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IRRADIATION OF POLYMERS FOR PHOTOELASTIC INVESTIGATIONS

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Abstract—The photoelastic behaviour of transparent polymers is studied with the objective to learn more about the correlations between molecular structure, mechanical properties, and stress–optical sensitivity on the one hand and to improve the fixation process by radiation at room temperature for practical purposes on the other hand. The fixation of three dimensional states of deformation allows the analysis of special states of elastic stresses even in the interior of complex constructional units and machine parts. As a continuation of former work, the polymeric system consisting of a well-known epoxy resin and maleic anhydride has been modified by the addition of mono- and polyfunctional acrylates and of an inhibitor to protect the reactive double bonds. Preliminary results are presented. This work is part of the Austrian contribution to an international co-operation between scientists from Hungary, Croatia and Austria.

INTRODUCTION

Transparent plastic material showing photoelastic properties has for a long time served as a means of studying states of deformation as well as stresses of a loaded body. In order to analyze such deformations or stresses even in the case of three-dimensional problems, it is necessary to obtain some sort of fixation of these states of deformation or stress. The classical method of fixation is the so-called "freezing procedure". This method is however limited in its application by the following reasons:

(1) Large deformations are needed in order to get readable results; that means that the ratio between the strains in the plastic model and the original part must be greater than in the classical two-dimensional procedure of fixation.
(2) At the freezing temperature it is not possibl e to obey Poisson's law of similarity; that means that in all cases where Poisson's ratio plays a certain role the results obtained are more or less incorrect.
(3) If, as in many cases, the original part or construction—and therefore the model too—consists of parts with different mechanical and thermal behaviour and/or anisotropic materials, the freezing method is practically not applicable.

In order to get any information of a complex three-dimensional state of stress, these limitations are usually ignored. On the other hand many attempts have been made to overcome these problems by fixation procedures at room temperature. The most promising attempts in this direction probably have been made by the use of ionizing radiation. However this procedure needs to be studied also from the viewpoint of radiation and polymer chemistry more basically than has been done in the past. Early work on this subject goes back to the fifties and sixties (see Miki *et al.*, 1969, and papers cited there). The fixation by ionizing radiation has mainly been studied by Miyazono (1967, 1969) in Japan and by Savchenko and Shokotko (1971) and Shokotko *et al.* (1972) in the Soviet Union, followed by studies in other countries (e.g. Gross-Petersen, 1972; Ajovalasit and Oliveri, 1973). Mainly cast resins like epoxy and polyester resins have been investigated due to the possibility that from these materials models of different shape can easily be formed.

Especially the more recent studies of Jecic and co-workers (Jecic and Kuch, 1978; Jecic and Goja, 1984, 1987) stimulated the present search for polymers with better fixation of stress deformation. Before the fixation process can be influenced in a systematic way, it seems to be necessary to study more basic properties of selected polymers, especially the photoelastic sensitivity and creep behaviour.

EXPERIMENTAL

Polymeric materials

For the first series of investigations two types of resins were chosen. The one type is an epoxy resin modified by the addition of crosslinking agents like maleic anhydride (MAA) and/or mono- or polyfunctional monomers and oligomers. To the other type belong radiation-curable monomers and resins

with a tendency for crosslinking. Essential in both cases seems to be a curing process with minor crosslinking in the first step followed by a pronounced crosslinking process as a second step.

The resin mixtures of the first series—a continuation of "classical" work—are compiled in Table 1.

By adding the inhibitor *p*-methoxy phenol, four different mixtures are derived from the mixtures O, A, B and C, containing 0, 0.1, 0.5 and 1.0% of inhibitor. In a second series the same basic mixtures were used with the addition of 1.0 and 3.0% of inhibitor, since it turned out that concentrations lower than 1% were almost ineffective.

The third series of resins (monomers, oligomers) contained radiation-curable compounds which will be reviewed elsewhere.

Production of test specimens and irradiation

Models for photoelastic investigations have to be stressless before being exposed to load. A major problem was a proper demoulding of the test specimens. Therefore diverse hollow moulds were designed and tested, using different materials and parting compounds. A certain temperature programme was developed to avoid the formation of thermal stresses. For cast resins bigger plates or sheets were formed from which six to ten circular test specimens with a diameter of 44 mm and a thickness of 10 mm could be made. The circular test specimens were notched diametrically with semi-circular notches ($\emptyset = 5$ or 1 mm).

The irradiation of the test specimens was performed in a Gammacell 220 with dose rates between 2 and 6 kGy/h. So far mainly irradiation of the models in the unloaded state has been carried out.

Construction of load devices and arrangement of an automatic measurement and evaluation

At the beginning a classical photographic set-up was used. It soon turned out that for a fast and reliable measurement of the test specimens an automatic device needed to be developed. The first step was the construction of a pneumatic load unit instead of a simple weight-loaded unit. In the meantime two more units of this type were built, the compact form of which allows their use in the small irradiation chamber of the Gammacell 220.

Instead of a photographic camera a CCD camera and a PC equipped with a frame-grabber (for image processing) are used for the recording of the fringe patterns as a function of load and time. A special computer programme allows the determination of the stress–optical parameters as a function of time in the relaxation test. Thereby the test can be interrupted automatically when the recorded data follow the theoretical function for a certain time.

The principle of measurement is a dark-field procedure with an angle of 45° between the loading and the polarizer (and of course another 90° between polarizer and analyzer); the linear polariscope shows isochromates as well as isoclines. The complete set-up consists of monochromatic light source (Na), pneumatic load device, polarizer, analyser, CCD camera, control and dialogue monitors and PC, equipped with frame-grabber.

The evaluation of the isochromates in the x- and the y-axis by the formulas shown below yields the values of the stress-optical constant S (or f_σ), F being the force, R being the radius of the sample, x and y the coordinates and n the order of the isochromates.

$$S_x = \frac{1}{n} \times \frac{4F}{\pi R} \times \frac{1 - \left(\frac{x}{R}\right)^2}{\left[1 + \left(\frac{x}{R}\right)^2\right]^2}$$

$$S_y = \frac{1}{n} \times \frac{4F}{\pi R} \times \frac{1}{1 - \left(\frac{y}{R}\right)^2}$$

RESULTS AND DISCUSSION

In order to attain the declared aim, i.e. a better understanding of the involved processes and therefrom an improvement of the fixation of deformations at room temperature, it seemed to be necessary to study the photoelastic behaviour of selected polymers quite basically. Former research has revealed only to a small degree the correlation between polymer structure and stress–optical activity. To our knowledge no extended investigations on this matter have been published so far. The probably best studied polymer system is that of epoxy resins (Araldit B® type) modified by maleic anhydride in order to make it crosslinkable by radiation. This is the reason why

Table 1. Modification of polymers made from "classical" epoxy resin/maleic anhydride mixtures

Mixture	Components (parts by weight)	Inhibitor (%)/marking 0	0.1	0.5	1	3
O	100 Araldit B + 21 MAA	O1	O2	O3	O4	O5
A	95 O + 5 arom. PEA	A1	A2	A3	A4	A5
B	95 O + 5 GPTA	B1	B2	B3	B4	B5
C	95 O + 5 arom. EPA	C1	C2	C3	C4	C5

Araldit B® = epoxy resin (Ciba); MAA = maleic acid anhydride = maleic anhydride; PEA = polyester acrylate, functionality = 6; GPTA = propox. glyceryl triacrylate; EPA = epoxy acrylate, functionality = 1; Inhibitor = *p*-methoxy phenol = hydroquinone monomethyl ether.

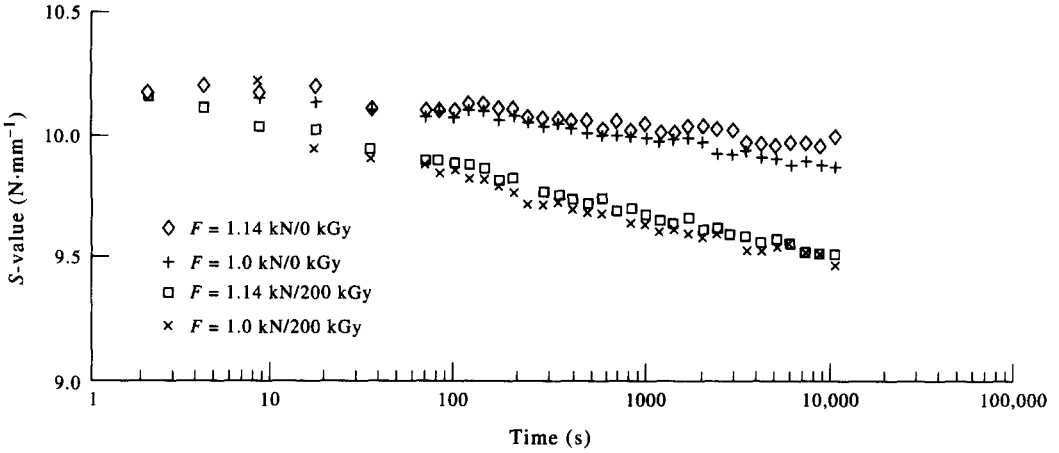

Fig. 1. Optical creep of thermally cured Araldit B®/MAA (system OI in Table 1) in non-irradiated and irradiated (γ, 200 kGy) state loaded with 1.0 and 1.14 kN, respectively.

we started our investigations with the further modification of the epoxy resin Araldit B®.

The suitability of polymeric material for three-dimensional models depends on different parameters of which the stress–optical constant f_σ or S (in the graphs), the modulus of elasticity (flexural modulus) E, the ratio E/f_σ, the transverse strain number and the creep behaviour of the material are the most important. The ratio E/f_σ should not be lower than about 3000 cm^{-1}, and creep, optical as well as mechanical, should be low or at least reproducible for proper correction.

The mixture of 100 parts by weight Araldit B and 21 parts by weight MAA shows, after thermal curing, some creep which can be seen in Fig. 1 by the decrease of f_σ (S-value) with time under constant load. When the cured material is irradiated with 200 kGy, the decrease and therefore the optical creep is stronger. In both cases it follows a logarithmic function (between about 5 and 12,000 s). Furthermore a small dependence on the applied load (force) can be observed. This non-ideal behaviour of the material makes it nevertheless a good object to study.

A thermally initiated curing may destroy a part of the crosslinking ability, which is to be introduced into the polymer by the addition of MAA (and acrylates). Therefore different amounts of the inhibitor p-methoxy phenol (=hydroquinone monomethyl ether) were added in order to protect the double bond in the MAA-unit as well as in the added mono- or polyfunctional acrylates. In Fig. 2 the S-value of polymer mixtures with 1 and 3% inhibitor in the unirradiated state and irradiated with doses of 100 and 200 kGy are shown for comparison. As can be seen, the higher concentration of inhibitor enhances the S-value in all four cases independent of irradiation. The increase of S is about 3–5%. The influence of the applied radiation doses is even less. In the "original" mixture O (without acrylates) no systematic change with dose can be observed. The small but distinct decrease of S with increasing dose seen in the polymers A and B, which contain

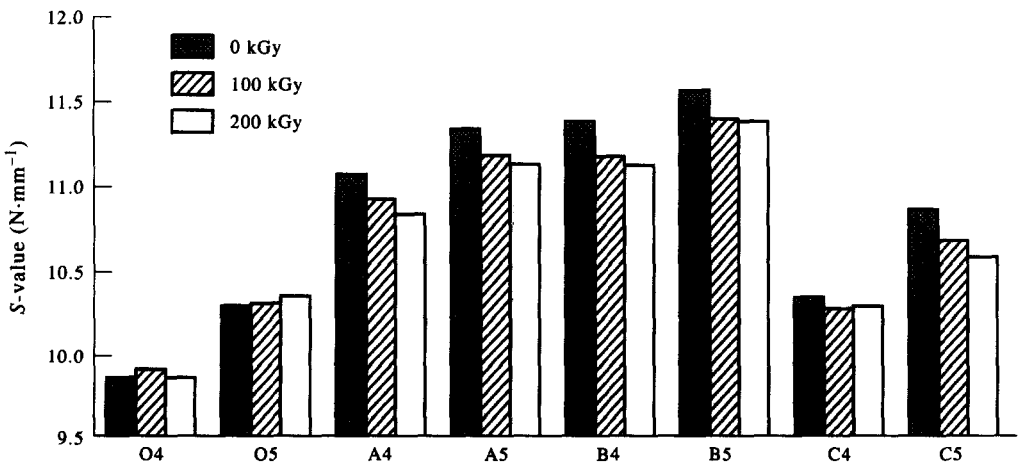

Fig. 2. Dependence of the stress–optical constant S from the composition of the polymer, especially from the content of inhibitor, and from the radiation dose. For the composition of O, A, B and C see Table 1.

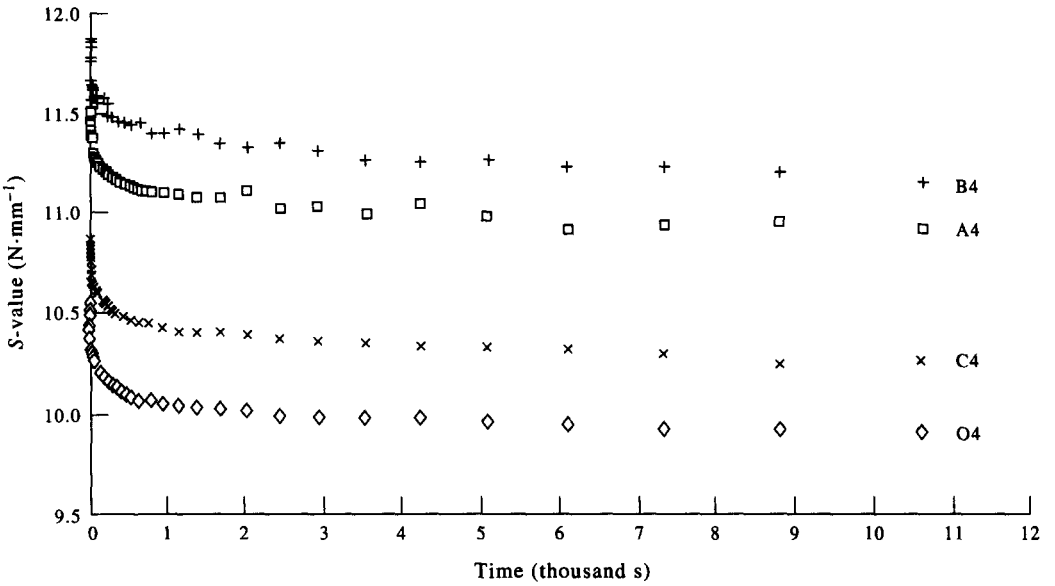

Fig. 3. Influence of the polymer composition on the stress–optical sensitivity in the case of 1% inhibitor content at a dose of 100 kGy. For the composition of O, A, B and C see Table 1.

polyfunctional acrylates, seem to be characteristic, although the interpretation of the data is difficult and needs further investigation. The general increase of S in A and B compared to O and C can be attributed to the higher functionality of the acrylates (see Table 1). The decrease of S with time is shown in Fig. 3 and Fig. 4 for polymers containing 1% inhibitor and irradiated to 100 kGy. Within a period of time less than 1000 s (about 15 min) the rheological process seems to slow down substantially (Fig. 3). following again a logarithmic function as can be seen in Fig. 4. This figure clearly presents the influence of the three acrylic modifiers. Probably according to their crosslinking efficiency, described by their double bonds per molecule and by their double bond content in moles double bonds per kg (GPTA: 3/6.8, PEA: 6/5.6, monom. EPA 1/2.5), distinct S-values are found. While GPTA yields the highest value for S, the addition of the PEA results in a lower creep of the S value. The difference between the initial and the "final" values in the curves can be taken as a measure for the optical creep, which not necessarily correlates with the mechanical creep process. Mechanical measurements are in progress and can hopefully contribute to reveal correlations between optical and mechanical creep processes. In addition the molecular structure and the alignment of dipoles under load within it, which causes the optical birefringence, need

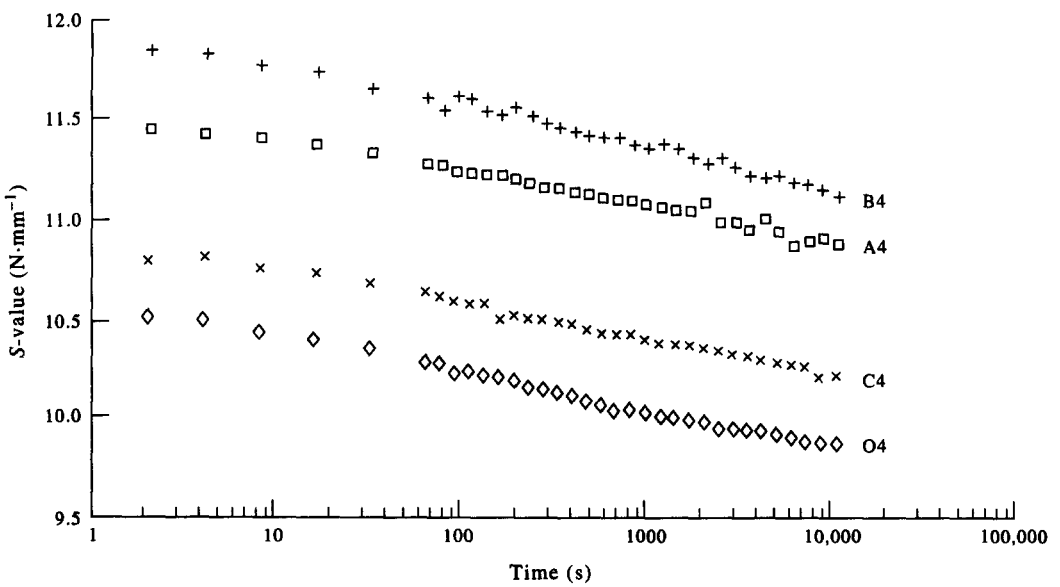

Fig. 4. Semi-logarithmic plot of the influence of polymer composition on the stress–optical sensitivity at a dose of 100 kGy. For the composition of O, A, B and C see Table 1.

to be studied in order to better understand the complex correlations. The work is in rapid progress.

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