INTERFACES IN FLAX FIBRE-POLYMER COMPOSITES

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SUMMARY: This paper aims to study the interface of flax fibre reinforced composites based on different matrices (thermoplastic or thermosetting) according to the Kelly-Tyson model using the pull-out test. Pull-out tests of two types of single flax fibres: untreated Green and treated Duralin embedded in different polymer matrices: unsaturated polyester, epoxy and low density polyethylene were carried out. Higher interfacial shear stresses were observed for the single fibre composites with Green flax whereas Duralin-epoxy composites exhibited the highest interfacial shear stress. The inhomogeneity of the structure in lignocellulose fibres makes the interpretation of pull-out tests difficult. Results suggested that the interfaces within the treated and untreated flax fibres are weaker compared to the interface between the fibre and the matrix itself.

KEYWORDS: natural fibres, polymer composites, interface, interfacial shear strength, pull-out tests, micromechanical analysis.

INTRODUCTION

Ecological concern has resulted in a renewed interest in natural materials. Issues such as recycling and environmental safety are important for the introduction of new materials and products. Lignocellulosic natural fibres such as flax exhibit excellent mechanical properties, low density and low price. This excellent price-performance ratio at low weight in combination with the environmental friendly character is very important for the acceptance of natural fibres in large volume engineering markets such as construction industry. The emphasis on research to date in natural fibre polymer composites has been on the important area of the interface between constituent components [1-5]. The properties of the interface which are affected by the chemical nature as well as the physical and thermodynamic compatibility between the constituents components limit the overall performance of the composite and therefore a carefully examination of the microstructure gives important information for the development of high performance composites. There is a number of experimental techniques developed to characterise the interfacial properties of fibre reinforced composites. Such techniques include micro-debonding, pull-out tests and micro-fragmentation tests.

The structure of flax is very complicated because it is comprised of different structures of cellulose, hemicellulose and lignin. Hemicellulose acts as a coupling agent between lignin and cellulose. The outer skin is porous and consists also of pectin and other non-structural carbohydrates. The elementary fibres are bound together in a single fibre with the help of the lignin-hemicellulose matrix.

Kelly and Tyson [6] initially modelled the interfacial debonding and frictional pull-out test for ductile matrices assuming a uniform interfacial shear strength. According to this model the interfacial shear strength τ can be calculated by the following equation:

$$\tau = P / D \pi L \qquad (1)$$

where P is the maximum debonding stress, D the diameter of the fibre and L the embedded length. This study aims to characterise the interface of flax fibre reinforced composites based on different matrices (thermoplastic or thermosetting) according to the Kelly-Tyson model using the pull-out test. The lack of homogeneity in this kind of fibres makes the interpretation of micromechanical analysis and in this case the pull-out test very difficult.

EXPERIMENTAL

The flax fibres used in this study were of two types: Green and Duralin (supplied by CERES BV, The Netherlands). The latter is an upgraded flax fibre [7,8]. During this upgrading process the hemicellulose and lignin are depolymerised into lower molecular aldehyde and phenolic functionalities, which are combined by a subsequent curing reaction into a water resistant resin. Individual Green and Duralin fibres were carefully separated out by hand. Single Green- and Duralin flax fibre-unsaturated polyester (Viapal UP 223 BS/65, Vianova Resins) specimens were specially prepared for pull out tests using a combination of ‘droplet’ and ‘thick film’ technique. The resin was left to be cured at room temperature for 24 h. Pull-out tests were also conducted for single Green- and Duralin flax fibre-polyethylene (LDPE, Riblene, Enichem, MR30) and Duralin-epoxy resin (Rutapox L20, Bakelite) specimens. The specimens were prepared by embedding a single fibre into a drop of polymer matrix at 200^{o}C in the case of polyethylene and at 80^{o}C for 6 h in the case of epoxy resin. The pull out tests were carried out with the help of a microtensometer with specially designed grips at room temperature. The pull-out tests of Green-, Duralin-polyethylene and Duralin-epoxy were carried out with the help of a single fibre pull-out test machine. After the pull-out tests the surface of the fibre and polymer were examined under a Scanning Electron Microscope.

RESULTS AND DISCUSSION

Table 1 shows the results from the analysis of the Load-Displacement curves taken during the pull-out tests for Green and Duralin flax fibres embedded in unsaturated polyester resin polyethylene and epoxy resin. It can be observed that the interfacial shear strength is higher for the untreated green fibres than the treated (Duralin). In the case of polyethylene the difference is very small. On the other hand the Duralin-epoxy single fibre composites exhibit the highest interfacial shear strength.

Table 1: Shear strength of single Green, Dew-retted and Duralin flax fibre reinforced polyester composites calculated from pull-out tests.

Composite	Average shear strength τ (MPa)
Green-polyester	17.1
Duralin-polyester	11.7
Duralin-epoxy	23.2
Green-polyethylene	5.4
Duralin-polyethylene	4.4

The Load –Displacement curves for Green and Duralin flax fibres embedded in polyester resin are similar in shape for both fibres suggesting small frictional forces. Unstable debonding and pull-out was also observed, however this may be due to sample preparation as well as to the sensitivity and rigidity of the microtensometer.

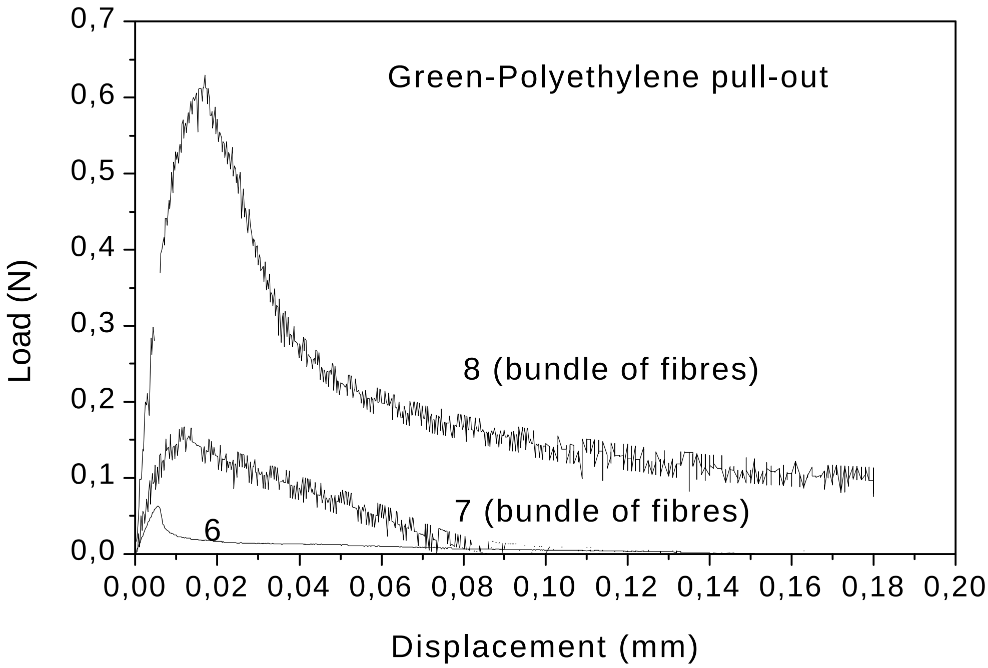

Figure 1: Load-Displacement curve for Green-Polyethylene single fibre composites

Figures 1 and 2 show Load-Displacement curves for Green and Duralin fibre, respectively, embedded in polyethylene matrix. The curves show an unstable debonding and pull out. In only one out of ten Green-polyethylene specimens, a part of the fibre broke before the pull out and remained within the matrix indicating weak interfaces within the fibre (SEM micrograph, Fig. 3). The friction stresses for all specimens were quite low between 1 and 4 MPa. An unstable debonding was also observed for Duralin-Epoxy specimens.

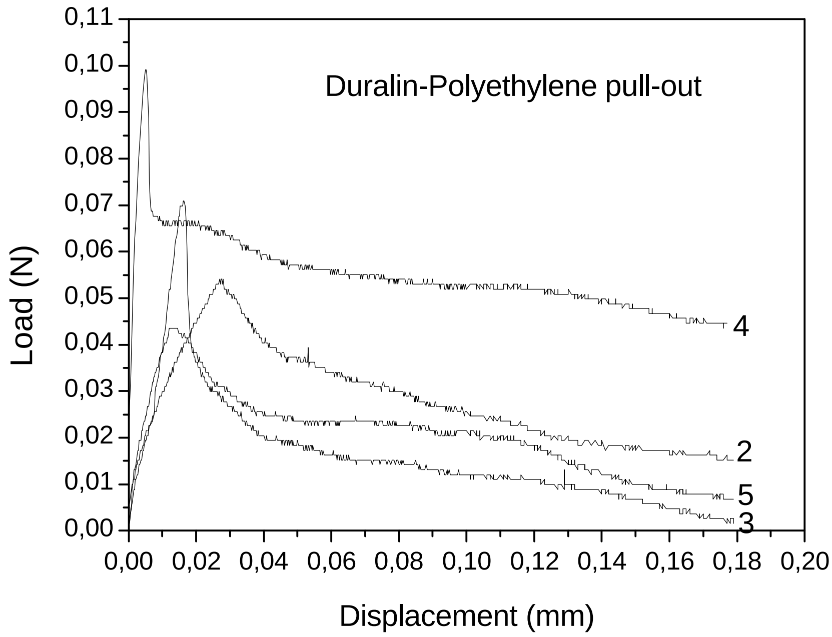

Figure 2: Load-Displacement curve for Duralin-Polyethylene single fibre composite

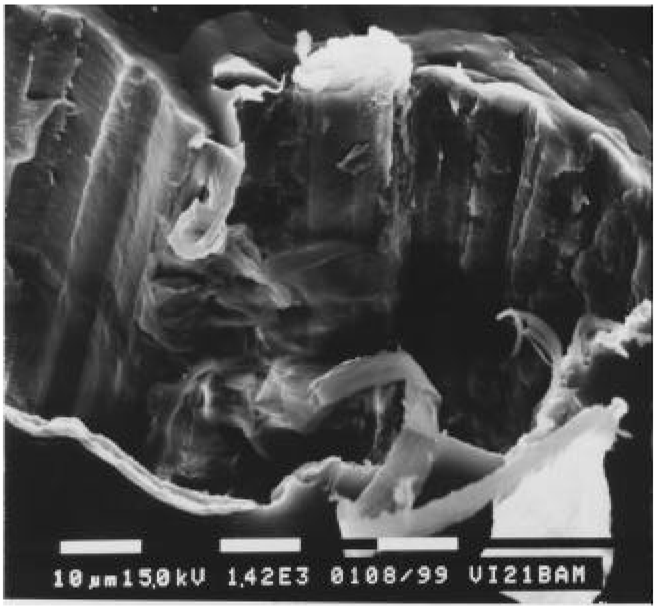

Figure 3: SEM Micrograph of Poyethylene matrix after pull-out . The Green fibre remained within the matrix.

SEM studies of the pulled out fibre surfaces indicated rougher surfaces in the case of Green-polyester than Duralin-polyester specimens. Generally, when the morphology of the fibre is smooth, the frictional force during the pull-out is small and the pull-out occurs in a gradually descending curve rather than an irregular curve in the case of a rougher fibre morphology.

Figure 4: SEM micrograph of Polyethylene matrix.The Duralin fibre has been pulled out

In the case of Duralin fibres SEM studies showed that a layer of hemicellulose-lignin matrix surrounded the fibre after the pull-out. There was no sign of rupture of the fibres or debonding in the interface between the cell fibres. It is possible that lignin from the bulk migrated to the outside of the cell wall increasing the amount of bonding medium in this region. The embedded lengths as measured from the pulled out fibres were longer than the lengths of the resin sockets from which they were pulled out, suggesting permanent elongation of the fibres.

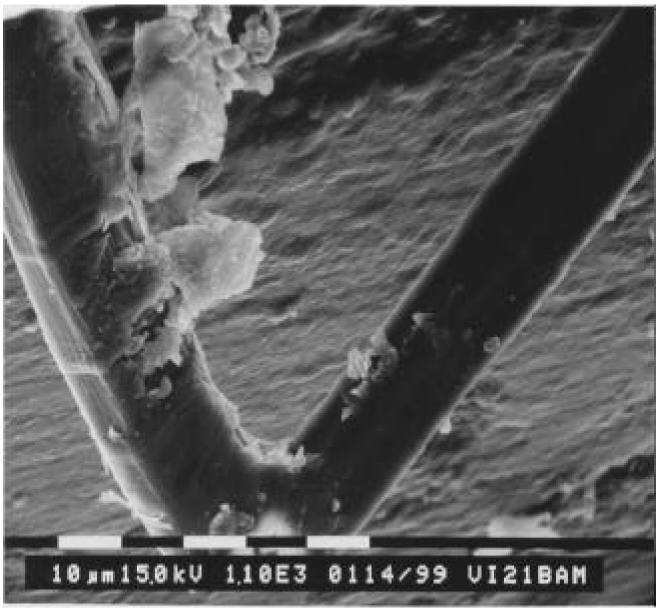

Figure 5: Green flax fibre pulled out from a Polyethylene matrix. The free length is on the left side and the embedded length on the right side.

This elongation could be explained by the uncoiling of cellulose microfibrils on the wall of the cell under tensile stress [9]. Figure 4 shows the SEM micrograph of a polyethylene matrix after the Duralin fibre has been pulled-out. Microfibrils were separated from the single fibre during pull-out and remained within the matrix suggesting that the interfaces within the fibre are weaker than the interface between the outer hemicellulose-lignin layer of the fibre and the matrix. Similar observation can be made (Fig. 5) from the micrograph of a Green fibre pulled-out from a polyethylene matrix. The outer hemicellulose-lignin layer, which surrounds the cellulose single flax fibre can be seen on the left side of the micrograph. This part of fibre is the free length during the pull-out testing. On the other hand the embedded fibre can be seen on the right side of the micrograph. It is obvious that the fibre is smoother and free from the outer hemicellulose-lignin layer compared to the free length part of the fibre. Studying the polymer matrix under the SEM it was obvious that the outer hemicellulose-lignin layer remained around and inside the hole of the polyethylene matrix.

Figure 6: Duralin fibre after being pulled-out from a polyethylene matrix

Figure 6 shows a single Duralin fibre after pull-out. The shape at the end of the fibre occurred possibly during separating and cutting the fibre in an appropriate length during sample preparation, giving reason to enhanced mechanical interlocking between the fibre and the polymer matrix. Figure 7 shows the polyethylene matrix after the pull-out of a single Duralin fibre. A small miniscus around the fibre is observed. Also, traces of the outer hemicellulose-lignin layer of the fibre show again that internal fibre interfaces break easier during the pull-out tests than the fibre-matrix interface itself. According to the above results it is suggested that more extended study of fracture surfaces by SEM and surface analytical methods is essential to generate information for the enhancement of behaviour of natural fibres as reinforcements in polymer composites.

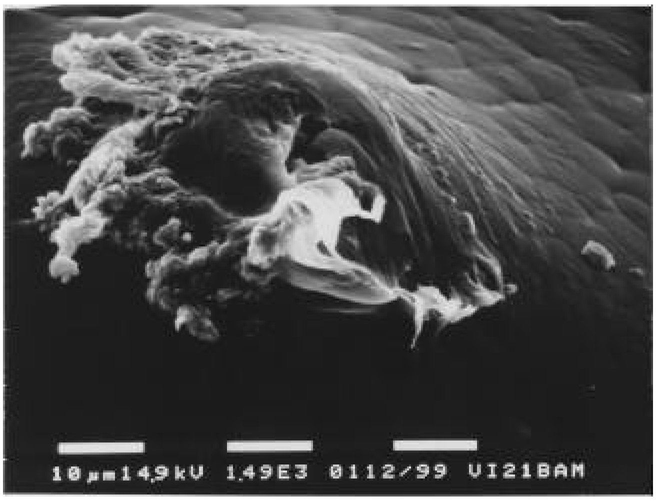

Figure 7: SEM micrograph of a Polyethylene matrix after the pull-out test of a single Duralin fibre.

CONCLUSIONS

Pull-out tests of two types of single flax fibres: Green and treated Duralin embedded in different polymer matrices: unsaturated polyester, epoxy resin and low density polyethylene were carried out. Results showed that single fibre polyester and polyethylene composites with Green flax fibres exhibited higher interfacial shear strength compared to the composites with Duralin flax. On the other hand the Duralin-epoxy resin single fibre composites exhibited the highest interfacial shear strength. For all composites the friction stress was considerably low. The pull-out tests of all the composites with polyethylene suggested that the interface within the fibres is weaker than the interface between the fibres and the matrix itself. More extended study of fracture surfaces by SEM and other surface analytical methods would generate important information for the enhancement of behaviour of natural fibres as reinforcement materials.

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