Elastic Properties of Adhesive Polymers. I. Polymer Films By Means of Electronic Speckle Pattern Interferometry

Johannes Konnerth,[1] Wolfgang Gindl,[1] Ulrich Müller[1,2]

[1]*Institute of Wood Science and Technology, Department of Material Sciences and Process Engineering, BOKU–University of Natural Resources and Applied Life Sciences, Vienna, Austria*
[2]*Wood Kplus – Competence Center for Wood Composites and Wood Chemistry, Linz, Austria*

Received 16 November 2005; accepted 2 March 2006
DOI 10.1002/app.24434
Published online in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: The elastic modulus and Poisson's ratio of seven different polymers frequently used as wood adhesives and/or matrix polymers in wood- and natural-fibre-reinforced composites, respectively, were determined by means of tensile tests. Specimen deformation during testing was measured by means of a mechanical extensometer and an electronic speckle pattern interferometry system, respectively. The results from both methods show an excellent correlation for the elastic modulus. The elastic moduli of the studied polymers cover a wide range from 0.47 GPa for polyurethane to 6.3 GPa for melamine–urea–formaldehyde, whereas Poisson's ratios show less variability. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 103: 3936–3939, 2007

Key words: adhesives; electronic speckle-pattern interferometry; mechanical properties

INTRODUCTION

In order to understand the transfer of mechanical loads across adhesive bonds in solid wood bond-lines, wood composites, and natural-fibre-reinforced composites, a broad variety of factors concerning topics as diverse as surface chemistry, polymer chemistry and physics, and wood and fibre science are to be considered. Recently, noncontacting methods for the measurement of strain distribution on material surfaces have been applied to study the micromechanics of adhesive bond-lines in solid wood.[1,2] Also, the strain distribution arising in a single-fibre reinforced model composite has been measured in a noncontacting way in order to study the transfer of load from the matrix to a reinforcing wood-particle in wood–plastic composites.[3] By measuring strain distribution, these studies aimed at a better understanding of the occurrence of stress concentrations in bond-lines and composites. However, any method of stress analysis requires some knowledge of the stress–strain behavior of the constituent materials. In case of an adhesive joint these properties are usually well known for the adherent, whereas adhesive properties are often less easy to determine[4–7] and data is scarce. For example, Wooley et al.[8] examined the stress concentration in lap joints as a function of the ratio of the elastic modulus of the adherent and the elastic modulus of the adhesive, considering also the thickness ratio of the adhesive layer and the adherent. In order to apply a similar analysis to wood bond-lines, for example, input parameters for elastic adhesive properties are required.

In the present work we use a mechanical extensometer and electronic speckle-pattern interferometry (ESPI) equipment for quantifying the elastic modulus and Poisson's ratio of seven different types of polymers frequently used as wood adhesives and/or matrix polymer in natural-fibre-reinforced composites.

MATERIALS AND METHODS

Specimen preparation

Films were prepared from four different representative wood adhesives: polyvinylacetate (PVAc, PV/H Holzleim Standard, Henkel Austria GmbH, Austria, Vienna), melamine–urea–formaldehyde (MUF, Dynomel L-435 with hardener H469, Dynea Austria GmbH, Krems, Austria), phenol–rescorcinol–formaldehyde (PRF, Aerodux 185 with hardener HR150, Friebe, Mannheim, Germany), and one-component polyurethane (1K PUR, Purbond HB110, Collano AG, Sempach, Switzerland). In addition, the following matrix polymers used for the production of fibre reinforced composites were tested: epoxy (Epoxidharz L Nr. 236349 with hardener L Nr. 236357, Conrad Electronic, Hirschau, Germany), polyester (Polyester-Laminierharz VIPAL VUP 4782 BEMT with hardener MEKP M300, Gerber GFK-Systeme, Stuttgart, Germany), and polypropylene (PP, PP301460/14 film, 0.5 mm, Goodfellow Cambridge Ltd, Huntingdon, U.K.).

Correspondence to: J. Konnerth (johannes.konnerth@boku.ac.at).

Contract grant sponsor: Austrian Science Fund FWF; contract grant number: P16837-N07.

Journal of Applied Polymer Science, Vol. 103, 3936–3939 (2007)

MUF, PRF, epoxy, and polyester were prepared between two polytetrafluoroethylene (PTFE) panels, ensuring a constant film thickness by means of 0.5-mm-thick glass plates placed between the PTFE panels. The films were cured overnight at ambient temperature and conditioned for one week between screw-clamped wood panels so as to prevent them from warping. In the case of MUF and PRF, which contain a considerable amount of water, absorptive paper sheets were placed between polymer films and wood panels and were changed repeatedly to ensure slow drying without distortion. PVAc and PUR films were produced by pouring the liquid adhesive onto a PTFE panel, where a film with a homogenous adhesive thickness of about 0.2 mm for PVAc and 1.4 mm for PUR was prepared by means of a spatula. PUR films were repeatedly turned over to ensure curing on both sides.

Tensile specimens were prepared by cutting strips with a width of 6 to 8 mm and a length of 60 mm from the cast films. To prevent edge stresses and therefore inhomogeneous stress distribution within the sample during testing, the edges of the strips were sanded with P500 sandpaper. Beech wood strips were glued to the ends of the polymer strips by means of a hot melt adhesive in order to fix the tensile specimens to the clamps of the universal testing machine, leaving a free sample length of 40 mm.

Tensile testing

Tensile tests were performed without fracturing specimens on a universal testing machine Zwick/Roell Z100 equipped with a mechanical extensometer (Macrosense, Zwick/Roell, Ulm, Germany) and a 2.5 kN load cell (Fig. 1). A zero span of 30 mm was chosen for the extensometer. At a constant cross-head speed of 0.5 mm/min, the specimens were loaded to a peak load of 8 MPa. Due to the lower maximum strength of PVAc and PUR, the peak load of these specimens was reduced to 4 MPa. The peak load for PP was 5 MPa. The Young's modulus was calculated by fitting a linear regression to the linear part of the recorded stress strain curve.

Using the same tensile specimens, tensile tests were also performed using an electronic speckle pattern interferometer (ESPI) system Q300 (Dantec-Ettemeyer, Ulm, Germany) (Fig. 1). The basic principle of the ESPI technique is explained in detail by Müller et al.[2] MUF, PRF, epoxy, and polyester specimens were loaded to a prestress of 4 MPa and then strained in 15 increments with constant step size, resulting in a final tensile stress of 8 MPa. Due to the small prestress and the small load steps, the specimen response was linear. Because of the high deformation for PUR, PVAc, and PP specimens noted from previous tensile tests, a preforce of only 1 MPa was applied. Twenty load increments of 0.2 MPa were used for PP, and load was further reduced for PUR and PVAc specimens, where 15 load increments of 0.1 MPa were applied. The displacement of each specimen was observed in two consecutive tests on both sides of the polymer film, and the results were averaged.

Speckle images of the specimen surface (the size of the observed field of view was 6 $\times$ 30 mm) were

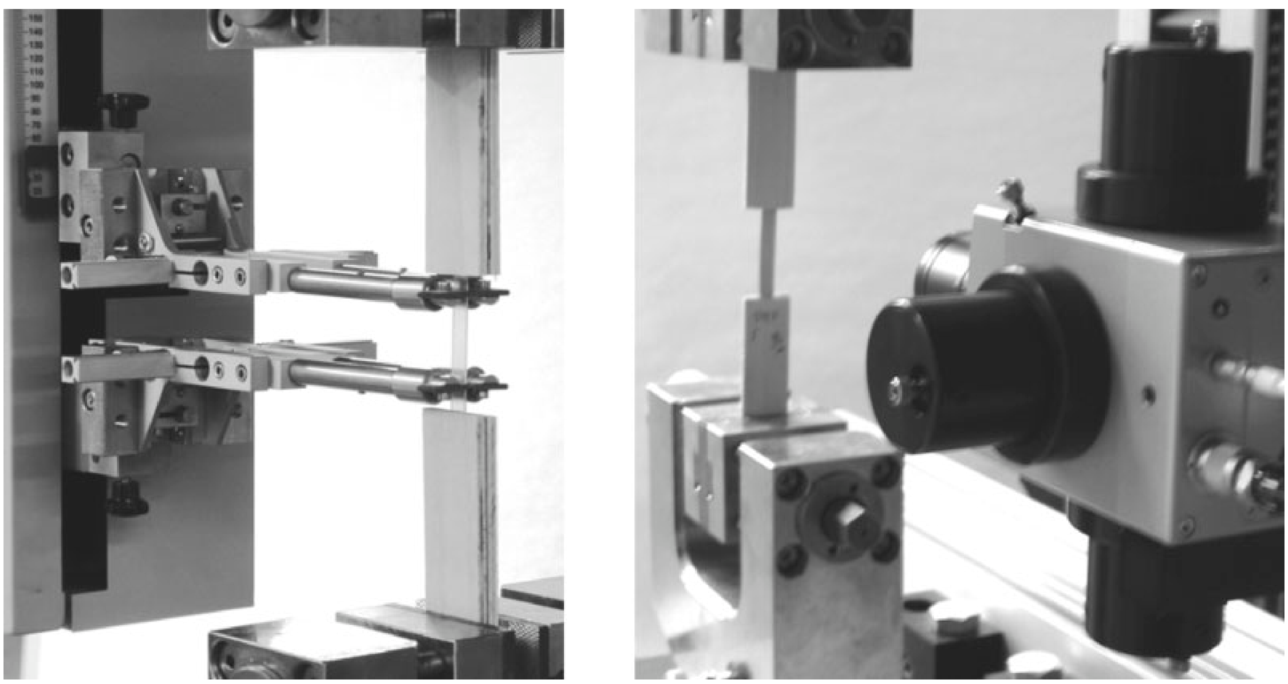

Figure 1 Tensile test setup with mechanical extensometer (left) and ESPI (right).

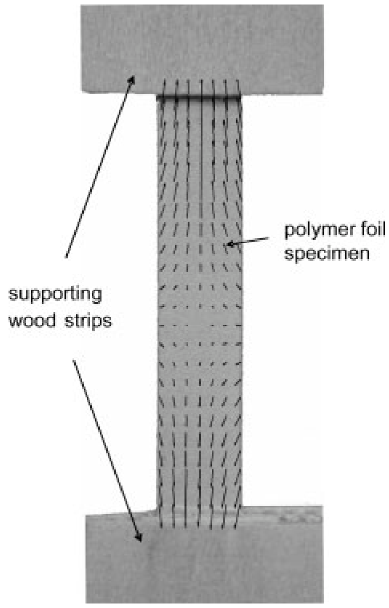

Figure 2 Image of a polymer tensile specimen with displacement vectors measured by means of ESPI equipment.

captured after each displacement step and 2D displacement maps were computed by summing up information from all (15 and 20, respectively) displacement steps. Vertical (i.e., tensile) and horizontal (i.e., transverse specimen contraction during uniaxial tensile testing) strain was calculated by differentiating displacement maps. Poisson's ratio ν was calculated by dividing the average of vertical strain across the field of view by the average horizontal strain. The elastic modulus *E* of each specimen was calculated by dividing tensile stress by the respective vertical strain:

$$G = \frac{E}{2(1+\nu)}. \quad (1)$$

From *E* and ν the shear modulus *G* was calculated using eq. 1, assuming isotropic and homogeneous material properties of the adhesive polymer films.

RESULTS AND DISCUSSION

Figure 2 shows a vector image of displacement obtained from ESPI measurements superimposed onto a speckle image of a tensile specimen. The displacement vectors clearly demonstrate strains in the load direction and the contraction of the sample perpendicular to the applied load. The vectors show a homogeneous deformation of the films throughout the whole observed field of view. Thus it is justified to calculate Poisson's ratio and elastic modulus from strain values averaged across the whole field of view. By comparing elastic moduli measured by means of ESPI with values obtained using the mechanical extensometer macrosense (Fig. 3), it is demonstrated that ESPI measurements show an excellent correlation with mechanically obtained data and are therefore equally valid.

Elastic moduli determined by using the mechanical extensometer macrosense and ESPI, respectively, Poisson's ratios measured with ESPI, and shear moduli calculated using equation 1 are summarized in Table I. While the elastic moduli of the studied polymers cover a wide range from 0.47 GPa for PUR to 6.3 GPa for MUF (macrosense measurement), Poisson's ratios show less variability.

Reference data of elastic moduli and Poisson ratios of resins and polymers are scarce in the literature and also seldom available from suppliers. Only a few studies investigating the mechanical properties of resins used for wood composites and the timber industry are available. Elastic moduli of resins were measured for MUF,[9] PUR,[10] and PRF.[11] The latter was determined by means of nanoindentation. This method entails an overestimation of the elastic modulus, which explains the higher values in reference [11] as compared to the ones measured in the present study. The elastic modulus of PUR measured with the mechanical extensometer (0.47 GPa) and with ESPI equipment (0.36 GPa) agrees well with the value reported by Broughton and Hutchinson[10] (0.5 GPa). An elastic modulus of 9.0 GPa was reported for MUF resin Landolt,[9] which is significantly higher than results from our own measurements with 6.3 GPa (mechanical extensometer) and 7.0 GPa (ESPI), respectively. Besides different testing conditions and methods, also the formulation of an adhesive has a significant influence on its elastic properties,[12] which could be the reason for the differences in stiffness observed in the respective studies.

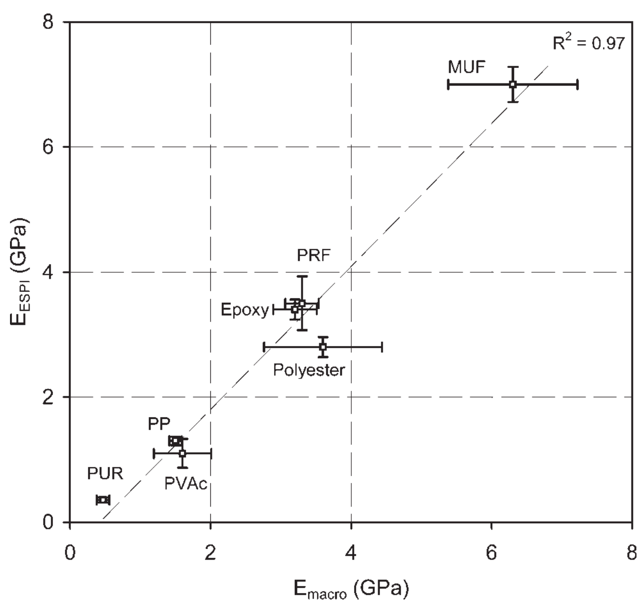

Figure 3 Correlation of elastic modulus measured using mechanical extensometer (E_{macro}) and elastic modulus measured using ESPI (E_{ESPI}).

TABLE I
Elastic Properties of Polymer Films Measured with Macrosense Displacement Sensors and ESPI (Arithmetic Mean and Standard Deviation; Number of Tested Samples in Brackets)

Type of polymer	E_{macro} (GPa)	E_{ESPI} (GPa)	Poisson's ratio	G (GPa)
Epoxy	3.2 ± 0.31 (9)	3.4 ± 0.16 (9)	0.36 ± 0.057 (8)	1.2
Melamine–urea–formaldehyde	6.3 ± 0.92 (8)	7.0 ± 0.28 (5)	0.34 ± 0.073 (5)	2.4
Phenol–resorcinol–formaldehyde	3.3 ± 0.24 (8)	3.5 ± 0.43 (7)	0.36 ± 0.083 (5)	1.2
Polyester	3.6 ± 0.84 (8)	2.8 ± 0.16 (8)	0.32 ± 0.036 (8)	1.4
Polypropylene	1.5 ± 0.09 (7)	1.3 ± 0.07 (7)	0.23 ± 0.059 (7)	0.61
1 K Polyurethane	0.47 ± 0.089 (7)	0.36 ± 0.041 (7)	0.30 ± 0.051 (7)	0.18
Polyvinylacetate	1.6 ± 0.41 (9)	1.1 ± 0.23 (6)	0.34 ± 0.062 (5)	0.60

Concerning the polymers tested in this study, datasheets of the producers are available for epoxy,[13] polyester,[14] and polypropylene.[15] For polyester the value published in the data sheet (3.5 GPa) corresponds well with results obtained from measurements with the mechanical extensometer (3.6 GPa). Also, values published by the producer for PP samples (0.9 to 1.5 GPa) correspond well with the elastic modulus of own measurements by mechanical extensometer (1.5 GPa). Measurements of Epoxy by means of the mechanical extensometer macrosense and ESPI resulted in an elastic modulus of 3.2 GPa (mechanical extensometer) and 3.4 GPa (ESPI), respectively, which is higher than the elastic modulus of 2.65 GPa given by the datasheet. However, an exact description of sample preparation, sample geometry and testing conditions determining the elastic modulus are not given in the datasheet. Therefore, a direct comparison of values is not reasonable due to different testing methods and conditions.

Jeandreau[16] identified possible Poisson's ratios of resins for the timber industry in the range of 0.3 to 0.5. The Poisson's ratios of polymers and resins of ESPI measurements range from 0.23 (PP) to 0.36 (polyester and epoxy). For epoxies, Poisson's ratios between 0.32 and 0.38[17,18] and for PUR values between 0.25 and 0.35[17] were reported, which agrees well with our own measurements of the Poisson's ratio of epoxy (0.36) and PUR (0.30), respectively.

CONCLUSION

It was shown that both macrosense measurements and ESPI are valid methods to measure elastic properties of adhesive polymers and matrix polymers frequently used in the wood and natural-fibre industry. Adhesives and matrix polymers cover a wide range of elastic properties which were determined successfully in this study and made available for further research.

W. Gindl and J. Konnerth gratefully acknowledge financial support by the Austrian Science Fund FWF.

References

1. Gindl, W.; Sretenovic, A.; Vincenti, A.; Müller, U. Holzforschung 2005, 59, 307.
2. Müller, U.; Sretenovic, A.; Vincenti, A.; Gindl, W. Holzforschung 2005, 59, 300.
3. Sretenovic, A.; Müller, W.; Gindl, W. Composites A, to appear.
4. Adams, R. D.; Coppendale, J. In Adhesion; Allen, K. W., Ed.; Applied Science Publishers: London, 1997; Vol. 1, p 1.
5. Bolton, A. J.; Irle, M. A. Holzforschung 1987, 41, 155.
6. Irle, M. A.; Bolton, A. J. Holzforschung 1991, 45, 69.
7. Da Silva, L. F. M.; Adams, R. D. J Adhesion Science Technol 2005, 19, 109.
8. Wooley, G. R.; Carver, D. R. J of Aircraft 1971, 8, 817.
9. Landolt. Zahlenwerke und Funktionen aus Physik, Chemie, Geophysik und Technik; Landolt-Börnstein: Berlin, 1950.
10. Broughton, J. G.; Hutchinson, A. R. Int J Adhesion Adhesives 2001, 21, 177.
11. Gindl, W.; Schöberl, T.; Jeronimidis, G. Int J Adhesion Adhesives 2004, 24, 279.
12. Jialanella, G. L.; Shaffer, E. O. J Adhesion Science Technol 1993, 7, 1171.
13. Conrad Electronic; Epoxidharze, Klebstoffe http://download.r-g.de/handbuch/kapitel02.pdf, 19.7.2005.
14. Gerber GFK-Systeme; Polyester–Laminierharz VIAPAL VUP 4782 BEMT, http://www.beigerber.de/Datenblatt%204782.pdf, 19.7.2005.
15. Goodfellow. Polypropylene PP film, PP301460/14 film, Goodfellow Cambridge Ltd.: Huntingdon, U.K.; http://www.goodfellow.com/csp/active/gfHome.csp, 19.07.2005.
16. Jeandrau, J. Int J Adhesion Adhesives 1991, 11, 71.
17. Duncan, B.; Dean, G. Int J Adhesion Adhesives 2003, 23, 141.
18. Jeandrau, J. Int J Adhesion Adhesives 1986, 6, 229.