FIMATEST – A NEW TESTING SYSTEM TO DETERMINE THE FIBRE-MATRIX ADHESION STRENGTH BY MEANS OF PULL-OUT TESTS

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ABSTRACT

Textechno, Germany, the world market leader in testing instruments for man-made fibers and yarns, has a focus on the development of instruments for the characterization of the physical properties of reinforcement fibers, rovings and the fiber-matrix adhesion.

We discuss in detail the measurement of the fiber-matrix adhesion strength by means of a new testing system which has been developed together two research institutes, the Leibniz-Institut für Polymerforschung (IPF) in Dresden and the Faserinstitut Bremen (FIBRE). The system determines the adhesion between fiber and matrix in terms of the local interfacial shear strength, the interfacial toughness and further parameters through a reproducible pull-out test and has won the JEC innovation award 2016. It is suited for all kinds of fibers as well as all kind of thermoset and thermoplastic matrices with curing and melting temperatures up to 400°C.

1. CHARACTERIZATION OF THE FIBER-MATRIX ADHESION

Due to the wide range of applications, composite materials continue to gain importance. Composites consist of at least two different materials, in which the advantages of both are combined. The adhesion and failure behaviour between the different materials must be well adjusted to lead to enhanced composite performance [1]. The mechanical properties, e.g. toughness and strength, are mostly determined by the interface between reinforcement fibers and matrix material. Therefore, the influence on the fiber-matrix adhesion is of great interest, especially in the development of new fibers, their sizings as well as matrix materials.

1.1 Macromechanical vs. Micromechanical Testing

Macro- and micromechanical test methods have been employed to characterize the fiber-matrix adhesion. For macromechanical testing, composites with unidirectional aligned fibers are manufactured to perform a tensile test perpendicular to the fiber orientation (transverse tensile test). However, the maximum stress found in macroscopic tests is not only depending on the fiber-matrix adhesion, but also by additional factors: the fiber content, orientation, length, diameter and fiber distribution homogeneity, the pore void of the test specimens, and the mechanical properties of the fiber and the matrix. To achieve repeatable results for the fiber-matrix adhesion through macromechanical tests, it is hence necessary to keep tight control of the manufacturing process of the specimen, making it difficult to compare the results across laboratories.

In the case of micromechanical testing techniques, a fiber-matrix compound is usually created using just a single reinforcement fiber. Compared to the micromechanical testing, the additional factors listed above that result from the composite processing are excluded. Suitable analysis techniques allow to receive a direct measure of the adhesion strength alone.

1.2 Micromechanical Testing Techniques

Various micromechanical testing techniques (Figure) to characterize the fiber-matrix adhesion strength are discussed in the literature. An overview of the different approaches is given in Ref. [2].

Among those techniques, the microbond and the pull-out test are probably most widely used [3] due to their experimental simplicity, well-defined test geometry and high reproducibility of experimental results. The fiber-matrix adhesion can relatively easily be characterized by these testing methods. Beyond the fiber-matrix adhesion strength, the initially measured data is influenced by the diameter and mechanical properties of the fiber and by the mechanical properties of the matrix. The mechanical properties of the matrix as well as of the fiber are determined separately and their influence can be removed from the data as for instance demonstrated in Refs. [4] and [5] for the pull-out test.

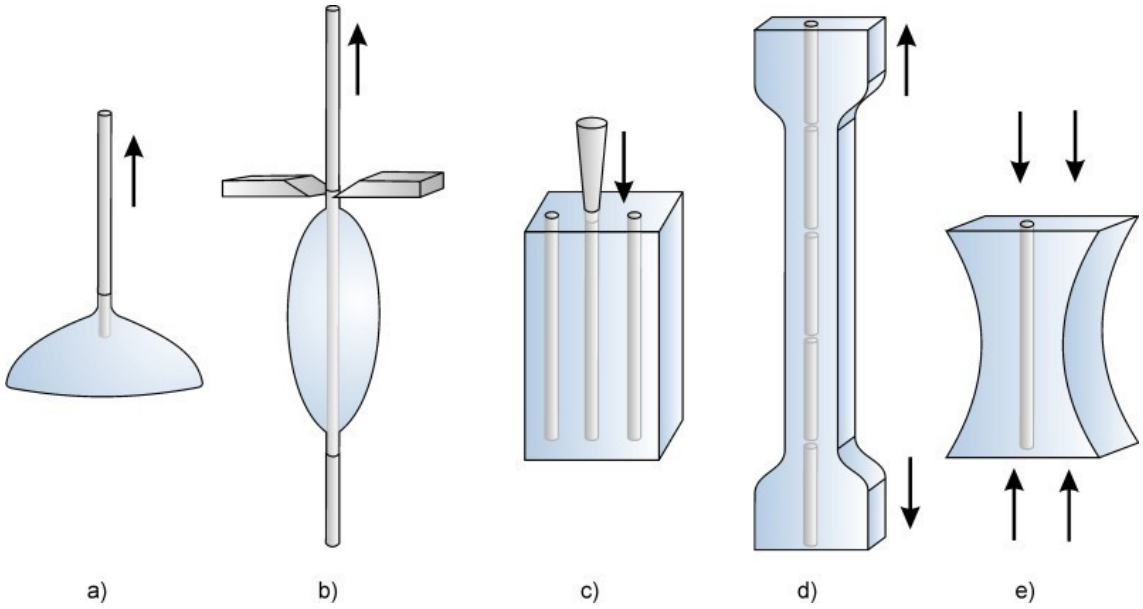

Figure 1. Micromechanical tests can be divided into two groups: 1) tests where an external load is applied directly to the fiber: pull-out (a), microbond (b) and push-out (c); and 2) tests where the load is applied to the matrix: fragmentation test (d) and Broutman test (e) [6].

Finally, the pull-out test is distinguished from the other micromechanical tests in the advantageous fact that it is applicable to most soft and stiff fibers in combination with ductile and brittle matrices that can be both thermoplastic and thermoset.

1.3 Pull-Out Test

When utilizing the pull-out test, this micromechanical technique can determine the interfacial interaction between fibers and matrices. Many test procedures to prepare the samples and perform the pull-out test have been developed by research institutes. However, since those are not standardized, the measured values are hard to compare [6]. For the following discussion, we employ the FIMATEST system [7] developed by Textechno together with the Leibniz-Institut für Polymerforschung (IPF) in Dresden and the Faserinstitut Bremen (FIBRE). FIMATEST is the first commercially available system to characterize the fiber-matrix adhesion strength by means of a pull-out test.

1.3.1 Sample Preparation for the Pull-Out Test

Figure shows the FIMABOND device of the FIMATEST system that is used to prepare the single fiber composite samples. This is a partially automated embedding station, suitable for all kind of matrices such as thermoset, thermoplastic and mineral matrices as well as all types of fibers.

Figure 2. The FIMABOND embedding station of the FIMATEST system.

The embedding of the fiber into the matrix is the most critical point to gain reproducible results through the pull-out test. The fiber must be embedded exactly in the center of the matrix droplet perpendicular to the surface of the matrix to avoid additional undesired shear forces during the measurement. Figure demonstrates an embedding process as performed with the FIMABOND. First, the fiber is approached to the top of the matrix (Figure , left) until contact is made. For typical fiber-epoxy systems a concave meniscus forms around the fiber (Figure , center). The fiber is embedded to predefined length into the matrix where typical embedding lengths range up to a few hundred microns. The embedding length is determined by the force that is necessary to fully debond the fiber from the matrix which should not exceed the tensile strength of the fiber, leading to fiber failure. Finally, the matrix is cured respectively consolidated (Figure , right).

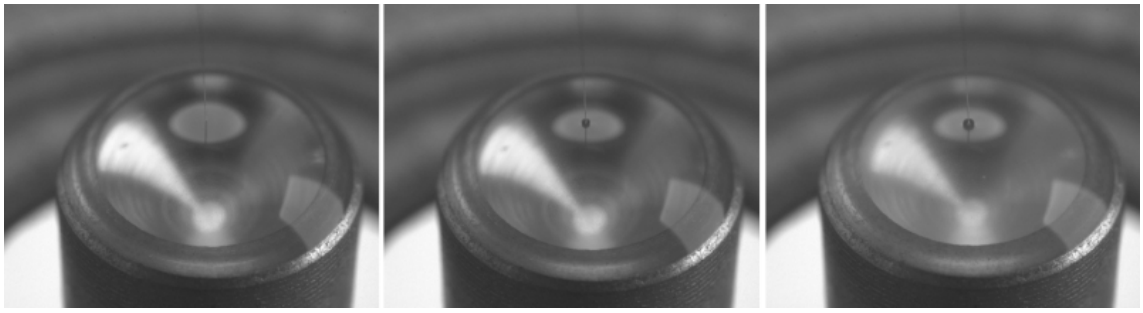

Figure 3. Process of embedding a glass fiber into epoxy resin with the FIMABOND, left: fiber approaching the matrix, center: contact between the fiber and the matrix surface and embedding, right: curing of the epoxy resin.

To process thermoplastics, the sample chamber of the FIMABOND can be flushed with inert gas, e.g. argon or nitrogen (Figure). After preparing the specimens, the sample is ready to pull out the fiber and to record the applied forces as a function of the displacement.

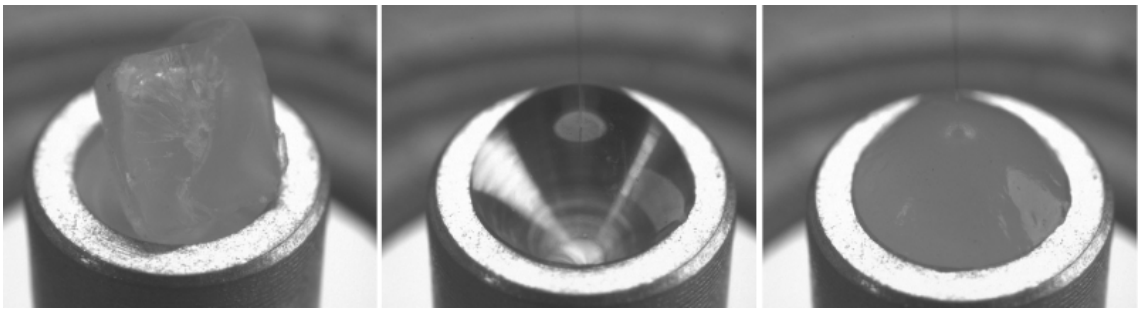

Figure 4. Process of embedding a glass fiber into PA 6 with the FIMABOND, left: flushing sample with inert gas, center: melting of the matrix material and contact between the fiber and the matrix surface and embedding, right: cooling and consolidation of the sample

1.3.2 Performing the Pull-Out Test

Textechno's single fiber linear-density and tensile tester FAVIMAT+ [10] is used to perform the pull-out test (Figure 5, left). The FAVIMAT+ is equipped by a high-resolution load cell (1 µN at 200 cN full range) as well as a highly precise and sturdy mechanics (cf. Sec. 1). The cross section of the embedded fiber must be known to fully evaluate the measured data. The cross section is determined – before embedding and pull-out test – by the FAVIMAT+ as well.

A special clamping mechanism (Figure 5, right) is installed in FAVIMAT+ to hold the single fiber composite samples that have been prepared with the FIMABOND.

Figure 5. Left: The single fiber linear-density and tensile tester FAVIMAT+. Right: The clamping mechanism installed in FAVIMAT+ to perform the pull-out test.

For the pull-out test, the prepared single fiber composite sample is put upside-down in the direct clamping system of the pull-out device. To ensure a precise alignment of the fiber to the jaw faces

and the matrix surface, a microscopic camera is integrated in the pull-out device. With the help of the camera, the fiber is adjusted parallel to the clamps and with minimal distance between the jaws and the matrix (Figure).

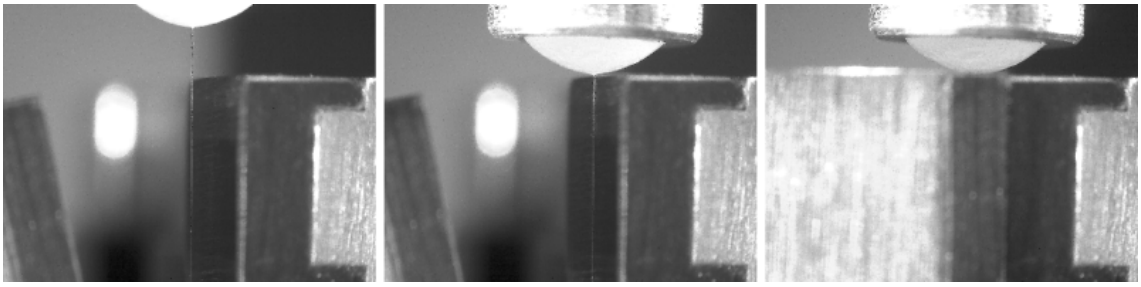

Figure 6. Process of adjusting the sample to the clamping system.

Then the pull-out test is started, the force-displacement curve is recorded, and evaluated automatically by the installed software.

1.3.3 Evaluation of the Pull-Out Test

Figure shows a typical force displacement curve recorded by the FIMATEST system. The forces are applied to the fiber. Due to the crack initiation at the interface at a critical value, the first marked force F_d (debonding force) is of special interest. From this point on the force continues to increase with a smaller slope due to the additional frictional forces occurring between the debonded part of the fiber and the matrix. Here, the crack still propagates along the interface. At the maximum force F_{max} the fiber is completely debonded from the matrix and the force drops rapidly to the remaining frictional force F_b followed by instable crack propagation. Since the embedded fiber is now fully debonded, the remaining force is only the friction between the fiber and the matrix. The actual embedding depth l_e is reached, when the applied force drops to zero – the fiber has been completely pulled out of the matrix.

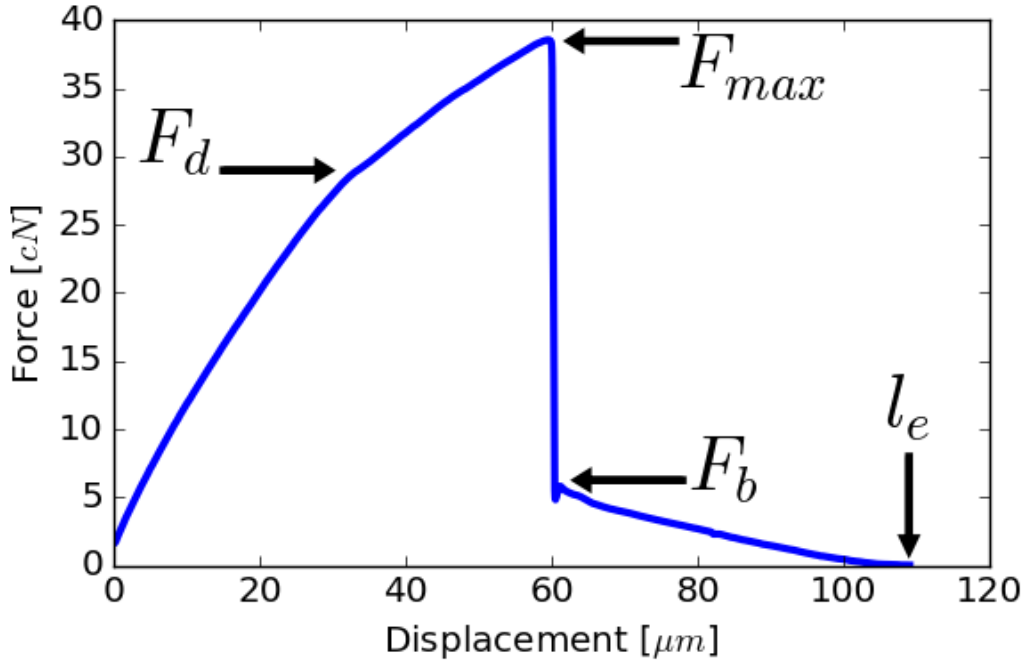

Figure 7. Force-displacement curve of glass fiber and epoxy resin.

After recording the force-displacement curve a next step is to evaluate it according to the scheme shown in Figure , yielding the maximum (F_{max}), minimum (F_b) and debonding (F_d) force as well as the actual embedding length (l_e). Based on these measured values and the individual fiber diameters d_f, the following data reduction is used according to different methods that are extensively discussed in literature [4], [5], [8]. This is done automatically by the FIMATEST software.

The apparent interfacial shear strength (τ_{app}) is the maximum force normalized on the wetted area of the fiber (Eq. 1). It is sufficient for a qualitative estimation, if different types of fiber-matrix adhesion strength are to be compared:

$$\boldsymbol{\tau_{app}} = \frac{\mathbf{F_{max}}}{\boldsymbol{\pi \cdot d_f \cdot l_e}} \quad , \tag{1}$$

where $\boldsymbol{d_f}$ ($\boldsymbol{r_f}$) is the individual fiber diameter (radius) measured by FAVIMAT+. Based on the strain-based method, the local interfacial shear strength ($\boldsymbol{\tau_d}$) is calculated (Eq. 2). Compared to the apparent interfacial shear strength, this excludes the influence of the friction between the fiber and the matrix and considers the specific geometry of the sample:

$$\boldsymbol{\tau_d} = \frac{\mathbf{F_d \cdot \beta}}{\mathbf{2 \cdot \pi \cdot r_f}} \cdot \mathbf{coth}(\boldsymbol{\beta \cdot l_e}) + \boldsymbol{\tau_T} \cdot \mathbf{tanh}\left(\frac{\boldsymbol{\beta \cdot l_e}}{\mathbf{2}}\right) \quad , \tag{2}$$

with $\boldsymbol{\beta}$, the shear lag parameter as defined in Ref. [9] and $\boldsymbol{\tau_T}$, considering thermal stresses. The critical interfacial energy release rate ($\boldsymbol{G_{ic}}$) – based on the energy-based method – allows to

consider the bonding on its own as well. The energy release rate is calculated as a function of the crack length and takes the deformation of fiber and matrix during the pull-out into account (Eq. 3):

$$G_{ic} = \frac{r_f}{2}\left\{C_{33s}\cdot\overline{\sigma}^2 + 2\cdot D_{3s}\cdot\overline{\sigma}\cdot\Delta T + \left(\frac{{D_3}^2}{C_{33}} + \frac{V_m\cdot(\alpha_T-\alpha_m)^2}{V_f\cdot A_0}\right)\cdot\Delta T^2 + \left[\frac{\sigma_0}{2}\cdot\left(\frac{1}{E_A}-\frac{1}{E_m}\right) + D_{3s}\cdot\Delta T\right]\cdot\left[\left(\overline{\sigma} + \frac{(1+m)\cdot D_3\cdot\Delta T}{C_{33}}\right)\cdot C'_T(a) - k\cdot C_T(a)\right]\right\} \quad . \tag{3}$$

As seen in the equations (2) and (3), parameters like the modulus E, the poisson ratio and the coefficient of thermal expansion α of the fiber and the matrix as well as the difference between reference stress-free temperature and the testing temperature ΔT are considered parameters in the evaluation of the local interfacial shear strength and the critical energy release rate. A detailed description of the parameters in given in Refs. [4] and [5].

After the debonding of the fiber from the matrix, the fiber will be completely pulled out. Thereby only friction, expressed by the interfacial frictional stress (τ_f), will occur (Eq. 4).

$$\tau_f = \frac{F_b}{\pi\cdot d_f\cdot l_e} \quad . \tag{4}$$

1.3.4 Selected Data for Different Fiber-Matrix Combinations

Using the FIMATEST system described in the section above, we determine the local interfacial shear strength τ_d for several fiber-matrix combinations. Figure shows the results for standard and treated fibers of both carbon (CF) and glass (GF) embedded in different matrices – polyetheretherketone (PEEK), polyamide (PA) and an epoxy resin (Epoxy). Moreover, a glass fiber embedded in different modified polypropylene matrices is shown. For these examples, the kind of fiber treatment or modification to the matrix or fiber is not specified in detail.

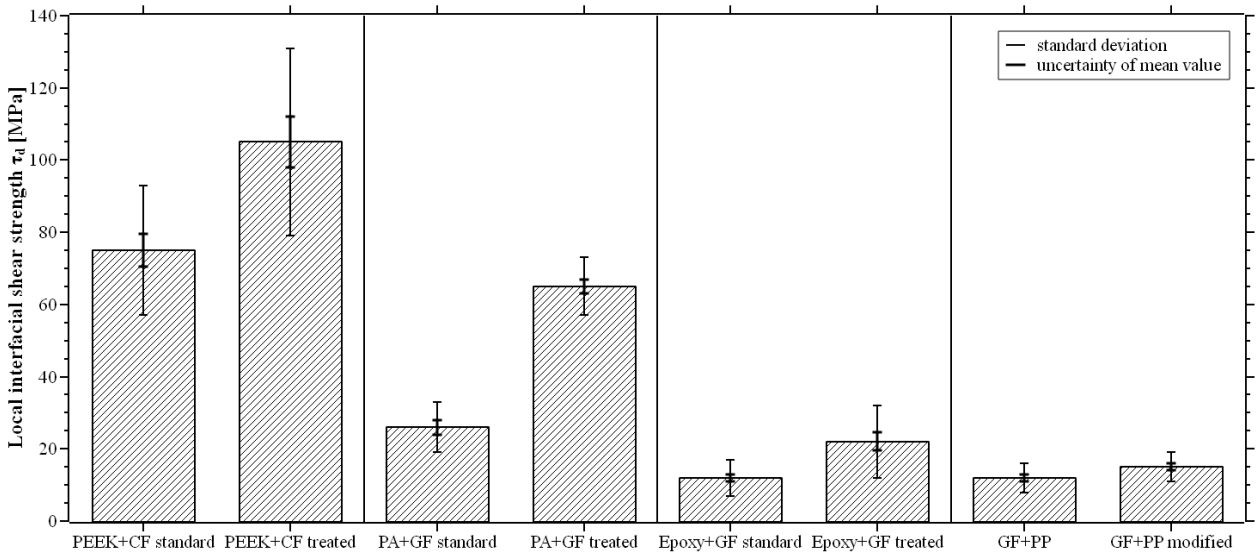

Figure 8. Local interfacial shear strength τ_d of standard and treated fibers in different matrices

We note that in all cases a significant improvement of the adhesion strength by treating the fibers or modifying the matrix could be detected. Figure displays the local interfacial shear strength, whereas Figur shows the critical energy release rate for the same set of fiber-matrix combinations.

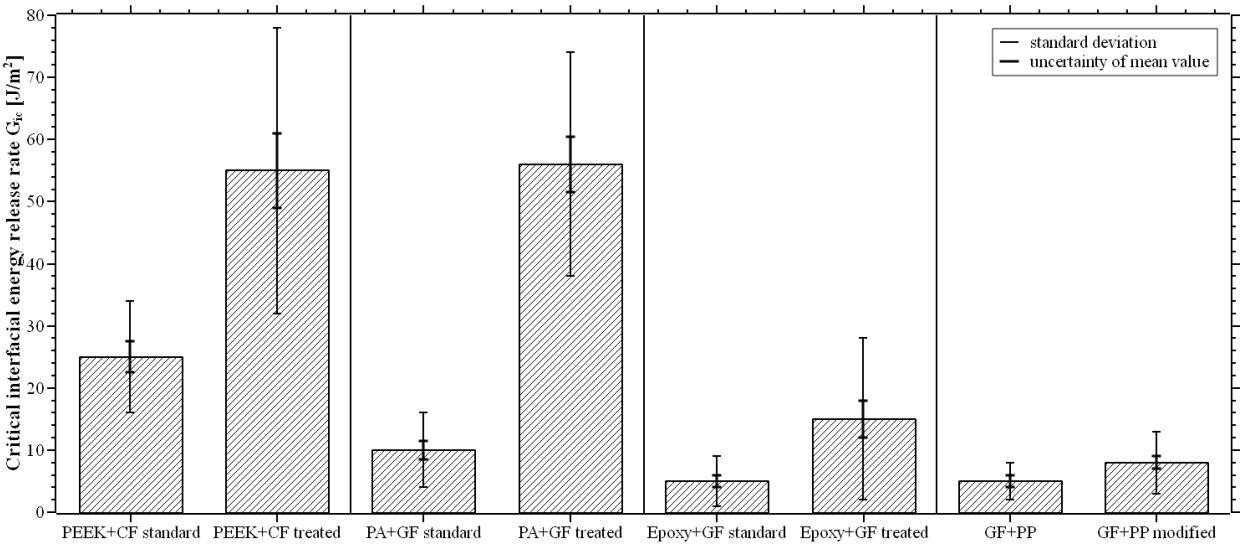

Figure 9. Critical interfacial energy release rate G_{ic} of standard and treated fibers in different matrices

For some fiber-matrix combinations the differences in adhesion strength become more clearly visible in the energy based model (Figure 9) than by the evaluation through the strain-based model (Figure 8). Again, it is noted that the adhesion strength increases significantly by the treatment of the fiber. Also, the use of different modified PPs leads to a significant difference in the results.

Another example for a glass fiber embedded in PP is given in Fig. 10. This study has been performed together with PHP Fibers GmbH with a focus on the amount of maleic anhydride masterbatch that is added to the PP fibers in hybrid tapes to optimize the adhesion to glass fibres in the consolidated composite. Starting without any maleic anhydride, both the strain-based (Fig. 10, left) and the energy based (Fig. 10, right) evaluation of the pull-out test show that the highest fiber-matrix adhesion strength is reached at 6 wt% fraction masterbatch in the PP. The data indicates, that at 10 wt% masterbatch the optimum amount of maleic anhydride has been exceeded.

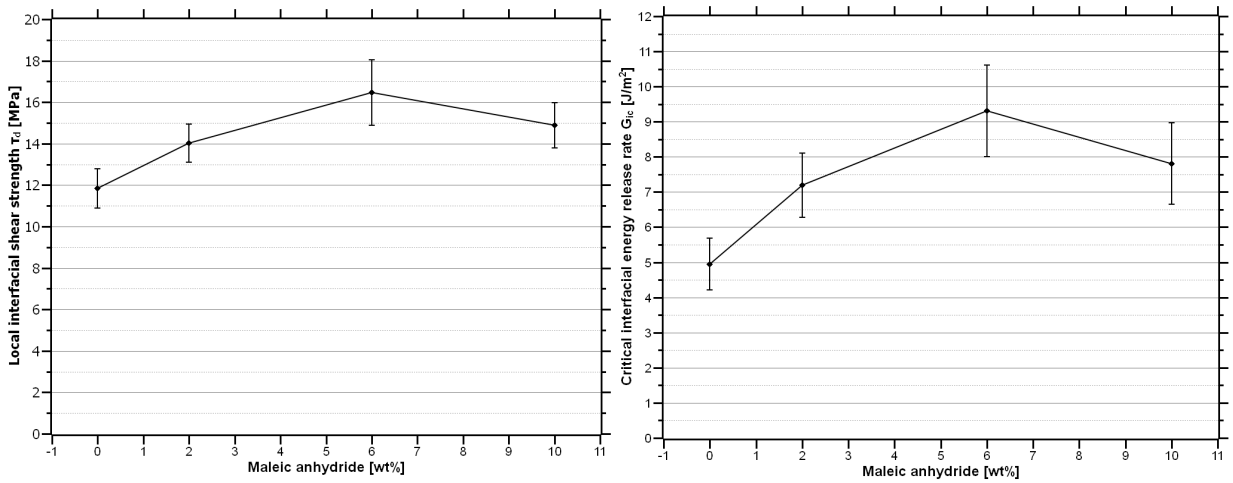

Figure 10. Fibre-matrix adhesion strength between GF and PP as a function of wt% maleic anhydride masterbatch in the PP. The error bars indicate the uncertainty of the mean value using 15 trials. Left: strain-based evaluation applied to the pull-out data, right: the energy based evaluation is used. In both cases the maximum adhesion is found at 6 wt% masterbatch.

2. CONCLUSION

In Textechno's FIMATEST system the micromechanical pull-out test technique to characterize the fiber-matrix adhesion strength is combined with a highly repeatable and reliable sample preparation using the all-new FIMABOND device. In this way, it is possible to characterize the fiber-matrix adhesion strength in terms of the local and the apparent interfacial shear strength as well as the critical interfacial energy release rate (interfacial toughness). Differences in the fiber-matrix adhesion are usually due to the treatment of the fiber (e.g. sizing, plasma treatment) or the modification of the matrix material. With this advanced equipment, differences in the fiber-matrix adhesion can be traced on all kinds of fiber materials in combination with all thermoplastic, thermoset and cement matrices.

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