MEASUREMENT OF MICRO-MECHANICAL INTERFACIAL PROPERTIES OF SiC (Nicalon^{TM}) FIBRE/BOROSILICATE GLASS MATRIX COMPOSITES AT ELEVATED TEMPERATURES BY THE SEM-PUSH-OUT TECHNIQUE

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SUMMARY: Results on both room and elevated temperature interfacial testing conducted on SiC (Nicalon^{TM})-fibre reinforced borosilicate glass matrix composites by fibre push-out are presented. A scanning electron microscope push-out apparatus enabling testing and simultaneous observation of the experiment was employed. Using a conical diamond indenter with a flat end, single fibres were pushed out of thin polished composite sections in the temperature range from 23 to 500 °C. Applied force and displacement of the fibres were digitally monitored during debonding and frictional sliding. It was found that with increasing testing temperature, the interfacial shear stresses changed. Between 350 and 500 °C, a significant increase of the characteristic stresses was measured. This stress increment was very pronounced for T > 450 °C, and it was attributed to the variation of the properties of the glass matrix as the temperature approached the glass transition temperature (T_g = 530 °C). A slight decrease of interfacial shear stresses was measured at lower temperatures (T < 350 °C). This was attributed to the relaxation of internal stresses created in the composite due to thermal expansion mismatch.

KEYWORDS: interfacial properties, fibre push-out technique, interfacial shear stresses, glass matrix composites, high-temperature interfacial properties

INTRODUCTION

It is well-known that the mechanical behaviour of fibre reinforced composite materials is controlled to a great extent by the properties of the interfaces [1]. In ceramic and glass matrix composites, in particular, a relatively weak interface is required for interfacial debonding to occur in the stress field of a propagating crack [1,2]. Crack bridging and deflection along the interface result in desired fibre toughening. The properties that characterize the interface are

the interfacial shear strength, which controls the occurrence and extend of fibre/matrix debonding, and the frictional shear stress, which affects the load-carrying capability and the energy dissipation process during fracture [2].

A number of experimental methods have been and are being developed in order to measure with high accuracy the interfacial properties of composite materials. Among these, the push-out indentation technique is gaining wide acceptance among researches and engineers because it is simple to perform and highly reproducible. In this test, the fibre/matrix interfacial properties are obtained from load-displacement curves of indented (pushed) fibres [2,3]. The test was introduced by Marshall employing a diamond pyramid loading probe and a slice cut from the composite such that the faces are perpendicular to the axis of the fibres [3].

Experimental techniques have evolved with the increasing acceptability of the push-out test. Earlier work was conducted using metallurgical microhardness testers [4]. One of the disadvantages of these testers is the achievement of sufficient precision for positioning of the indenter above the fibres. To overcome this problem optical microscopes and video systems were integrated within the test instrument, increasing the sight field resolution. The first push-out tester allowing in-situ visual monitoring of the test was developed by Eldridge et al. in 1989 [5]. Later, Daniel and Lewis [6] developed a scanning electron microscope (SEM) based microindentor, which allowed microscopic observation of the fibre indentation process. This system was successfully used for interfacial testing of various ceramic matrix composites at room temperature [6]. However, the apparatus lacked a high-temperature testing facility, which is an essential requirement when testing ceramic composites due to their application potential at high temperatures. A push-out apparatus housed in a chamber of a SEM, with the capability to perform *in situ* tests at high temperatures (up to 1100 ºC), became available commercially in 1994 and first results of its use on metal, ceramic and glass matrix composites have been reported [7-10]. Another instrument with the capability to conduct push-out tests at high temperatures was described by Eldridge [11]. Using these devices, the applicability of the push-out technique can be broadened significantly and measurements on ceramic and glass matrix composites under conditions close to that expected to be encountered in practical situations are possible. Besides being useful in understanding the composite behaviour at elevated temperatures, high-temperature push-out testing also allows to distinguish between chemical and mechanical bonding at the interface [11]. In the present article, we report on the use of the SEM-push-out indentation technique to obtain the interfacial properties of SiC-fibre reinforced glass matrix composites at room and elevated temperatures.

EXPERIMENTAL METHODS

Material

The material investigated was a commercially available unidirectional SiC Nicalon^{TM} (NL202) fiber reinforced borosilicate (DURAN) glass matrix composite fabricated by Schott Glaswerke (Mainz, Germany); the composites were prepared by the sol-gel-slurry method [12]. Nominal properties of the matrix and fiber are given in Table 1. The density of the composites was 2.4 g/cm^3, and the fiber volume fraction ~ 0.4. A scanning electron microscopy (SEM) image showing the microstructure of an as-received sample in a plane perpendicular to the fiber axes is shown in Figure 1. The composite exhibits a fairly homogeneous distribution of the fibers and absence of porosity in the glass matrix. Since the thermal expansion coefficients of the matrix and fiber are very close (see Table 1), the internal residual stresses that develop in the samples upon cooling from the fabrication temperature are

low. The magnitude of such residual stresses in composites similar to those studied here were calculated by Klug et al. [13] using a simplified coated-cylinder model and found to be less than 20 MPa. Figure 2 shows a transmission electron microscopy (TEM) image of the fibre/matrix interface. The typical carbon-rich interface (about 20 nm thickness) can be appreciated. This interface provides a weak matrix-fibre bonding and therefore imparts the favourable non-catastrophic fracture behaviour to this material [9,12].

Table 1: Properties of the composite constituents [9]

Property	Matrix: DURAN™ (borosilicate glass)	Fibre: SiC Nicalon™ (NL 202)
Density (g/cm^3)	2.23	2.55
Young's modulus (GPa)	63	198
Poisson's ratio	0.22	0.20
Thermal expansion coefficient (K^{-1})	3.25×10^{-6}	3×10^{-6}
Tensile strength (MPa)	~ 60	2750
Fibre volume fraction: 0.4 Fibre radius: 7-11 μm Density of the composites: 2.4 g/cm^3		

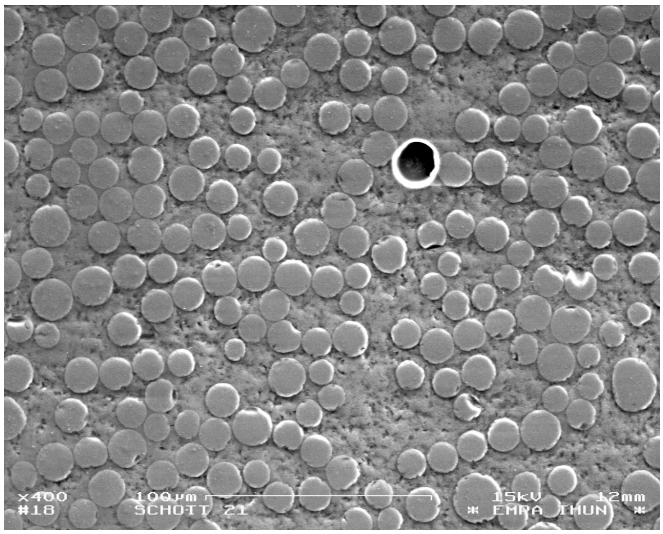


Fig. 1: SEM micrograph showing the microstructure of the investigated glass matrix composite. A hole left by a fibre after having been pushed-out is also shown.

The SEM-Push-out Apparatus

The instrument used combines the materials analysis capabilities of a SEM with mechanical testing [14]. It was constructed by adding a mechanical loading device to a SEM chamber (Zeiss DSM 962). With the indentor in the field of view of the SEM at high magnification, a single fibre from the composite specimen can be aligned with the indentor using the stage controls of the microscope. The resolution and depth of field of the SEM allow for detailed observation of the fiber/matrix interface while under dynamic loading. A hot stage is attached to the SEM chamber allowing elevated temperature tests to be conducted. In the present study,

using a conical diamond indenter with a flat end, single fibres were pushed out of thin (approx. 0.4 mm) polished composite sections in the temperature range from 23 to 500°C. The displacement rate used was 0.18 μm/s. For each temperature at least ten fibres were tested. Applied force and displacement of the fibres were digitally monitored during debonding and frictional sliding. The interfacial debonding shear strength (τ_{db}) and the interfacial friction shear strength (τ_{fr}) were determined from the load data using a linear model [15]:

$$\tau = P/2\pi rs \qquad (1)$$

where: τ is interfacial shear stress, P is the load at the point of interest, r is the fibre radius and s the specimen thickness.

The linear model is based on a force balance equation and assumes an uniform shear stress distribution, which in practice is only partially valid. However, the goal of the investigation was to compare the change of interfacial properties with temperature and not to determine the intrinsic absolute values. Thus, because of its simplicity, the linear model was used and the test conditions other than temperature were kept constant for comparison. For example, in order to eliminate the so-called Poisson effect when testing different fibres [10,16] (lateral expansion of the fibre on push-out), only fibres with similar diameters (in the range 17-21 μm) were tested. Moreover, all samples were polished to a similar thickness (in the range 390-410 μm). A more detailed description of the indentation push-out methodology is presented elsewhere [8].

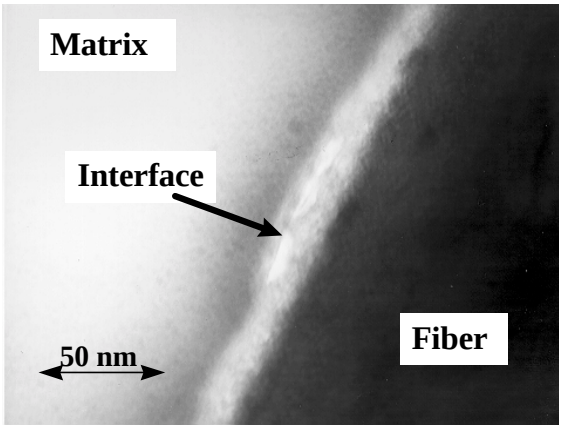


Fig. 2: TEM micrograph showing the carbon-rich layer at the fiber/matrix interface.

RESULTS AND DISCUSSION

Room temperature testing

Figure 3 shows the indentor acting on a SiC-Nicalon fibre in an as-received sample. The apparatus offered high accuracy for the indentor positioning, allowing the push-out process to be carried out without fibre or matrix damage. Figure 4 shows a typical interfacial shear stress-displacement curve during push-out indentation for a room temperature test. The interfacial shear stresses are calculated according to Eq. 1. It is seen that the shear stress increases linearly until the curve shows a kink and a slope change, indicating the onset of fibre-matrix debonding. The load continues to increase at a lower rate until reaching the maximum, at which point the debonding process has finished and the whole fiber starts to slide out. The quality of the interface is characterized by the value of the interfacial frictional shear strength (τ_{fr}), which is visualized by the plateau reached by the shear stress versus

displacement curves at large fiber displacements. It is seen that τ_{fr} has a value of τ_{fr} = 14 MPa ± 1 MPa, which is similar to values measured in equivalent glass matrix composites [10,17]. The debonding shear strength (τ_{db}) can also be obtained from the stress-displacement plot. A value τ_{db} = 21 MPa ± 2 MPa was determined, also in agreement with published data [10,17]. These measurements confirm the presence of a weak interfacial bonding in the as-fabricated composites, which is given by the carbon-rich layer at the matrix/fiber interface, as shown in Figure 2. This relatively weak bonding leads to fibre pull-out during composite fracture and thus to a typical "composite" failure behaviour with high fracture strain, as shown elsewhere [9].

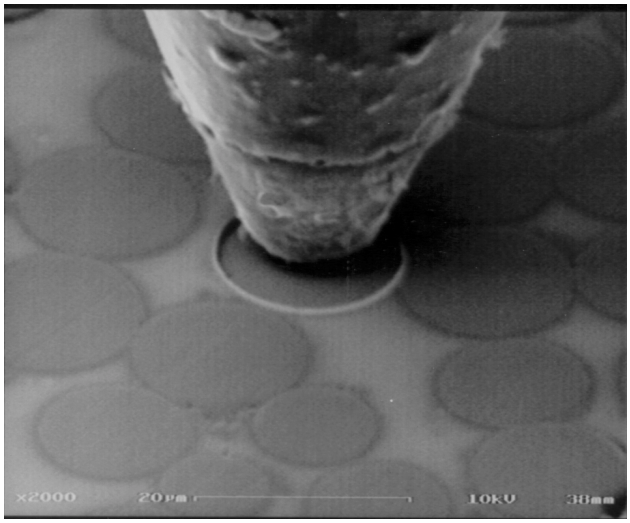


Fig. 3: SEM image of the indentor acting on a SiC-Nicalon^{TM} fibre during push-out test.

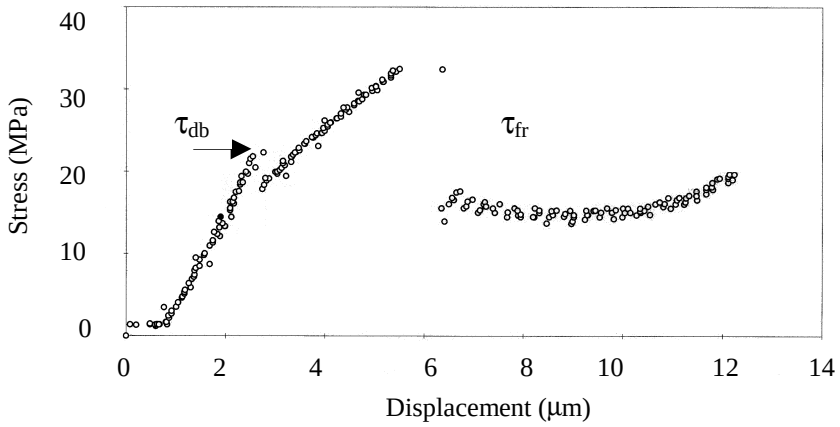


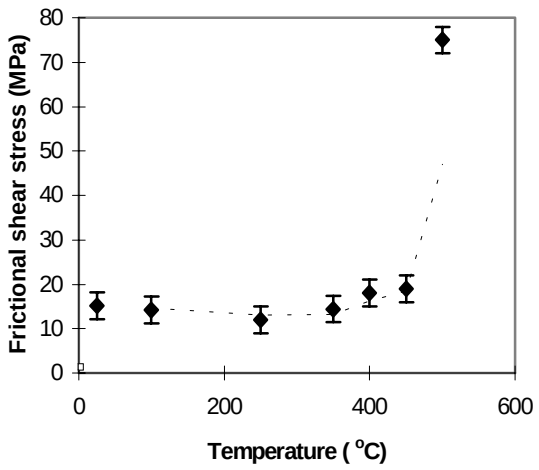
Fig. 4: Typical stress-displacement curve recorded during fibre push-out at room temperature in SiC (Nicalon^{TM}) fibre /borosilicate glass matrix composite

Elevated temperature testing

Since the present composite materials are designed for applications at temperatures between room temperature and about 550 °C [9], the variation of the interfacial properties in this

temperature range was investigated using the high-temperature stage of the push-out apparatus. The results for the variation of the interfacial frictional shear stress as a function of the testing temperature are shown in Figure 5. It is found that between 350 and 500 °C, a significant increase of the characteristic stresses occurred. This stress increase is very pronounced for T > 450 °C. A slight decrease of the interfacial shear stresses between RT and 300 °C can also be inferred from the measured values. However, due to the scatter of the data, this decrease may be an artefact of the measurement. Indeed more data are necessary to confirm this trend. Nevertheless the relaxation of internal residual stresses in the composite with increasing temperature may be the mechanism responsible for this initial transient decrease of the interfacial stresses. The strong effect of internal stresses on the interfacial properties of fibre reinforced glasses has been documented in the past [17]. Considering that the coefficient of thermal expansion of the matrix (α_m) is slightly higher than that of the fibre (α_f) (see Table 1), residual clamping stresses are develop, as calculated by Klug et al. [13]. These stresses will tend to diminish as the temperature increases, thus resulting in lower interfacial shear stresses, as measured by fibre push-out. Similar behaviour has been found for other composite systems having the same thermal expansion mismatch sign [11]. For composites with $\alpha_m < \alpha_f$ the opposite behaviour occurs, i.e. the interfacial shear stresses increase with increasing temperature [18]. In the present composites, for higher testing temperatures (T> 300 °C), however, other effects may become relevant, which are related to the change of the properties of the glass matrix at temperatures increasingly approaching its glass transition temperature (T_g = 530 °C) [19]. Although the Young`s modulus of borosilicate glass remains nearly constant from RT to T = 530°C [19], the viscosity of the glass will decrease and simultaneously the thermal expansion coefficient will increase as the temperature approaches T_g. The expansion and softening of the matrix at 500 °C may be responsible for the significant increase of the interfacial frictional shear stress, i.e. the fibres become firmly attached to the soft matrix as the sliding friction coefficient increases with decreasing matrix viscosity. A similar effect relating viscous flow of the glass matrix with the interfacial characteristics in model fibre reinforced glass matrix composites has been suggested by Barsoum and Elkind [20]. Yielding of the matrix at elevated temperatures complicates the interpretation of the push-out results because matrix yielding plays a major role in the interfacial failure process [11]. The qualitative explanation of the interfacial behaviour of glass matrix composites at elevated temperatures, based on the decrease of matrix viscosity and increase of thermal expansion coefficient of the glass matrix near T_g, may form the basis for developing a quantitative model to describe the problem, this being the focus of current research.

Fig. 5: Variation of the interfacial frictional shear stress (τ_{fr}) with testing temperature



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