery state temperature corresponding to each plasticized mixture.

Indeed, the curing temperatures, T_c, for each polymer formulation were taken to be 20° to 30°C above the glass transition temperature T_g for the mixture. Since the values for the glass transition temperatures of these materials were approximately known from previous tests[18] to be in the range between 120° and 60°C the corresponding cure temperatures for the various polymers were selected: T_{c_0} = 140°; $T_{c_{10}}$ = 130°; $T_{c_{20}}$ = 110°; $T_{c_{30}}$ = 95°; and $T_{c_{40}}$ = 90°C.

Since the rubbery plateau temperatures depend directly on the corresponding T_g selection of curing temperatures above T_g by 20° to 30°C resulted in values of T_c very close to the corresponding rubbery plateau temperatures for each mixture and this had, as a consequence, a uniform heat treatment for each polymeric substance.

The curing procedure applied may be described as follows. The temperature was first raised by 5°C h^{-1} from room temperature up to the corresponding T_c for the substance and then this maximum temperature was maintained for 72 h. The constant temperature plateau was followed by a progressive decrease of temperature at a rate of 1°C h^{-1} back to ambient.

This fast curing cycle was followed by a second of 1 month duration at ambient temperature. In this way stable and stress-free materials with mechanical and optical repeatable properties were obtained for each type of polymer. Otherwise, the properties of the polymeric materials tested may vary with time and from batch to batch not only due to the time-dependent viscoelastic behaviour, but also because of crosslinking. Each polymeric sample was designated by three numbers in sequence, each of the two first indicating the wt % epoxy prepolymer of either the epoxy polymer, or the polysulphide plasticizer. The amount of hardener is subsequently included, although it remained constant for all mixtures and was sufficient to give a smooth and satisfactory polymerization.

Simple tensile specimens were cut from each completely

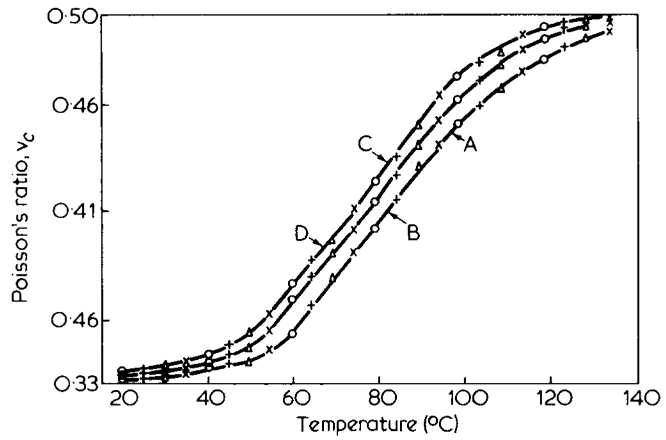

Figure 1 Variation of lateral contraction ratio (Poisson's ratio) *versus* temperature in creep at three different time intervals t = 1/4 min, 4 min and 32 min for an unplasticized epoxy polymer (C-100-0-8). A, t = 1/4 min; B, t = 4 min; C, t = 32 min

cured plate with the following dimensions: width, b, between 41.90 and 42.10 mm and length along the reduced section of the tension specimens l equal to 200 mm. All specimens were tested on a photoelastic bench before viscoelastic testing, in order to ensure that their shape and the preparation procedure did not introduce residual stresses at the reduced section, so that pure tension viscoelastic tests can subsequently be executed.

The testing equipment was simple. An electric furnace with temperature-controlling equipment was used as the environmental chamber. The front and back of the furnace were covered with glass plates especially annealed and having parallel lateral faces, in order to minimize any distortion of the light rays passing through the plates and to avoid any error in the measurement of strains by the projected moiré image method[17]. Furthermore, any moisture was excluded from the furnace, while the temperature level was kept constant at the desired temperature by means of automatic thermoregulators capable of keeping temperature constant for long periods when the system was in equilibrium with an accuracy of ±0.2°C up to temperatures of 150°C.

The loading system consisted of a simple lever device with dead weights added outside the furnace, which assured an instantaneous, but not hammering loading and unloading of the specimens. The loading process consisted of tensile creep tests for periods of 32 min. Measurements of longitudinal extension and lateral contraction were executed at equal time intervals on a logarithmic scale from 1/4 min up to 32 min at each temperature step. The loads applied at each test were small and care was taken not to create excessive deformation beyond the linear behaviour of the material. Furthermore, as the temperature of the test increased, the applied load was decreased according to a sigmoid curve in order to compensate for the increase of the creep compliance of the material and to minimize the effect of the lateral contraction on the assumed constancy of the applied stress within the linear domain of the material.

The amount of longitudinal strain applied in each test varied between 3 x 10^3 and 1.5 x 10^3 micro-strain for substances between C-100-0-8 and C-100-40-8 for the tests of ambient temperature (t = 25°C). As the temperature increased in the isothermal tests and the corresponding moduli reduced, the applied loads were reduced accordingly, so that the amount of deformation in each specimen remained both elastic and the same.

All the data were measured at least three times and the reproducibility of strains was checked to be satisfactory. Since the substances tested did not flow in the range of linear viscoelastic behaviour single specimens for each type of material were used over the whole temperature range, thus eliminating errors due to the variability of material in different specimens. Moreover, each specimen after the end of each loading step was left to recover completely before the execution of the next test while the rise in temperature facilitated and speeded up the recovery of the specimen.

The projected moiré image method was used for the measurement of strains. However, for greater accuracy, each specimen was equipped with two identical strain gauges of special type, which were both cemented at the back of the specimens and at places in the reduced section not interfering with the moiré measurements. One gauge was oriented along the longitudinal axis of the specimens thus measuring the longitudinal extension, while the second had its axis along a transverse section of the specimen, thus measuring the lateral contraction.

For the optical measurement of both strains, crossed line gratings were photoprinted on the front surface of specimens with densities varying between 8 and 20 lines/mm with their principal axes parallel to the principal axes of the undeformed specimen. A point source of light and a system of collimating lenses created a projection of the specimen grating with a one to one magnification on a ground-glass screen outside the furnace. Superposition of a second identical grating with the specimen grating (the reference grating) formed an initial moiré pattern by the interference of the two gratings in contact. By adjusting the optical apparatus it was possible to annul the initial moiré pattern with the specimen unloaded at the time when thermal equilibrium was achieved. Thus, it was possible, at each temperature level, to exclude the influence of the thermal expansion of the specimen and to measure directly the components of strain from the moiré pattern corresponding at each time station. The evaluation of strains from the displacement curves given by the two simultaneous moiré patterns was easy since the strain field was uniform and a large gauge length was selected.

The values of strains derived from the moiré patterns were always in satisfactory agreement with the corresponding values from the strain-gauge measurements, the discrepancies between relative values being of the order of 1–3%, especially at small values of strains.

RESULTS

Isothermal creep tests were carried out at various stages from ambient temperature up to the rubbery state for each composition. A temperature of 25°C (298K) was taken as a reference temperature for plotting the composite curve. Moreover, the instantaneous measurements of Poisson's ratio at a specified time instant (t_m = 15 sec) for each temperature level gave the *LCR* (Poisson's) function in terms of temperature for tensile creep. The time interval of 15 sec after the abrupt application of the load was selected so as to exclude inertia effects due to loading.

The curves of Poisson's ratio *versus* temperature at three different time intervals t_1 = 1/4 min, t_2 = 4 min and t_3 = 32 min for composition C-100-0-8 are plotted in *Figure 1*. All three composite curves are of sigmoid type and each curve lags in time relative to any other curve corresponding to a longer time. However, all three curves present the same shape. It is therefore sufficient to compare the *LCR* (Poisson)

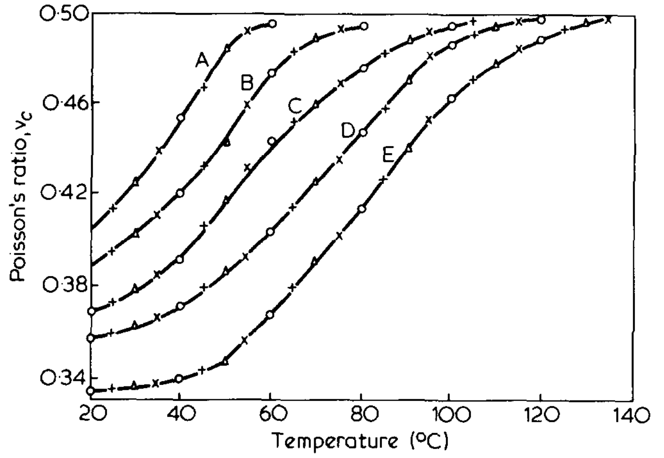

Figure 2 Variation of lateral contraction ratio (Poisson's ratio) *versus* temperature in creep for a series of plasticized epoxy polymers; amount of plasticizer varies between 0 and 40% of the epoxy prepolymer. A, C-100-40-8; B, C-100-30-8; C, C-100-20-8; D, C-100-10-8; E, C-100-0-8; $\nu_c = f(T)$, $t = 4$ min

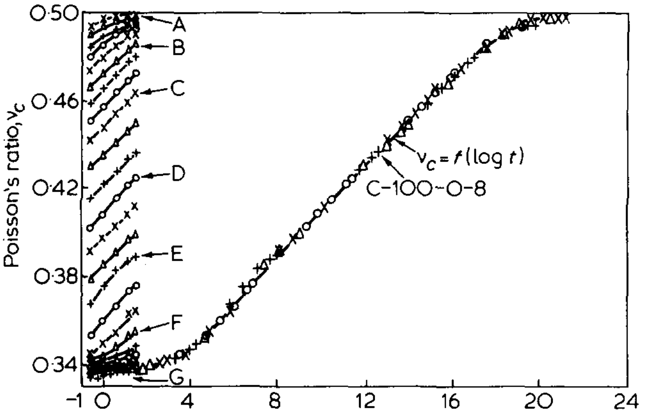

Figure 3 Individual isothermal creep curves and master or composite curve of the variation of lateral contraction ratio $\nu_c(\log t)$ in creep *versus* log(time) for an unplasticized epoxy polymer (C-100-0-8)

ratio behaviour for each composition with respect to temperature by plotting only one representative curve for each composition. The curves selected were those corresponding to the time interval $t = 4$ min.

Figure 2 presents the variation of Poisson's ratio for all compositions of epoxy polymers *versus* temperature (°C). From this Figure it may be seen that addition of plasticizer in the epoxy polymer results in a shifting of the Poisson's ratio *versus* temperature curves to lower temperatures.

Although the amount of shifting is approximately constant for constant amounts of plasticizer added to the epoxy polymer, there also exists a small, but perceptible, increase in the steepness of the respective curves as the amount of plasticizer is increased.

Figures 3 to *5* present the individual isothermal curves of variation of Poisson's ratio in creep (ν_c) *versus* log (time) (min) from 1/4 to 32 min for various steps of temperature and various amounts of plasticizer, starting from 0 up to 40%. 25°C was taken as reference temperature; this temperature was close to the ambient temperature of the tests and easily achievable. The composite (or master) curves of each composition were plotted in the same Figures by applying the time–temperature superposition principle. The validity of the principle was assured by applying the appropriate amount of stress at each step of temperature, so that the material always remained in the elastic region. Previous experimental evidence with the same materials for the evaluation of the transient mechanical creep and relaxation behaviour[12] [creep compliance and relaxation modulus in extension *versus* log(time)] yielded the limits of validity of the time–temperature superposition principle for the evaluation of the lateral contraction ratio in creep. However, in cases where doubts arose immediate application of proportional loading steps yielded similar creep curves for $\nu_c = f(\log t)$ with the respective multiplication factors.

The composite curves of each individual composition were juxtaposed in *Figure 6*, in order to evaluate the relative position of the master curves of all compositions. It can be derived from the comparison of these master curves that the softer or more plasticized materials may be considered as presenting composite or master curves, which resulted from the same standard curve, which is the curve corresponding to the unplasticized material, by a horizontal shifting of each curve along the log(time) scale. Addition of plasticizer corresponded to a shift of the respective curves toward

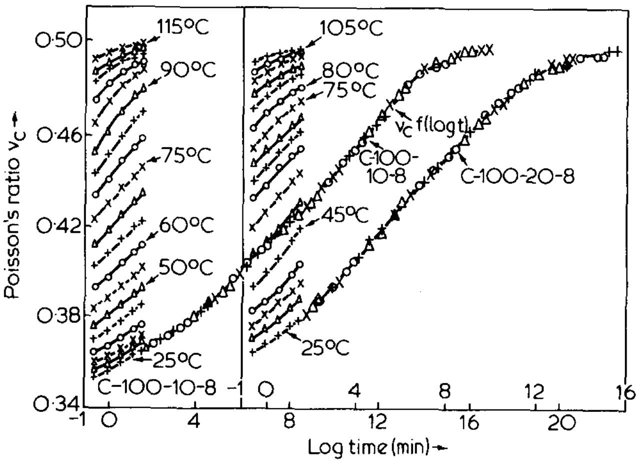

Figure 4 Individual isothermal creep curves and master or composite curves of the variation of lateral contraction ratio $\nu_c(\log t)$ in creep *versus* log(time) for two types of plasticized epoxy polymers with amounts of plasticizer equal to 10 and 20%, respectively to the epoxy prepolymer (C-100-10-8, C-100-20-8)

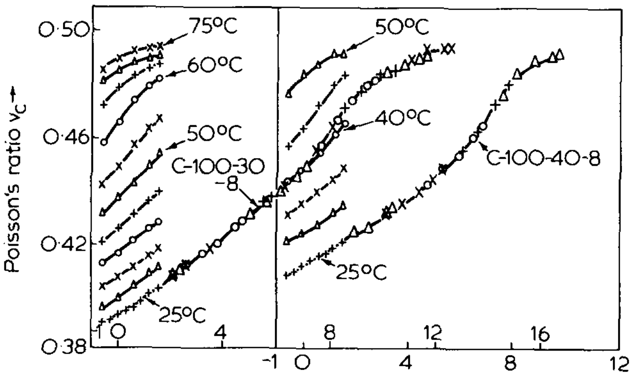

Figure 5 Individual isothermal creep curves and master or composite curves of the variation of lateral contraction ratio $\nu_c(\log t)$ in creep *versus* log(time) for two types of plasticized epoxy polymers with amounts of plasticizer equal to 30 and 40%, respectively, to the epoxy prepolymer (C-100-30-8, C-100-40-8)

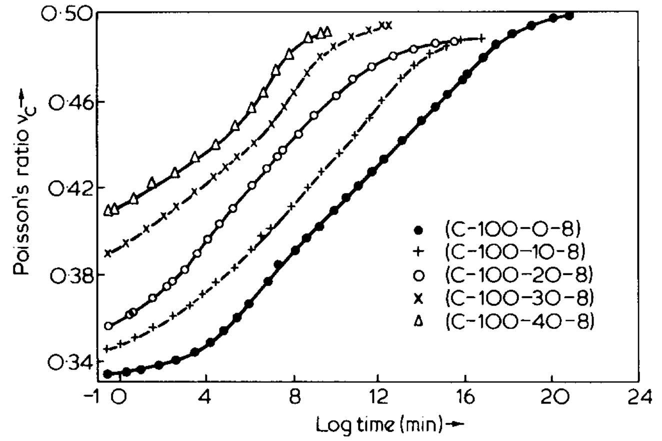

Figure 6 All five previous master or composite curves of the variation of the lateral contraction ratio (Poisson's ratio) in creep *versus* log(time)

shorter times along the abscissae. This means that if these materials were tested at temperatures lower than the reference temperature of 25°C, they would yield the missing parts of the master curves, which lie at shorter times. Otherwise, the plasticized materials behave at room temperature as if they were already in their transition region and it depends on the amount of plasticizer added how far they are in their glassy or transition regions.

This coincidence between the composite curves of the various substances counterbalances the lack of information of viscoelastic behaviour of highly plasticized substances, for which only parts of their composite curves appear in *Figures 4* to *6* for temperatures above 25°C. The missing glassy regions of these curves may be approximately replaced by values taken from the respective curves with smaller amounts of plasticizer, which present broader regions around T_g for ranges of temperature above the ambient.

However, it can be derived from *Figure 6* that, while the master curves for Poisson's ratio *versus* log(time) are all of a similar shape, the shift between successive curves with the same amount of plasticizer diminishes as the amount of plasticizer is increased. Besides this decrease in the shifting of the composite curves, there is also a slight increase of the slope of these curves as the amount of plasticizer increases.

These phenomena could be explained by the fact that, although equal amounts of plasticizer were added to each previous mixture to obtain the subsequent mixture for the same amount of epoxy prepolymer, these amounts of plasticizer corresponded to weight fractions of the corresponding mixture equal to 8.5, 15.6, 21.7 and 27%, respectively, for the four plasticized mixtures. This decreasing difference in weight fractions of the plasticizer in each polymeric mixture may be the main cause of the decrease of the shifting of the composite curves.

Figure 7 presents the variation of the time factor log a_T *versus* temperature for the various mixtures of epoxy polymers tested as it has been derived by plotting each composite curve from the corresponding individual curves. The time factor loga_T is defined as the amount of shifting in the log(time) scale of the individual isothermal creep curves to construct the respective composite curves. It can be derived from this plot that although at lower temperatures the values of time factor intertwine, curves of loga_T *versus* temperature for a higher percentage of plasticizer present a progressively increasing steepness and they seem to belong in succession to parts of the basic curve corresponding to the unplasticized material which lie always at the one extreme of the curve corresponding to high temperatures.

A comparison of the exact positions and forms of the composite curves for the lateral contraction ratio with the corresponding composite curves for the other characteristic functions of the viscoelastic behaviour of this substance (shear, extension and bulk compliances in creep) indicates that the viscoelastic retardation spectrum l_k in creep for the lateral contraction ratio is always in advance with respect to the other spectra. *Figure 8* presents the higher order approximations of the retardation spectra for the unplasticized epoxy polymer. The extension retardation spectrum was taken from ref 18, while the shear and bulk retardation spectra were taken from refs 15 and 19 and the approximation for the lateral contraction ratio retardation spectrum was evaluated by applying well-known approximation methods.

From these spectra it can be derived that:

(i) the transition regions in retardation appear in the order: lateral contraction ratio, bulk, extension and finally shear compliances. They terminate in the same order: lateral contraction ratio, bulk, extension and shear;

(ii) the broadest transition region appears in the lateral contraction ratio function and the shortest for the shear compliance;

(iii) the inflexion points of these spectra where their maxima appear are presented in the same order as the appearance of retardation spectra;

(iv) finally the widths of the bell-shaped protrusions of the spectra are larger for *LCR* functions and shorter for shear compliances.

These phenomena indicate the already expected pheno-

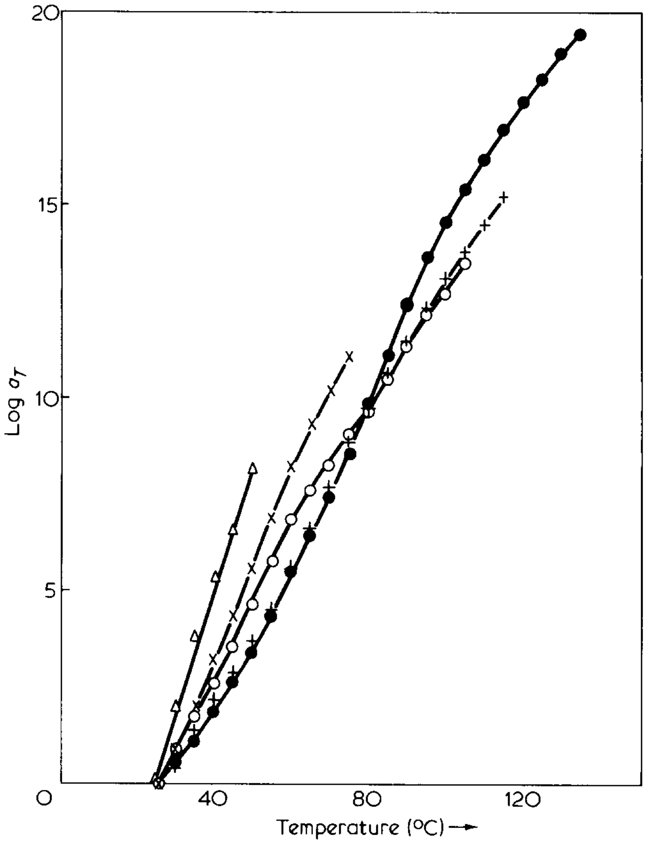

Figure 7 The variation of the log(time) factor a_T *versus* temperature for the five types of unplasticized and plasticized epoxy polymers: ●, C-100-0-8; +, C-100-10-8; ○, C-100-20-8; x, C-100-30-8; △, C-100-40-8

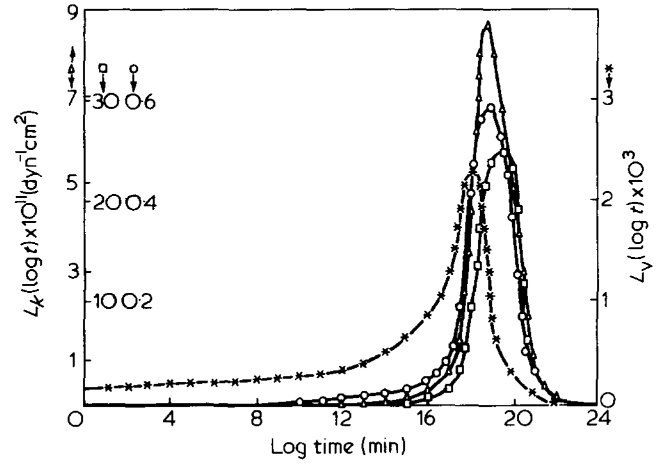

Figure 8 Higher order approximations of retardation spectra for the unplasticized epoxy polymer (C-100-0-8) ($L_s(t)$ = shear, $L_e(t)$ = extension, $L_v(t)$ = bulk and $L_\nu(t)$ = lateral contraction ratio function). Pure epoxy polymer: □, $L_s(t)$ ($k = s$); △, $L_e(t)$ ($k = e$); ○, $L_v(t)$ ($k = v$); * $L_\nu(t)$ ($k = \nu$)

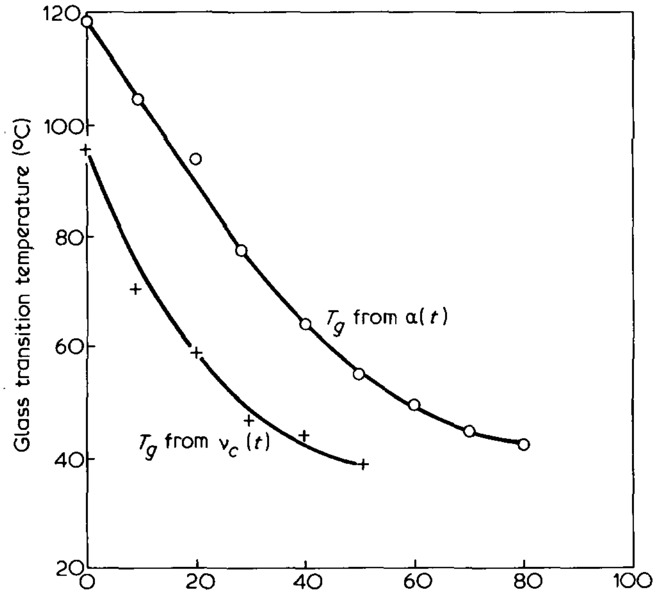

Figure 9 Variations of glass transition temperature T_g *versus* amount of plasticizer as they have derived either from measurements of the thermal expansion coefficient [$\alpha(t)$] of the substances, or from the master of composite curves of lateral contraction ratio in creep [$\nu_c(t)$]

menon that the *LCR* function in some way expresses the coupling between shear and bulk compliances which play a dominant role in the contribution of shear to the corresponding *LCR* function. In this way the *LCR* function should be advanced in time from bulk and shear compliance spectra, as should also occur for the extension compliance spectrum. For an explanation of this phenomenon see ref 19. Moreover, the advance in appearance of the *LCR* function spectrum, which was indicated in the spectrum in *Figure 8*, is in agreement with similar behaviour for the dynamic compliances indicated in *Figure 3* of ref 4.

Another proof of the validity of the sequence of the retardation spectra may be derived by evaluating the glass transition temperatures T_g from the corresponding composite curves for the *LCR* function in creep. Indeed, from *Figures 3* to *5* the following values for T_g may be derived. For the unplasticized epoxy polymer it was found that T_{g_0} = 96°C while for the plasticized polymers it was valid that $T_{g_{10}}$ = 71°C, $T_{g_{20}}$ = 59°C, $T_{g_{30}}$ = 50°C, $T_{g_{40}}$ = 43°C and $T_{g_{50}}$ = 37°C. For the accurate evaluation of T_gs by measuring the variation of thermal expansion coefficients with temperature, use was made of results derived from tests executed on a thermomechanical analyser equipped with a flat-ended quartz rod adjusted to act in a direction normal to the surface of the specimens. A constant axial load, small enough to assure contact of the probe and the specimen, was applied to the probe and the displacement of the probe was recorded as a function of temperature. By a simple procedure described in ref 20 the glass transition temperature in bulk may be determined. *Figure 9* presents the effect of plasticizer on T_g derived from tests evaluating the thermal expansion coefficient. In the same Figure, T_g as derived from composite curves expressing the lateral contraction ratio function was also plotted. Although both curves are similar, the values of T_g derived from *LCR* in creep are always considerably in advance, a phenomenon which is similar to that indicated in *Figure 8*.

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REFERENCES

1 Schwarzl, F. *Kolloid Z.* 1956, **148**, 47–57
2 Freudenthal, A. M. and Henry, L. A. *Solid Propellant Rocket Research* (Ed. M. Sommerfeld), 34–66, 1961
3 Gottenberg, W. G. and Christensen, R. M. *Trans. Soc. Rheol.* 1963, 7, 171
4 Theocaris, P. S. *Kolloid Z. Z. Polym.* 1969, **235**, 1182
5 Rigbi, Z. *Rheol. Acta* 1966, 5, 28
6 Waterman, H. A. *Kolloid Z. Z. Polym.* 1963, **192**, 1
7 Waterman, H. A. *Kolloid Z. Z. Polym.* 1963, **192**, 9
8 Koppelmann, J. *Kolloid Z. Z. Polym.* 1955, **144**, 12
9 Heydemann, P. *Kolloid Z. Z. Polym.* 1963, **193**, 12
10 Theocaris, P. S. *Rheol. Acta* 1964, **3**, 299
11 Theocaris, P. S. *J. Mech. Phys. Solids* 1964, **12**, 125
12 Theocaris, P. S. and Hadjijoseph, C. *Kolloid Z. Z. Polym.* 1965, **202**, 133
13 Theocaris, P. S. *Exp. Mech.* 1965, 5, 105
14 Theocaris, P. S. *J. Polym. Sci. (A)* 1965, **3**, 2619; *Rubber Chem. Technol.* 1966, **39**, 375
15 Theocaris, P. S. *Revue Roumaine des Sciences Techniques (Mech. Appl.)* 1966, **11**, 1185; *Kolloid Z. Z. Polym.* 1966, **209**, 34
16 Kästner, S. *Kolloid Z.* 1958, **157**, 133
17 Theocaris, P. S. 'Moiré Fringes in Strain Analysis', Pergamon Press, London 1969, **4**, 160
18 Theocaris, P. S. and Hadjijoseph, Chr. *Proc. Fourth Int. Cong. Rheology, Part 3* 485–500, Interscience, London–NY, 1965
19 Theocaris, P. S. *Kolloid Z. Z. Polym.* 1970, **236**, 59
20 Theocaris, P. S., Paipetis, S. A. and Papanicolaou, G. *J. Appl. Polym. Sci.* 1978, **22**, 1417