

# Blends of epoxy resin and polyphenylene ether as a matrix material for high-performance composites

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# BLEND OF EPOXY RESIN AND POLYPHENYLENE ETHER AS A MATRIX MATERIAL FOR HIGH-PERFORMANCE COMPOSITES

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Poly(2,6-dimethyl-1,4-phenylene ether), PPE, has been introduced as a matrix material in carbon fabric reinforced composites, using epoxy resin (DGEBA) as a reactive solvent. After impregnation the epoxy resin solvent is converted into non-solvent and phase separation is initiated. The resulting thermoplastic composites reveal a unique morphology of epoxy coated fibres in a pure PPE matrix. The epoxy interphase provides a high level of interfacial adhesion. Additionally, it is shown that multiple brittle failure of the epoxy interphase during mode-II loading (impact) is effective in delocalizing damage. This could be a new toughening mechanism for thermoplastic composite materials.

## INTRODUCTION

An interesting polymer to apply as matrix material for high performance composites is poly(2,6-dimethyl-1,4-phenylene ether) or PPE. PPE is a relatively low cost amorphous thermoplastic with a high  $T_g$  of approximately 220 °C and excellent mechanical properties e.g. in terms of toughness. However, as a result of its limited thermal and oxidative stability PPE can not be melt processed. The high temperatures required for melt processing (300 °C à 350 °C) will yield severe degradation and therefore PPE is usually classified as intractable. Consequently, a solution impregnation route has to be considered. However, solution impregnation techniques will introduce additional problems related to the complete elimination of residual solvent from the composite material.

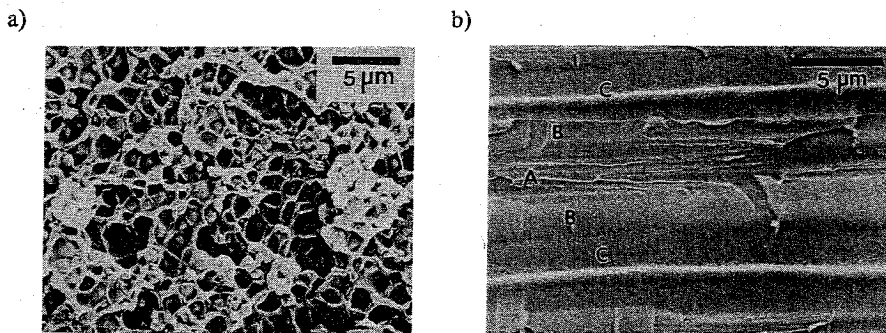
A challenging solution to these problems can be found in the use of reactive solvents. In a previous study [1], a reactive processing technique was explored for PPE, based on the use of standard bisphenol A epoxy resin as reactive solvent. In contrast to the literature [2-4] on blends of epoxy resins and engineering plastics, which mainly aims at toughened epoxy systems or hybrids of thermosets and thermoplastics, in this processing technique, basically, the epoxy resin is only introduced in order to tune the processability of PPE. In accordance with conventional solvent techniques this results in enhanced flow and reduced processing temperatures. However, instead of the complicated solvent recovery step required in conventional solvent techniques, the homogeneous solution is cured after processing and consequently the epoxy resin solvent is converted into non-solvent. At the high PPE contents of interest, > 40wt.%, phase separation is accompanied by phase inversion. Consequently, neat PPE is regained and the solvent is converted into a dispersed phase. Due to this morphology the properties of the final material are dominated by PPE.

In this investigation [5,6] the reactive processing technique is studied as a route to introduce PPE in high performance composite structures. Attention is mainly aimed at the composite morphology and toughness.

## RESULTS AND DISCUSSION

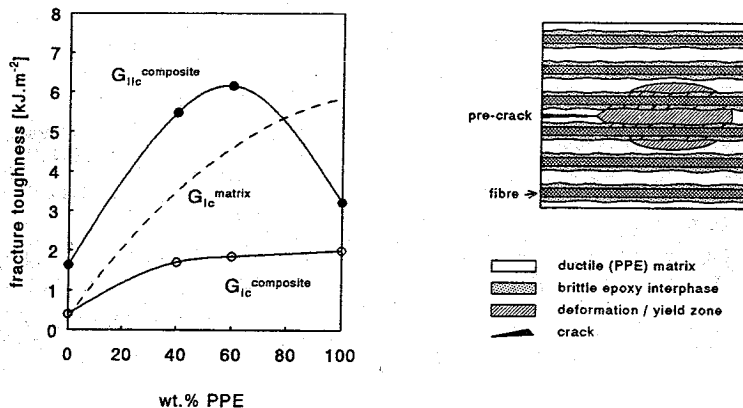
The epoxy/PPE system (Epikote 828EL, Shell, PPE 803, 30 kg·mol<sup>-1</sup>, GE plastics) was applied as a matrix material in continuous carbon fibre reinforced composites. Using epoxy resin as a reactive solvent, the viscosity of PPE at an impregnation temperature of 225 °C could be reduced to 30 and 500 Pa.s for 40 and 60 wt.% PPE, respectively. These medium viscosity compounds can be relatively easily used in a film-stacking technique to prepare carbon fabric (CD168, Ten Cate) reinforced composites with a fibre volume percentage of 50 % under low pressure conditions. Optical microscopy studies demonstrated that the reactive processing technique resulted in PPE based composites exhibiting a low void content and an excellent degree of impregnation.

A more detailed study of the morphology of the carbon fabric laminates revealed a pronounced influence of the presence of fibres on the matrix morphology. In composites epoxy resin preferentially accumulates at the fibre surface resulting in an epoxy 'interlayer' around the fibre. This effect is shown to be dependant on the fibre volume fraction. In contrast to the phase inverted morphology of epoxy spheres dispersed in a PPE matrix as observed in the unreinforced epoxy/PPE system (*Figure 1a*), in fibre-rich areas of the composite, in the PPE-rich matrix no epoxy spheres are distinguished (*Figure 1b*). Consequently, the majority of epoxy resin is migrated to the fibre surface resulting in a unique composite morphology of epoxy coated fibres in a nearly pure PPE matrix. Based on these morphological results it is concluded that epoxy resin is not only effective as a solvent enabling the preparation of PPE based composites, but additionally provides an important structural part of the final composite, i.e. the interphase, resulting in excellent adhesion.



**Figure 1:** SEM micrograph of fracture surface of cured epoxy/PPE (60 wt% PPE) blend; a) in the pure material and b) in a carbon fabric reinforced composite (50 vol.% fibre). A: fibre(imprint), B: epoxy interphase, C: PPE matrix.

In *Figure 2* the mode-I and mode-II interlaminar fracture toughness,  $G_{Ic}$  and  $G_{IIc}$ , obtained for the composites based on the neat constituents and the blends are presented together with the  $G_{Ic}$  of the pure matrix materials. As shown by Bradley *et al*[7] no linear correlation exists between matrix and composite mode-I fracture toughness. For ductile matrix materials the neat resin fracture toughness is only partly translated into the composite fracture toughness and an 'upper limit' for  $G_{Ic}^C$  is found of approximately 2 à 3  $\text{kJ}\cdot\text{m}^{-2}$ . The low  $G_{Ic}^C$  values are related to the high volume fraction of high modulus fibres which constrain the development of a large plastic zone at the crack tip. For the epoxy/PPE based composites this limit is reached already at low PPE contents. A positive consequence is that the penalty in matrix toughness due to the use of epoxy resin as a reactive solvent is negligible and barely affects the composite performance. The mode-II tests, however, reveal a remarkably strong synergy in  $G_{IIc}$  for the epoxy/PPE based laminates. In contrast to the pure PPE composites where mode-II tests yield only a minor increase in  $G_c$  ( $G_{IIc}=1.6 \times G_{Ic}$ ), the pure epoxy as well as the epoxy/PPE composites reveal in mode-II test conditions pronounced higher  $G_c$  values compared to mode-I test conditions ( $G_{IIc}=4 \times G_{Ic}$  and  $G_{IIc}=3.3 \times G_{Ic}$ , respectively). In the basically brittle epoxy resin composites this is the result of a deformation mechanism involving extensive microcracking ahead of the crack tip, referred to in the literature [7] as 'hackling'. This cracking mechanism is very effective in increasing the amount of energy absorbed. Obviously, such a change in deformation mechanism has also to occur in the epoxy/PPE composite. Taking into account the unique composite morphology this may give the explanation for the synergy observed. Provided that cracks in the epoxy interlayer can initiate yielding in the PPE-rich phase, multiple brittle fracture of the epoxy interlayer during mode-II loading can effectively delocalize yielding of the PPE-rich phase and subsequently increase the deformation volume and  $G_{IIc}^C$ . A schematic presentation of the influence of brittle interphases on the mode-II deformation mechanism in ductile thermoplastic composites is shown in *Figure 3*.



*Figure 2:* Matrix mode-I fracture toughness and composite (50 vol.% fibre) mode-I and mode-II fracture toughness versus matrix composition.

*Figure 3:* Schematic presentation of the mode-II deformation mechanism in ductile thermoplastic composites including brittle epoxy resin interphases.

In order to evaluate the toughening effect observed, a new epoxy resin solvent system is applied, based on a range of homogeneous blends of bisphenol A and polypropylene oxide epoxy resins (Epikote 828 and Epikote 877, Shell). By means of this reactive solvent system, the strain at break of the in-situ polymerized glassy interphase can be varied from 5% up to 80%. In Figure 4 the mode-I and mode-II fracture toughness of an epoxy/PPE (60 wt% PPE) composite as a function of the ductility of the interphase is presented. With increasing ductility,  $G_{Ic}$  increases slightly while a pronounced drop in  $G_{IIc}$  is observed. The results clearly support the toughening effect of brittle interphases: Upon increasing the interphase ductility, multiple fracture of the interphase and the consequent increase in  $G_{Ic}$  is suppressed, yielding in the end a deformation mechanism which is independent of the loading conditions and corresponds with the deformation of a composite based on a homogeneous ductile matrix material.

Surprisingly, the results reveal that a brittle interphase is effective in increasing the toughness of a ductile thermoplastic composite. This observation is of particular interest, since in the existing literature attention is mainly directed towards the introduction of ductile or rubbery interphases [8]. In future, more results will be presented on this intriguing feature.

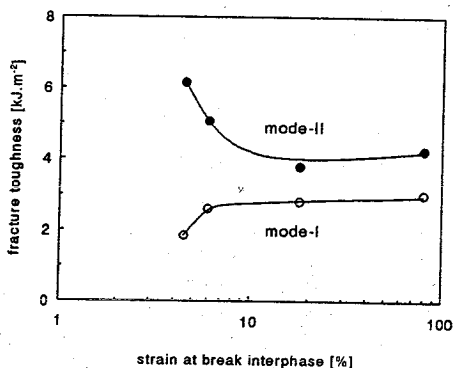


Figure 4: Mode-I and mode-II fracture toughness of an epoxy/PPE (60 wt% PPE) carbon fabric reinforced composite versus the strain at break of the interphase.

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