INTERLAMINAR FRACTURE TOUGHNESS OF PEI AND PEEK/CARBON FIBRE COMPOSITES IN RELATION TO MATRIX VISCOELASTICITY

R. Frassine, M. Rink and A. Pavan *

The rate and temperature dependence of the interlaminar fracture toughness of two unidirectional carbon fibre composites having PEI and PEEK as matrices are studied in relation to the fracture toughness of the relevant matrix. The "time-temperature correspondence principle" has been applied to obtain master curves covering several decades of crack speed. While in the case of PEEK the interlaminar toughness of the composite is close to that of the pure matrix for all conditions examined, for PEI the toughness of the neat resin is not completely exploited in the corresponding laminate.

INTRODUCTION

Interlaminar crack propagation resistance is a fundamental property for the design of damage-resistant composite materials and parts. However, the resin fracture toughness can only partly be translated into delamination fracture toughness, owing to the fibre constraint to the development of large plastic zones [1]. This work is aimed at comparing the effect of rate and temperature on fracture toughness of two very ductile thermoplastic matrices and of their composites.

MATERIALS

Neat resins: Polyetherimide (PEI) produced by General Electric (ULTEM 1000), and polyetheretherketone (PEEK) produced by ICI (PEEK 150P).

Composites: AS4-3K-PEI prepreg produced by Ten Cate AC, with nominal fibre content of 58% by weight, and AS4-3K-PEEK hybrid yarn produced by BASF, with nominal fibre content of 61% by volume.

* Dipartimento di Chimica Industriale e Ingegneria Chimica
Politecnico di Milano, Piazza Leonardo da Vinci 32, 20133 Milano (Italy)

TESTING

Neat Resin Tensile testing: tests at varying displacement rates and temperatures were conducted on specimens prepared by saw-cutting of compression-moulded plates, following ASTM D638M-82 recommendation (Type M-II).

Neat Resin Fracture Testing: Double Torsion (DT) fracture tests were conducted at different displacement rates and temperatures on 50 mm wide and 150 mm long specimens saw-cut from 3.5-5 mm thick compression-moulded plates and V-grooved on both faces.

DT testing has been reviewed in [2]. By applying conventional Linear Elastic Fracture Mechanics, the strain energy release rate, G_{Ic}, and crack speed, $\dot{a}$, can be expressed as follows:

$$G_{Ic} = \frac{P_c}{2B}\frac{dC}{da} \qquad (1) \qquad\qquad \dot{a} = \frac{\dot{x}}{P_c \frac{dC}{da}} \qquad (2)$$

in which P_c is the load during crack propagation, B the specimen thickness under the groove, C the specimen compliance, a the crack length and $\dot{x}$ the applied displacement rate. Corrections proposed by Leevers [3,4] to account for crack curvature and load moment arm reduction during the test were applied to Eq. 1.

Interlaminar Fracture Testing: tests were conducted on composite laminates, 3-4 mm thick, using Double Cantilever Beam (DCB) testing geometry. Specimens having 20 x 170 mm in-plane dimensions with fibres oriented parallel to the major axis of the specimen were obtained by saw-cutting compression-moulded plates. A starting edge crack, about 50 mm long, was generated by introducing a very thin film (about 15 μm) in the interlaminar region at the mid-plane of the laminate. The crack growth during the test was videorecorded from the lateral side of the specimen.

The critical value of the strain energy release rate, G_{Ic}, was calculated by using the following expression [5]:

$$G_{Ic} = \frac{3\,\delta\,P_c}{2\,B\,a} \qquad (3)$$

in which P_c is the load, δ is the crack opening displacement, B is the specimen width and a is the crack length. Corrections were applied to account for end-tabs rotation during the test and for beam stiffening due to the bonded end-tabs (see [5]).

YIELDING RESULTS

Tensile yield stress values were obtained at varying strain rate and temperature on the two neat resins examined: a decreasing trend with increasing time and temperature is observed, which is a common behaviour for polymers.

For both materials, the single yield stress vs. time-to-yield isotherms were reduced by a horizontal shift along the logarithmic time axis to single master curves. Figs. 1 and 2 shows the master curves and the shift factors obtained. For PEEK, some measurements in plane-strain compression were also performed to obtain shift factor values at low temperatures, at which the material becomes brittle in tension.

For PEEK, as it can be seen in Fig.2, the shift factors derived from yield data in tension and compression superimpose very well with literature data [6] obtained from small-strain dynamic-mechanical measurements. These shift factors have been used in the following to reduce fracture data obtained at varying rate and temperature.

FRACTURE RESULTS

Neat resins - Experimental compliance calibrations, as in [7], were performed in DT at the different rates and temperatures examined, in order to obtain the dC/da factor to be introduced into Eq. 1.

During fracture, PEI showed continuous crack propagation at all deformation rates and temperatures examined, while PEEK showed continuous crack propagation at temperatures above 70°C and unstable (stick-slip) propagation below this temperature. At 70°C both types of fracture were observed, depending on deformation rate. The values of the fracture load, P_c, obtained during the tests, together with the values of dC/da previously derived, were used to calculate G_{Ic} and $\dot{a}$ via Eqs. 1 and 2. For PEEK, in the case of stick-slip behaviour, values of G_{Ic} at both initiation and arrest were determined and associated with a crack speed calculated using the value of P_c at crack initiation. Neither of the two materials showed clearly defined temperature and rate dependences.

As performed by other authors previously (see e. g. [8-10]), in Figs. 3 and 4 the fracture toughness data obtained at different temperatures have been reduced to a single reference temperature (23°C) shifting them along the logarithmic crack speed axis by the factor $\log(a_T^{23})$. Shift factors, a_T^{23}, where those obtained from the yield measurements. For both materials a far clearer picture of the fracture behaviour is so obtained. PEI (Fig. 3) shows a non monotonic trend in toughness, suggesting that different viscoelastic mechanisms may take place during fracture, depending on rate and temperature. For PEEK (Fig. 4), a monotonic decrease in toughness with increasing rate is observed and, with regard to the type of propagation, two regimes can be identified. At low rates (or high temperatures), stable ductile behaviour takes place, while at higher rates (or lower temperatures) unstable stick-slip is observed.

Composites - Crack propagation in DCB testing at varying displacement rate and temperature was always stable for PEI (as for its corresponding matrix), and always unstable (stick-slip) for PEEK. For the latter, at high temperatures and/or low rates, stable propagation was often observed between crack-jumps.

By shifting the data along the crack speed axis, as previously made for the neat resins, a master curve at a reference temperature of 23°C was derived for each material (Figs. 5 and 6). PEI toughness regularly increases with crack speed, which does not seem to reflect the trend observed on the neat resin. PEEK toughness shows a decreasing trend similar to that observed for neat resin.

DISCUSSION

Extensive investigation by Bradley [1] during the past decade clearly pointed out that resin toughness transfer to continuous-fibre reinforced composite materials may be a non-trivial question, owing to fibre constraint on the plastic zone. In the absence of additional dissipative mechanisms such as extensive microcracking or fibre pullout and break, the interlaminar fracture toughness of a composite is expected to be less, or at most comparable, to that of the matrix, depending on morphology and thickness of the interlaminar layers, and on spacing between fibres. In the present work, some conclusions could be reached as regards the use of PEI and PEEK as matrices for composites.

With PEI, the interlaminar fracture toughness of the unidirectional composite turned out to be lower, and less rate sensitive, than the fracture toughness of the relevant resin. With PEEK, the toughness of the composite is essentially the same as that of the neat resin over the entire range of testing conditions explored. It can be observed that, in the range in which the matrix showed unstable crack propagation, the composite appears somewhat tougher than the matrix: this effect may be attributed to defects or inhomogeneities introduced in the matrix resin during moulding (which is especially difficult with PEEK), which may play a major role in the case of brittle fracture of the neat resin. Finally, despite the fact that the PEEK composite showed unstable crack propagation over the entire rate and temperature ranges explored, its fracture toughness at crack arrest was found substantially higher than that of the matrix. In conclusion, only for PEEK does the composite structure appear optimised as regards the interlaminar fracture resistance.

ACKNOWLEDGEMENTS Special thanks are due to Drs. P. Rossi, S. Drali and A. Comini for contributing to the development of this work during their degree theses.

REFERENCES

[1] Bradley W. L., "Application of Fracture Mechanics to Composite Materials", edited by K. Friedrich, Elsevier Publ., 1989, pp. 159-187.

[2] Frassine R., Riccò T., Rink M., Pavan A., J.Mater.Sci., Vol.23, 1988, pp.4027-4036.

[3] Leevers P. S., J. Mater. Sci., Vol. 17, 1982, pp. 2469-2480.

[4] Leevers P. S., J. Mater. Sci. Letters, Vol. 5, 1986, pp. 191-192.

[5] ESIS-TC4 Polymers and Composites: "A Protocol for Interlaminar Fracture Testing of Composites. Mode I (DCB)" (May 1992).

[6] Ogale A. A., "Thermoplastic Composite Materials", edited by Carlsson, L.A Elsevier Publ., pp. 205-232, 1991.

[7] Frassine R., J. Comp. Mater., Vol. 26, 1992, pp. 1339-1350.

[8] Marshall G.P., Coutts L.H., Williams J.G., J. Mater. Sci., Vol. 9, 1974, pp 1409-1419.

[9] Kinloch A. J., Adv. Polymer Sci., Vol.72, 1985, pp. 45-67.

[10] Popelar C.F., Popelar C.H., Kenner V., Poly.Eng.Sci., Vol.10, 1990, pp.577-86

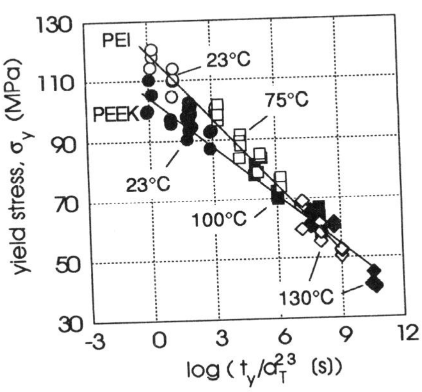

Figure1 Yield stress master curves at the reference temperature $T_0 = 23°C$.

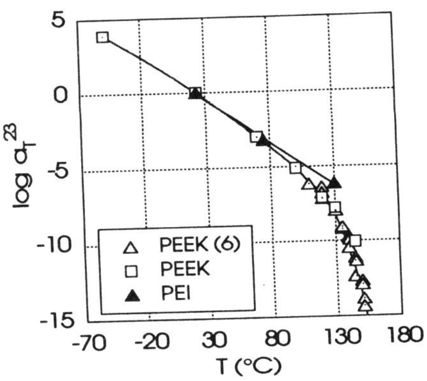

Figure 2 Shift factors referred to 23°C from time-temperature superposition.

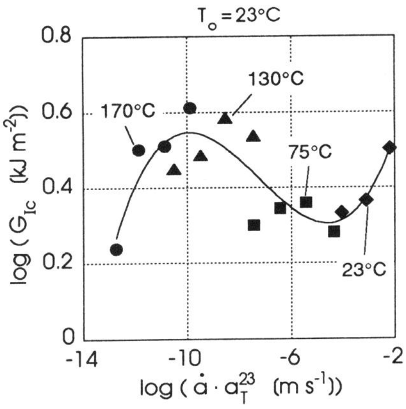

Figure 3 Fracture toughness vs. reduced crack speed master curve for PEI.

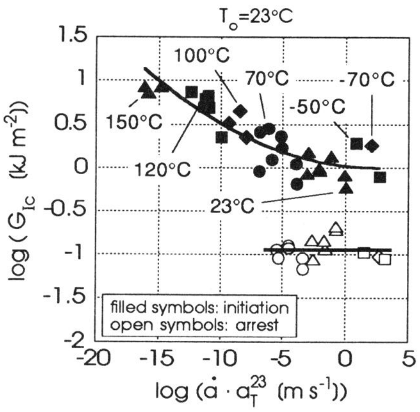

Figure 4 Fracture toughness vs. reduced crack speed master curve for PEEK.

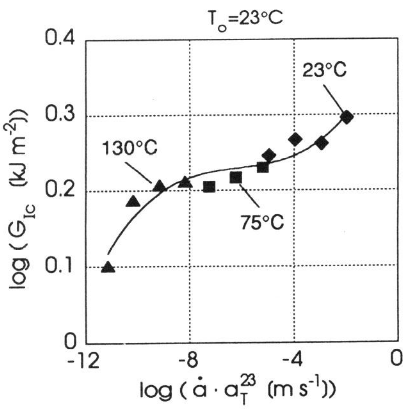

Figure 5 Fracture toughness vs. reduced crack speed master curve for PEI laminate.

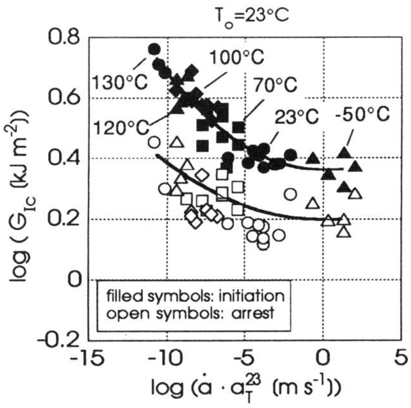

Figure 6 Fracture toughness vs. reduced crack speed master curve for PEEK laminate.