Polymer Bulletin 27, 109–116 (1991)

Polymer Bulletin

Microstructural aspects of fracture and fatigue behavior in short fiber-reinforced, injection-molded PPS-, PEEK- and PEN-composites*

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ABSTRACT

Both the fracture and fatigue behavior of temperature-resistant thermoplastic matrix composites with discontinuous fiber reinforcement are strongly affected by the microstructure. The molding-induced microstructure of the composites can be characterized by a reinforcing effectiveness parameter (R). This parameter treats a short fiber reinforced injection-molded composite as a laminate, in the layers of which the fibers are present in different amounts, in various orientations and in addition, in various aspect ratios and aspect ratio distributions. The relative change in the fracture toughness can be predicted by the microstructural efficiency concept (M). Due to analogies between static fracture and fatigue crack propagation (FCP) results, this concept seems to work well when the dependence of FCP on microstructural details is considered.

INTRODUCTION

Injection-molded, short glass (GF) and carbon fiber (CF) reinforced composites with temperature resistant thermoplastic matrices such as polyetheretherketone (PEEK), polyphenylenesulfide (PPS) and polyethernitrile (PEN) are potential candidates for being used in technical domains, so far occupied by metallic and ceramic materials. Short fiber reinforcements provide high stiffness, strength, heat and dimensional stability to these matrices making them suitable for applications in the automotive, transportation, electric and electronic industry. These applications require from the materials a high resistance to fatigue and to fracture at both dynamic (impact) and static loadings. In the present paper it is outlined how the fracture and fatigue performance of composites with discontinuous fiber reinforcement can be correlated with their microstructural details (fiber volume fraction (V_f), layer structure, fiber orientation, fiber aspect ratio and its distribution) generated by the injection molding process.

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*Work presented at the ICCM-VIII conference (15–19, July, 1991) in Honolulu, USA

EXPERIMENTAL

Materials Film-gated injection-molded plaques of 3-4 mm thickness with GF and CF reinforceemnt in the range of 10 to 30 wt.% were supplied by the LNP Engineering Plastics, Malvern,PA,USA (PPS composites based on a Ryton grade of Phillips Petroleum,Bartlesville,OK,USA), by ICI Ltd, Wilton, UK (Victrex PEEK 450 G based composites) and by Idemitsu Kosan Co, Chiba,Japan (PEN ID 300 based composites). Their characterization - with the only exception of PEN-composites - can be found in references [1-3].

Testing Compact tension (CT) specimens were cut from the injection molded plaques and notched either parallel or perpendicuar to the mold filling direction (MFD). Static fracture tests were performed on a Zwick 1445 type tensile machine at different temperatures and deformation rates. The fracture toughness (K_c), was calculated according to the ASTM E 399 standard. Fatigue crack propagation (FCP) measurements were carried out in tensile-tensile mode (R=0.2) on a Schenck PSA 10 kN servohydraulic machine at ambient temperature using the same CT specimens and a sinusoidal wave function of 5 Hz frequency. Other necessary experimental details can be taken from our previous works (e.g.[1-4]).

RESULTS AND DISCUSSION

Microstructure Development The molding-induced microstructure both of the unfilled matrices and the short fiber reinforced grades can be approached by the flow model of Tadmor [5] and Rose [6]. This model considers the shear and elongational flow field developed during the cavity filling process. The molding-induced matrix morphology of these high melting temperature plastics of polycondensation type and with a T_g far beyond the mold temperature is a rather simple one composed of skin and core layers [1,2,4]. It was suggested [4] that this skin-core structure affect the fracture and especially the fatigue performance considerably.
The reinforcing fibers, on the other hand, may adopt a complex alignment due to the dominating flow field. As a result, several layers with different planar fiber orientation can be distinguished across the specimen thickness. Due to the rather low molecular mass of the virgin polymers - which hinders the formation of a shear zone with different fiber orientation - the fiber layering can well be approximated by a 3-ply laminate model composed of one central (C) and two surface (S) layers. In these layers the mean fiber orientation (f_p) was characterized by a modified Hermans-type parameter (Figure 1). It has been also shown that the fiber layering and orientation depend on V_f, on the fiber aspect ratio (l/d) and - provided that the diamater of the fiber is the same - on the fiber length distribution (l_W/l_N- where subscripts W and N relate to the weight and number average) [7]. The microstructure of the short fiber reinforced composites with respect to the loading direction can be characterized by the reinforcing effectiveness parameter (R) [8-9]:

$$R=\left(\frac{2S}{B}\cdot f_{p,eff,S}+\frac{C}{B}\cdot f_{p,eff,C}\right)\cdot V_f\cdot\left(\frac{l}{d}\right)\cdot\left(\frac{l_N}{l_W}\right) \tag{1}$$

where B is the specimen thickness and all other terms were defined before.

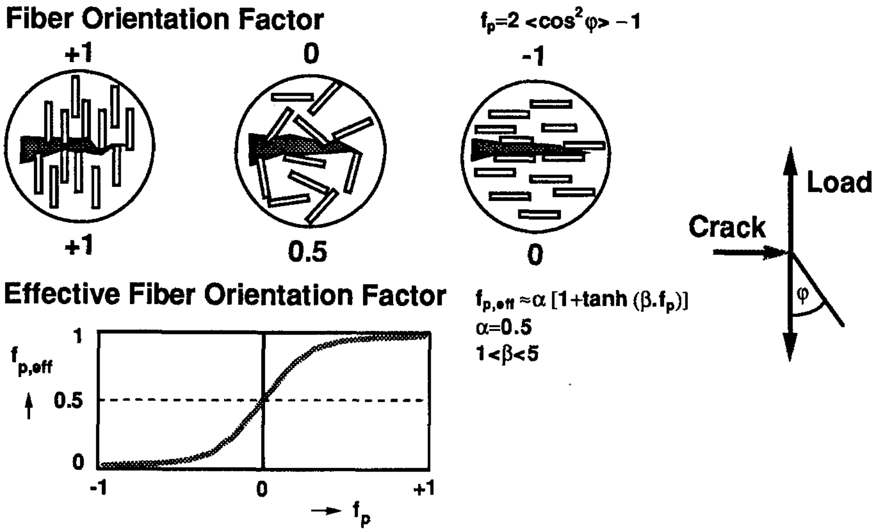

Figure 1. Relation between fiber orientation and effective fiber orientation with respect of the loading direction

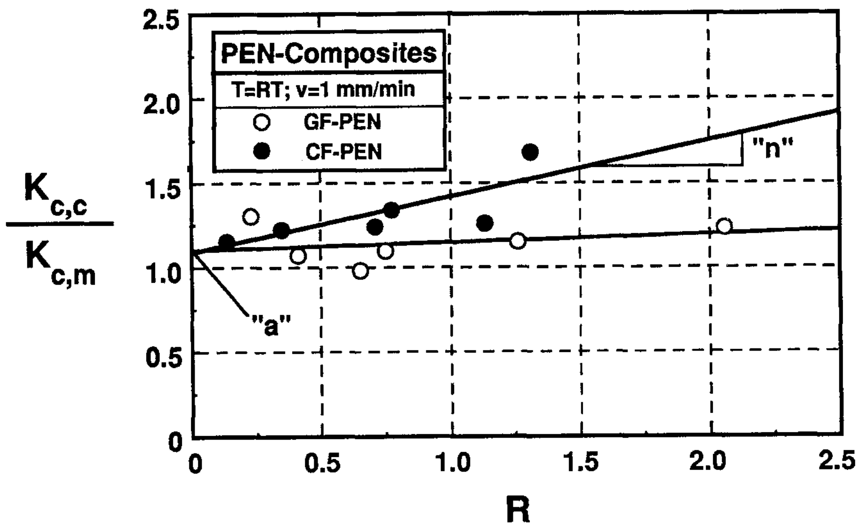

Figure 2. Evaluation of the factors "a" and "n" on the example of GF- and CF-reinforced PEN composites (Testing conditions: T=20 °C, v= 1 mm/min crosshead speed)

Fracture Toughness The microstructural efficiency concept [8] seems to be a powerful tool to estimate the change in fracture toughness due to reinforcement. According to this concept, wich belongs to the scope of the linear elastic fracture mechanics (LEFM), the fracture toughness of the composite related to the matrix at a given testing condition linearly depends on R (cf.Figure 2):

$$\frac{K_{c,c}}{K_{c,m}} = M = a + n.R \tag{2}$$

where

$K_{c,c}$ and $K_{c,m}$ - are the fracture toughness of the composite and matrix, respectively

a - is the matrix stress conditon factor

n - is the energy absorption ratio.

The relative improvement in the toughess of the composite is plotted against the reinforcing effectiveness (R) for the PPS-, PEN- and PEEK-composites in Figure 3. Based on Figure 3 one can conclude that with increasing matrix toughness (as inherent property or generated by the testing conditions) the energy adsorption ratio (n) decreases.The sign of "n" is positive in these materials indicating that the energy absorption by fiber-related failure events (i.e. fiber fracture, pull-out and fiber/ /matrix debonding) is higher than the matrix-related one (e.g. crazing, shear deformation,fracture). For GF-PEEK and GF-PEN n≈0 which means that the failure is practically matrix-dominated, i.e. incorporation of fibers does not change the failure mode due to the high matrix ductility.

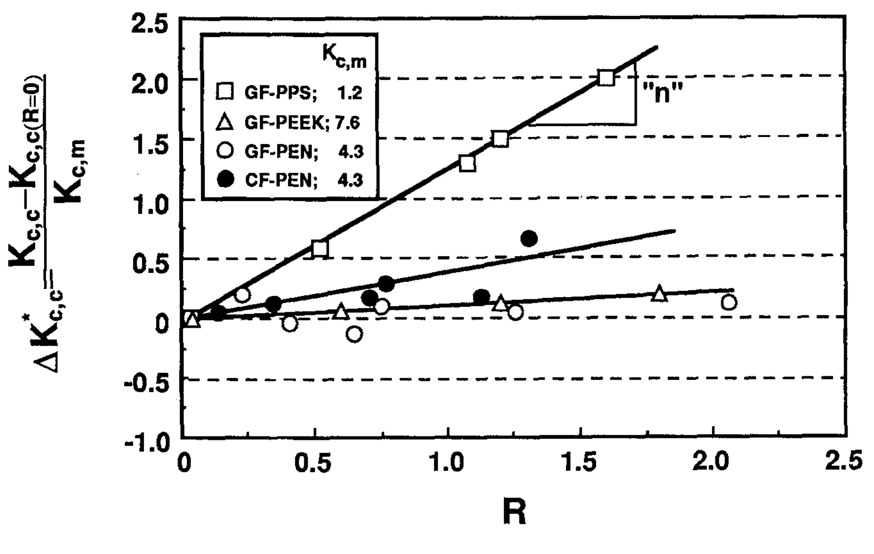

Figure 3. Relative effectiveness of toughness improvement by fiber reinforcement in the polymer matrices studied (Testing conditions: T=20 °C, v= 1 mm/min crosshead speed); Note:$K_{c,m}$ is given in $MPa.m^{1/2}$ units.

The characteristic failure in function of temperature and deformation rate are often summarized in failure maps [1,2,10-11]. Such a failure map (cf. Figure 4) gives instructions for example

i- how the high toughness range of PEEK could be extended towards higher temperatures (e.g. by copolymerization, crosslinking) or towards higher deformation rates (via toughening, increasing the molecular weight) - as it can be worked out from the related patents.

ii- which are the basic fiber related failure events to be studied for further material improvement (in the high toughness range pull-out processes are of basic importance according to Figure 4).

Mapping of the fracture toughness or fracture energy as a function of the temperature- and frequency-dependant E-moduli (fracture maps) delivers the right informations for design and construction purposes.For PEEK and PPS such fracture maps were constructed and published [1,2,11].

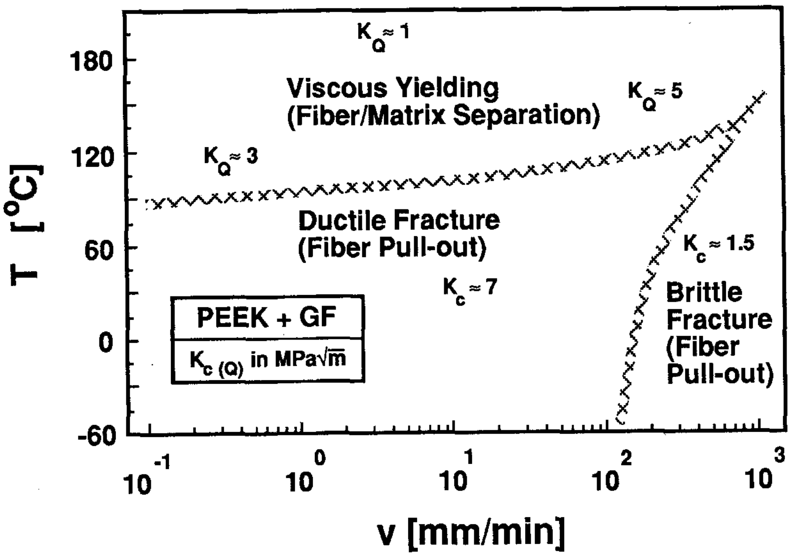

Figure 4. Failure map showing characteristic failure modes and fracture toughness values for the GF-reinforced PEEK; Note:K_Q is out of the valid range of the LEFM.

Fatigue Crack Propagation (FCP) Under cyclic loading conditions final breakdown of the short fiber reinforced composites is controlled by FCP instead of a crack initiation process. This is due to the fact that inherent failure sites ("notches") in form of structural inhomogeneities are always present [12]. The stable FCP range generally can be described by the well-known Paris-Erdogan power law [13]:

$$\frac{da}{dN}=A.(\Delta K)^{m} \tag{3}$$

where
A and m - are constants
da/dN - is the FCP rate and
ΔK - is the amplitude of the stress intensity factor.

The effects of the microstructural constituents on the FCP run are summarized schematically in Figure 5 on the example of GF- and CF-PEN composites. Since the upper bound of the validity of the Paris range is at about K_{cc} and the related failure events are also very similar at subcritical (FCP) and critical loadings (static fracture), the fatigue and fracture results are interrelated. Thus increasing R results in improved resistance to FCP of these materials [3-4,11] (cf.Figure 6).

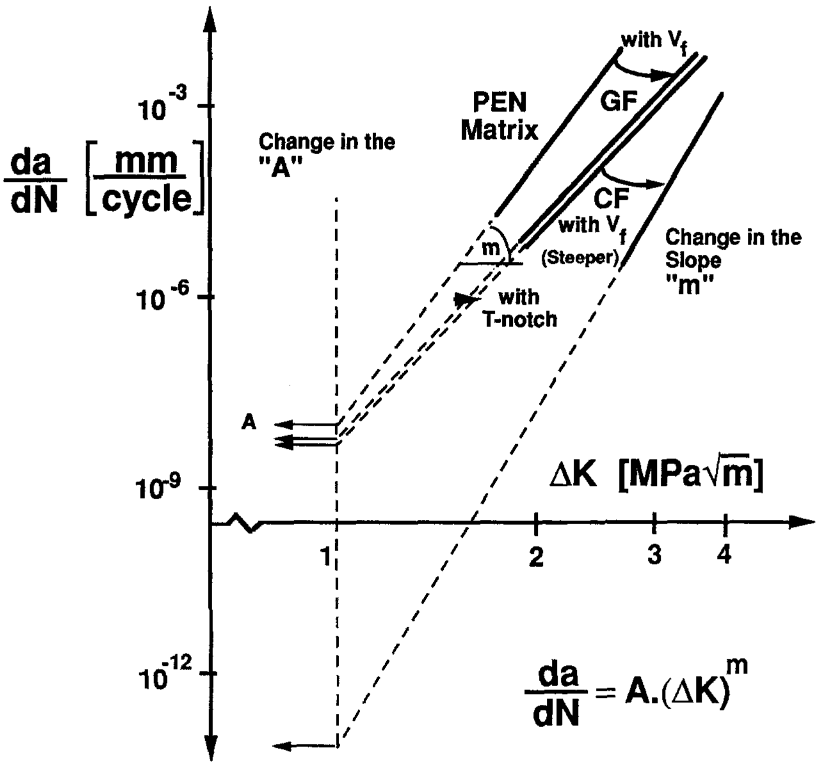

Figure 5. Effects of microstructural factors on the the FCP response on the example of short fiber-reinforced PEN-composites, schematically

Based on Figure 5 Equation 3 can be modified assuming that its constants are functions of M:

$$\frac{da}{dN}=\left(\frac{A}{M}\right).(\Delta K)^{m(M)} \tag{4}$$

$$\log\left(\frac{da}{dN}\right)=\log A-\log(a+n.R)+m(M).\log(\Delta K) \tag{5}$$

Equation 5 simplifies further as m(M) or m(M).log(ΔK) are considered as constants.This occurs when "m" only slightly changes with "M", which is the case when "n" tends to 0. In this way, the log(da/dn) vs. log M data pairs lay on a straight line, which was reported for PEEK-composites, indeed [3]. It seems that the last assumption is the easier fulfilled the lower is the energy absorption ratio (n), or with other words, the higher is the ductility of the matrix. It is worth mentioning, that the simplified version of Equation 5 works well, provided that the valid Paris-range on the ΔK-scale is broad enough.

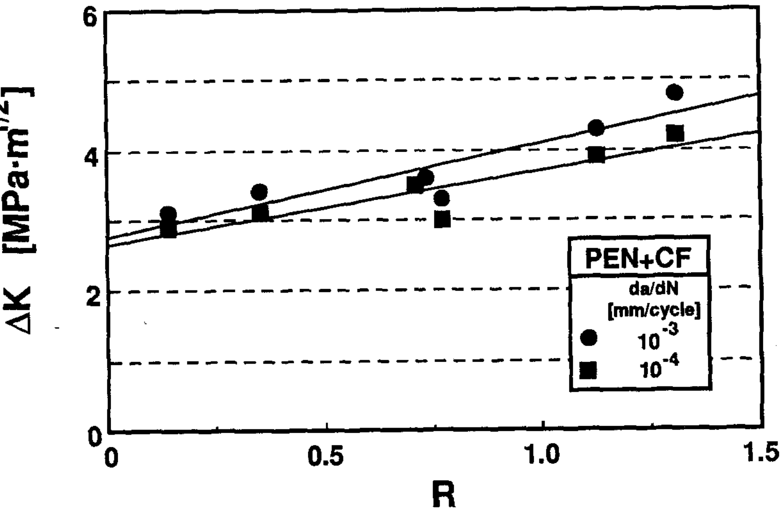

Figure 6. ΔK ranges at selected FCP rates for the CF-PEN composites

CONCLUSIONS

Improvement in fracture toughness and fatigue crack propagation behavior achieved by fiber incorporation highly depends on the matrix characteristics. This effect is the stronger the lower is the matrix toughness. Both the fracture toughness and the fatigue crack propagation (FCP) response can be correlated to the microstructure of the composites by means of the microstructural efficiency factor (M). This "M" term counts for the effects of the reinforcing fibers incorporated, by considering their relative effectiveness with respect of the loading direction (R), the change in the matrix stress state (a) and the variation in the energy absorption balance between fiber- and matrix-related failure events (n) induced by the fibers present. Fractographic analysis revealed that the dominant failure events are practically the same - excluding the effects of crack tip heating - in static fracture and FCP tests. This is a further argument for the reliability of this microstructural efficiency concept.

ACKNOWLEDGEMENT

Financial support of this study by the Deutsche Forschungsgemeinschaft (FR 675/2-2) is gratefully acknowledged. Thanks are also due to ICI Ltd. and Idemitsu Kosan Co.for providing the materials investigated.

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Accepted July 23, 1991 C