Interrelation of Mechanical and Optical Properties of Plasticized Epoxy Polymers

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Synopsis

A series of cold-setting epoxy polymers, plasticized with different amounts of plasticizer, ranging between 0 and 90% by weight of the amount of the epoxy prepolymer, were studied for their mechanical, optical, and fracture behavior properties. Quantities defining the mechanical properties were considered: the elastic modulus E, Poisson's ratio ν, and fracture tensile stress σ_f. These were accurately measured with electric strain gauges in specimens tested in a 5-ton Instron tester. The optical behavior was characterized by the stress optical coefficients of the materials in both principal directions, α and β, as well as by the coefficient of optical anisotropy, ξ. The values of these quantities were measured by a Fizeau interferometric method. Finally, the optical method of caustics was applied to cracked epoxy polymer specimens to provide a new experimental technique for determining the stress optical properties of these polymers in terms of their mechanical properties. This method was used to check the previous results found by established experimental methods.

INTRODUCTION

The study of the mechanical behavior of epoxy polymers has been reported from both the theoretical and the experimental points of view.[1,5–7] Epoxy polymers are crosslinked thermosetting polymers whose hardening is achieved by adding suitable agents. The thus prepared polymers are insoluble, unmeltable substances of high strength with effective adhesive properties.

The continuously increasing use of epoxy polymers in research and especially in connection with photomechanics made it necessary to study not only the mechanical but also the optical behavior of these substances. Actually, epoxy polymers are used as model materials in two- and threedimensional photoelasticity[2] as well as in birefringent coatings techniques.[3,4] Also their mechanical and optical behavior can be changed between broad limits by adding different amounts of plasticizer. The mechanical and optical properties of epoxy polymers are strongly time dependent, so that these materials can be used to simulate the viscoelastic behavior of other engineering materials.

Theocaris has studied the mechanical and optical viscoelastic behavior of epoxy polymers.[5–9] However, the stress optical behavior of epoxy polymers in these studies was restricted only to cases of linear behavior of polymers useful in classical photoelasticity, where the variations of stress optical coefficients, relating the temporary birefringence to the principal stress difference, was determined by time and temperature.

In the present paper the mechanical and optical behavior of epoxy polymers plasticized with different amounts of plasticizer at ambient temperature was studied. The mechanical behavior was defined by the elastic modulus E and Poisson's ratio ν. The optical behavior was characterized by the stress optical coefficients,[6,7] which relate the variation of the optical paths of either transmitted

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or reflected light rays from the rear face of the specimen. The variation of all these properties with the amount of plasticizer to the epoxy prepolymer was studied in detail.

Finally, the caustics obtained by the reflected and traversing specimen light rays for cracked epoxy-polymer specimens were used for the evaluation of the stress optical constants in terms of the mechanical properties of epoxy polymers. Good agreement between the values of the mechanical and stress optical constants of epoxy polymers and those obtained by other experimental methods was achieved.[8,9]

Stress Optical Coefficients

Let us consider a light beam normally incident on the lateral face of a loaded specimen. The impinging optical rays at any point on the specimen are analyzed along the directions of the principal stresses at the point examined. If we consider the interference of the light rays reflected from the front and rear faces of the specimen, then the difference of the optical paths $\delta_{1,2}$ along either of these directions is given by[6]

$$\delta_{1,2} = 2[(n_{1,2} - n)d + n\Delta d] \tag{1}$$

where n and $n_{1,2}$ are the refractive indices of the material of the specimen at the unloaded and the loaded state, respectively; 1 and 2 are the directions of either of the two principal stresses σ_1 or σ_2; and d and Δd are the thickness and the variation of the thickness of the specimen due to loading.

Introducing into relation (1) the variations of the refractive index $\Delta n_{1,2} = n_{1,2} - n$ due to loading along the principal stress directions, as given by the Maxwell-Neumann strain optical law expressed by

$$\Delta n_{1,2} = b_1\epsilon_{1,2} + b_2(\epsilon_{2,1} + \epsilon_3) \tag{2}$$

where b_1 and b_2 are the strain optical constants and $\epsilon_{1,2,3}$ are the principal strain components, we obtain for the order of fringes $N_{1,2}$ along the directions of the principal stresses σ_1 and σ_2 the following relation[6]:

$$N_{1,2} = 2d(\alpha^*\sigma_{1,2} + \beta^*\sigma_{2,1}) \tag{3}$$

where the stress optical coefficients α^* and β^* are given by

$$\alpha^* = \frac{1}{E\lambda}(-b_1 + 2\nu b_2 + \nu n)$$

$$\beta^* = \frac{1}{E\lambda}[-b_2 + \nu(b_1 + b_2) + \nu n] \tag{4}$$

and ν is Poisson's ratio.

Similarly, we obtain for the variation of the optical paths of the light rays, either traversing the specimen or reflected from its rear face, the following respective equations:

$$\Delta s_{t_{1,2}} = C_t[(\sigma_1 + \sigma_2) \pm \xi_t(\sigma_1 - \sigma_2)]d$$

$$\Delta s_{r_{1,2}} = 2C_r[(\sigma_1 + \sigma_2) \pm \xi_r(\sigma_1 - \sigma_2)]d \tag{5}$$

where

$$C_t = \frac{\alpha_t + \beta_t}{2} \qquad C_r = \frac{\alpha_r + \beta_r}{2} \qquad \xi_{t,r} = \frac{\alpha_{t,r} - \beta_{t,r}}{\alpha_{t,r} + \beta_{t,r}} \tag{6}$$

with

$$\alpha_t = \frac{1}{E}[b_1 - 2\nu b_2 - \nu(n-1)]$$
$$\beta_t = \frac{1}{E}[b_2 - \nu(b_1 + b_2) - \nu(n-1)] \tag{7}$$

and

$$\alpha_r = \frac{1}{E}\left[b_1 - 2\nu b_2 - \nu\left(n - \frac{1}{2}\right)\right]$$
$$\beta_r = \frac{1}{E}\left[b_2 - \nu(b_1 + b_2) - \nu\left(n - \frac{1}{2}\right)\right] \tag{8}$$

The above established stress optical constants can be readily measured by using a simple calibration test, for example, a tension specimen. For this simple case and from eq. (3) we obtain for the stress optical coefficients α^* and β^*

$$\alpha^* = \frac{N_1}{2d\sigma} \qquad \beta^* = \frac{N_2}{2d\sigma} \tag{9}$$

where σ is the tensile stress applied to the specimen.

When the coefficients α^* and β^* are measured, then all the above defined stress optical coefficients can be readily calculated through eqs. (6)–(8).

METHOD OF CAUSTICS FOR THE EVALUATION OF THE MECHANICAL AND STRESS OPTICAL CONSTANTS OF POLYMERS

The optical method of caustics, developed by Theocaris for the analysis of singular elastic stress fields,[9] has been used for the determination of crack-tip stress intensity factors as well as for the formulation of a new experimental technique for the evaluation of the stress optical constants of epoxy polymers in terms of their mechanical properties. The method of caustics consists in allowing a light beam to impinge on the lateral face of the specimen and receiving the reflected or the transmitted light rays on a reference screen placed at some distance from the specimen. These reflected or transmitted light rays, because of the variations in thickness and refractive index of the loaded specimen, are scattered and concentrated along a highly illuminated curve on the screen, which is called the *caustic*.

For the case of a cracked tension specimen, the stress intensity factor, which is the governing quantity of the stress field near the crack tip,[9] is expressed by the following equation in terms of the transverse to the crack axis diameter D_t of the caustic formed by the light rays reflected from the rear face of the specimen:

$$K_I = \frac{1.671}{2z_0 d C_r (\lambda_m^r)^{3/2}} \left(\frac{D_t^r}{3.16}\right)^{5/2} \tag{10}$$

where d is the thickness of the specimen, z_0 is the distance between the specimen and the reference screen, and λ_m is the magnification factor of the optical arrangement, defined by

$$\lambda_m = \frac{z_0 + z_i}{z_i} \tag{11}$$

and z_i is the distance between the focus of the impinging light beam and the specimen.

Relation (10) gives the opening mode stress intensity factor K_I in terms of the transverse diameter D_t^r of the caustic formed by the reflected light rays from the rear face of the specimen. Similarly, the stress intensity factor K_I can be expressed in terms of the transverse diameter D_t^f of the caustic, formed by the light rays reflected from the front face of the specimen by a relation similar to relation (10):

$$K_I = \frac{1.671}{z_0 d C_f (\lambda_m^f)^{3/2}} \left(\frac{D_t^f}{1.87}\right)^{5/2} \tag{12}$$

where

$$C_f = \frac{\nu}{E} \tag{13}$$

The stress intensity factor $\mathrm{K_I}$ is also expressed in terms of the transverse diameter $\mathrm{D_t^t}$ of the caustic formed by the light rays passing through the specimen:

$$K_I = \frac{1.671}{z_0 d C_t (\lambda_m^t)^{3/2}} \left(\frac{D_t^t}{3.16}\right)^{5/2} \tag{14}$$

From eqs. (10) and (14) it can be concluded that

$$\frac{C_r}{C_t} = \frac{1}{2} \left(\frac{D_t^r}{D_t^t}\right)^{5/2} \left(\frac{\lambda_m^t}{\lambda_m^r}\right)^{3/2} \tag{15}$$

As can be observed from relations (5) to (8), the stress optical constants C_t and C_r are connected by the following:

$$C_t = C_r + \frac{\nu}{2E} \tag{16}$$

From eqs. (15) and (16) it can be derived that

$$C_t = \frac{\nu/2E}{1 - 1/2(D_t^r/D_t^t)^{5/2}(\lambda_m^t/\lambda_m^r)^{3/2}} \tag{17}$$

Equation (17) enables the calculation of the stress optical constant C_t related to the light rays passing through the specimen in terms of the ratio ν/E of Poisson's ratio ν to the elastic modulus E. Thus, eq. (17) establishes a relationship between the stress optical behavior of the material, expressed by the constant C_t, and its mechanical behavior, expressed by the ratio ν/E. This relationship between constants C_t and ν/E is established through the diameters of the caustics formed by the light rays reflected and transmitted through the specimen.

After calculating the stress optical constant C_t from the ratio ν/E, eq. (16) enables the calculation of the other stress optical constant C_r through the same ratio ν/E.

Moreover, eqs. (10), (12), and (14) can be used for the determination of the stress intensity factor K_I by measuring the transverse diameters of the caustics formed by the light rays reflected either from the rear or the front faces of the specimen, or those traversing the specimen.

Theoretical values of stress intensity factors have already been determined for a large variety of specimen configurations and loading conditions. For the simple case of an infinite cracked plate loaded in tension normally to the crack axis, the stress intensity factor K_I is expressed by[10–12]

$$K_I = \sigma\sqrt{\pi\alpha} \tag{18}$$

where α is the half-crack length for an internal crack or the whole-crack length for an edge crack. For the case when the plate does not extend up to infinity, K_I is given by[11]

$$K_I = k\sigma\sqrt{\pi\alpha} \tag{19}$$

where the correction factor k expresses the influence of the ratio of the crack length to the width of the specimen on K_I. It has been found that $K_I > 1$ always.

PREPARATION OF SPECIMENS

The specimens were prepared from a pure cold-setting commercial epoxy prepolymer, (Epicote 828), polymerized by addition of 8% triethylenetetramine (TET) hardener per weight of the epoxy prepolymer. The amounts of plasticizer, consisting of a polysulfide polymer Thiocol LP3 added in the prepolymer, were varied between 0 and 90% by weight of pure epoxy prepolymer. The epoxy–plasticizer mixtures were thoroughly mixed in an open cup before the hardener was added. The mixture was then degassed in a vacuum chamber for 20 min to remove all bubbles. Then the mixture was ready for casting. The casting was made in orthogonal molds coated with a suitable oil in order to eliminate adhesion of the mixture to the mold. Plates with highly reflective surfaces can also be obtained in this manner. The ten types of epoxy polymer plates thus prepared were denoted as C-100-p-8, where the letter C stands for the type of the epoxy polymer, which is a cold-setting one, and the two numbers in sequence indicate the percentage by weight of the epoxy prepolymer and the amine hardener added in each preparation. The intermediate letter p indicates the percentage of the plasticizer added to each mixture, which varied from 0 to 90% by amounts of 10% for each batch.

After the removal of the molds, each plate was cured by heating each specimen at 100°C and then slowly cooling it to ambient temperature. The duration of each curing cycle lasted approximately seven days in order to insure complete polymerization of each plate.[5] From the completely cured plates, tension specimens of dimensions 4 × 50 × 200 mm were prepared for the determination of the elastic modulus, Poisson's ratio, and stress optical coefficients of each substance. For the determination of the stress intensity factors at the crack tip as well as for the evaluation of the stress optical constants in terms of the mechanical constants, thin-edge slits of length $a = 5$ mm and width between lips $w = 0.3$ mm were sawn in each specimen, which simulated edge cracks. Also, for the determination of the ultimate stress of the epoxy polymers, plain tension specimens of cross section 4 × 10 mm were tested.

EXPERIMENTAL PROCEDURE

The tension specimens were loaded in a 5-ton Instron tester at ambient temperature (20°C) for the determination of elastic moduli and Poisson's ratios. All specimens for all compositions of plasticized epoxy polymers were prepared and tested under exactly the same environmental conditions in order to yield comparable results. The measurement of the longitudinal and transverse deformations of the tension specimens was made by using electric strain gauges. Figure 1 presents the variation of the elastic modulus and Poisson's ratio at ambient temperature versus the percentage p of plasticizer of the epoxy polymers. It can be observed from this figure that, while Poisson's ratio ν increases with p, tending to its limiting value $\nu = 0.50$ for $p = 100\%$, the elastic modulus decreases as the percentage p of the plasticizer in the epoxy polymer increases. In the same figure, the variation of the fracture tensile stress σ_f of all mixtures of epoxy resins studied is also shown. We can see that σ_f decreases with p. All the above curves are of a sigmoid type, indicating that at low and high values of p the variations of the elastic modulus, Poisson's ratio, and fracture stress are negligible; there is an intermediate region of p values for which these quantities are very dependent on p (transition zone).

A He–Ne gas laser ($\lambda = 6328$ Å) was used to illuminate the specimen for the measurement of the stress optical constants α^* and β^*. The optical interference pattern formed by the reflected light rays from the front and rear faces of the specimen was received on a reference screen. The variation of the fringe order $N_{1,2}$ for a light beam polarized along the longitudinal and transverse axes of the specimen was detected by a photocell connected with an x–y plotter. The stress optical constants α^* and β^* were then calculated through relations (9). Figure 2 presents the variation of α^* and β^* versus the percentage p of plasticizer in the epoxy polymers. It can be seen that α^* and β^* increase with p. The variation of the stress optical coefficient C, related to the light rays traversing the specimen, and the coefficient ξ, expressing the optical anisotropy (birefringence) of the specimen due to loading, is also shown (Fig. 2).

After the determination of the stress optical coefficients, eq. (10) was used to calculate the crack-tip stress intensity factors in slotted plasticized epoxy polymer

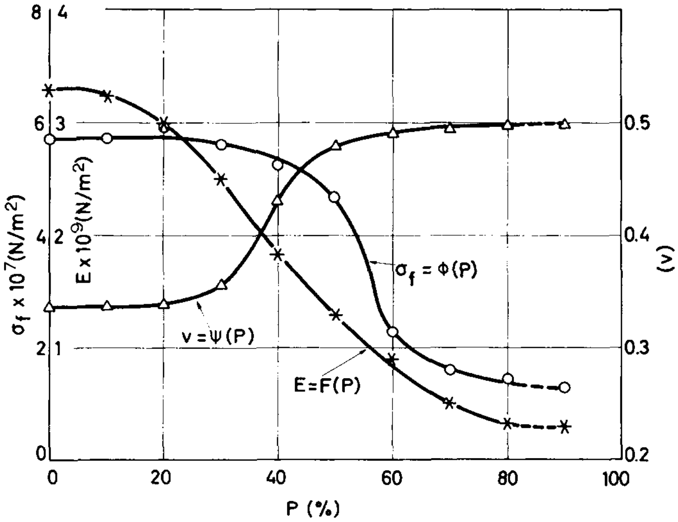


Fig. 1. Variation of elastic modulus E, Poisson's ratio ν, and fracture stress σ_f vs amount p of plasticizer of the epoxy prepolymer, ranging between 0 and 90% of the amount of the epoxy prepolymer.

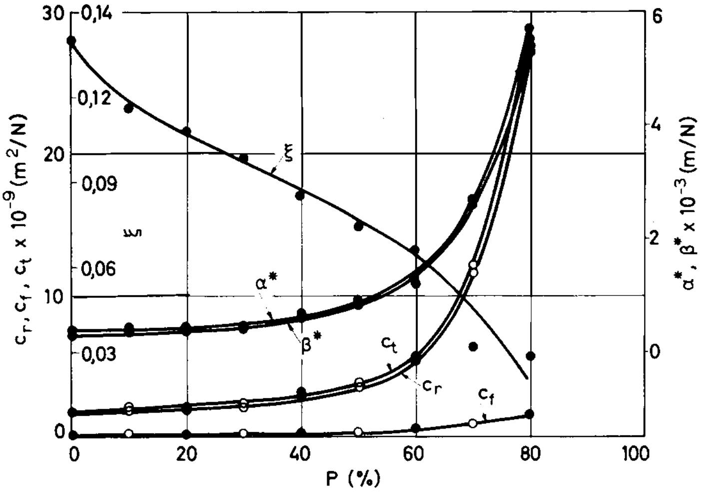


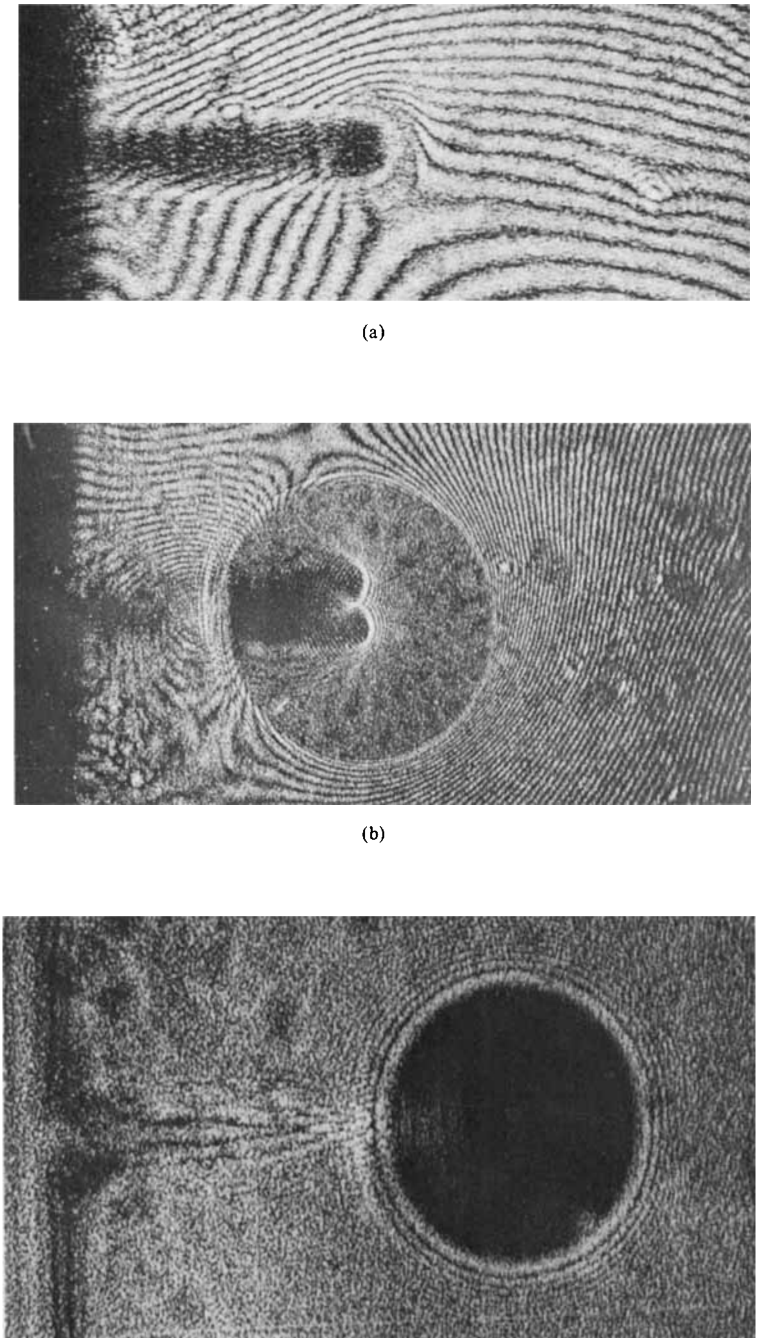
Fig. 2. Variation of stress optical constants α^*, β^*, C_r, C_f, C_t, and ξ vs amount p of the plasticizer of the epoxy prepolymer, ranging between 0 and 80% of the amount of the epoxy prepolymer; (●) and (○) indicate results obtained by the Fizeau interferometric method and the new technique based on the method of caustics, respectively.

specimens. These slotted specimens were interposed in a He–Ne gas laser light beam impinging on the specimen with a magnification factor of the optical system λ_m varying between 5.5 and 10.11. Figure 3 shows the optical patterns obtained by the light rays reflected from the front and rear faces, Figure 3(a) and 3(b) and those traversing the specimen, Figure 3(c), for a C-100-10-8 slotted specimen, illuminated by a light beam and subjected to a stress equal to $\sigma = 5 \times 10^6$ N/m^2.

The optical pattern formed by the light rays reflected from the specimen consists of a dense interferogram induced by the interference of the light rays reflected from the two faces of the specimen and two highly illuminated curves, the caustics, each of which formed by reflections from either face of the specimen (Fig. 3). The external caustic is formed by reflections from the rear face of the specimen, and the internal caustic, by reflections from the front face. In the corresponding optical pattern formed by the transmitted light rays, only one caustic is formed.[6] By measuring the three transverse diameters $D_t^{r,f,t}$ of all these caustics, the stress intensity factor K_I was calculated by applying eqs. (10), (12), and (14).

Figure 4 presents the variation of the stress intensity factor K_I for all compositions of the plasticized epoxy polymers. The theoretical values of K_I were also drawn (continuous straight line of Fig. 4). It can be observed that the experimental values of K_I are in close agreement with the theoretical curve.

By also measuring the transverse diameters of the caustics formed by the reflected light rays from the rear face of the specimen and those traversing the specimen, the stress optical constants C_t and C_r were determined in terms of the ratio ν/E of Poisson's ratio to the elastic modulus of the corresponding epoxy polymer. Thus, the stress optical constants C_t and C_r (Fig. 2) were determined by knowing only the corresponding values of ν and E. It can be observed that these values of C_t and C_r compare favorably with the values of these constants measured by the very sensitive Fizeau interferometric method.



(a)

(b)

(c)

Fig. 3. Optical patterns obtained by illuminating a slotted C-100-10-8 epoxy polymer by a light beam. Case (a) corresponds to the unloaded specimen. Case (b) to reflected light rays from the specimen in which a tensile stress equal to $\sigma = 5 \times 10^6$ N/m^2 has been applied and case (c), to light rays passing through the specimen loaded as in case (b).

The experimental values for the mechanical and optical constants of the epoxy polymers studied are listed in Table I.

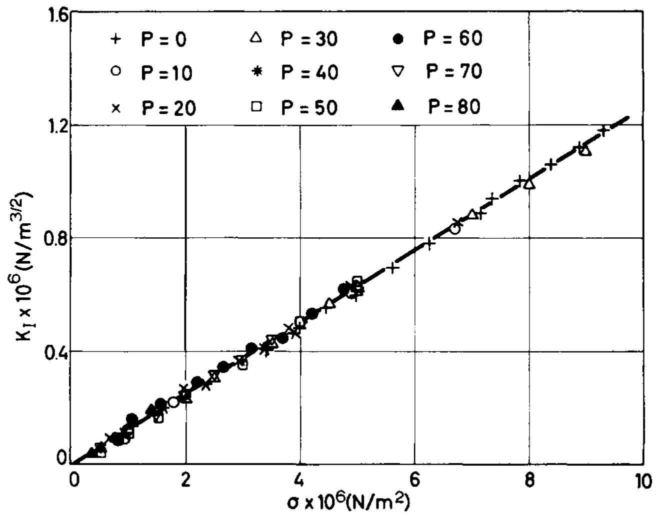


Fig. 4. Variation of stress intensity factors K_I vs applied stress σ for plasticized epoxy polymers where p indicates the percentage of the plasticizer contained in each mixture.

TABLE I

Mechanical and Optical Constants of Epoxy Polymers

p, %	$\alpha^* \times 10^{-3}$, mN^{-1}	$\beta^* \times 10^{-3}$, mN^{-1}	$C_r \times 10^{-10}$, m^2 N^{-1}	ξ	$E \times 10^9$, Nm^{-2}	ν	$\sigma_f \times 10^7$, Nm^{-2}
0	0.382	0.308	1.671	0.1401	3.30	0.338	5.73
10	0.429	0.357	1.965	0.1161	3.22	0.338	5.73
20	0.439	0.386	2.040	0.1080	3.00	0.340	5.98
30	0.482	0.416	2.125	0.0983	2.50	0.358	5.59
40	0.705	0.623	3.020	0.0859	1.82	0.430	5.24
50	0.902	0.818	3.596	0.0739	1.30	0.480	4.68
60	1.319	1.208	5.273	0.0666	0.90	0.490	2.27
70	2.670	2.556	11.585	0.0311	0.50	0.495	1.60
80	5.679	5.429	27.333	0.0289	0.32	0.500	1.46
90	—	—	—	—	0.30	0.500	1.29

[a] p is the percentage of the plasticizer; α^* and β^* are the stress optical coefficients; ξ is the coefficient of the optical anisotropy; E is the modulus of elasticity, ν is Poisson's ratio; and σ_f is the fracture tensile stress.

CONCLUSIONS

The mechanical and stress optical behavior of plasticized epoxy polymers was studied. As representative quantities of the mechanical behavior, the elastic modulus and Poisson's ratio were considered, whereas the stress optical behavior was determined by the stress optical coefficients connecting the variation of the optical path of a plane-polarized light ray along the principal stress directions to the individual values of the principal stress. The variation of the above quantities with the amount of plasticizer of the epoxy polymers, ranging between 0 and 90% of the amount of the epoxy prepolymer, was studied. It was shown that the curves representing the variation of elastic modulus, Poisson's ratio, and fracture tensile stress versus the amount of plasticizer p are all of a sigmoid type. They present an intermediate transition region which corresponds to values of p between 30 and 60%, for which the variation of all these quantities is strong. For values of p lying outside the above region, all the quantities expressing the mechanical behavior of the epoxy polymers vary insignificantly with p. Contrary to the mechanical properties, the stress optical properties of plasticized epoxy polymers, expressed by the stress optical coefficients, vary monotonically with p. The variation of these last quantities for values of p larger than 50% is extremely steep.

Apart from the determination of the mechanical and stress optical constants of the plasticized epoxy polymers with amounts of plasticizer varying between 0 and 80% per weight of the epoxy polymer, a new experimental technique, based on the method of caustics, for the calculation of the stress optical constants in terms of the mechanical constants was introduced. According to this technique, the caustics obtained by the light rays reflected from the rear face of a cracked tension specimen and those traversing the specimen were compared. The constants C_t and C_r were determined in terms of the ratios ν/E of the Poisson's ratio to the elastic modulus for each substance. The method of determining the stress optical constants in terms of the mechanical constants avoids the use of tedious and time-consuming interferometric methods.

Finally, the study was directed to the determination of the fracture behavior of the plasticized epoxy polymers. The crack-tip stress intensity factors, representing the controlling parameter for the rear to the crack-tip stress field, were determined for plasticized epoxy polymers. Good agreement with the theoretical predictions was obtained.

The determination of the mechanical and optical behavior of epoxy polymers is of interest in experimental mechanics where these substances are used as model materials. The mechanical and optical properties of epoxy polymers vary according to their particular composition and therefore they can be used to simulate the elastic or viscoelastic behavior of other materials (metals or plastics). Thus, the optical method of caustics can be extended to solve fracture mechanics problems for single or composite materials.

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