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ON THE INTERPLY FRICTION OF DIFFERENT GENERATIONS OF UNIDIRECTIONAL PREPREG MATERIALS

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ABSTRACT
With the aim of reducing cost of prepreg composite components, manufacturing methods are developed and refined. An automatic tape laying machine can shorten the process cycle by stacking prepreg flat and thereafter allow for forming into desired shapes. Forming of stacked prepreg requires knowledge about the uncured properties of prepreg, such as viscosity of the matrix, intra- and interply deformation properties. This study focus on the interply friction, i.e. the friction at the prepreg-prepreg interface, and how this affects the forming. The conclusions presented here show that the difference between prepreg material systems is significant. Further, it is concluded that the prepreg-prepreg friction is governed by a combination of Coulomb and hydrodynamic friction, where different mechanical phenomena dominate depending on the test conditions.

1 INTRODUCTION
With the increasing use of composite materials in civil aeronautical applications follow a demand for higher efficiency and reduced process cycle times compared to the more traditional manual processes developed for small series aeroplanes. Using prepreg materials, the focus should be on the lay-up process in order to avoid tedious manual steps and inter-mediate debulking. The automatic tape laying (ATL) machine opens up for efficient lay-up, however is not feasible for too complex component geometries. The pre-stacked sheet must therefore often be formed prior to (or as a first step in) the curing process.

Forming a stack of prepreg requires that the material deforms according to the desired shape in a predictable way without causing wrinkles or other faults. This is done by forcing the different layers to deform by intraply deformation (within prepreg ply) and interply deformation (in-between prepreg plies); both in the plane and out of the plane. This paper focuses on the interply deformation, or more exactly the resistance to interply deformation, by measuring the friction in-between separate prepreg plies. This friction influences both the non-homogenous deformation in the plane during a pin-jointed-net deformation behaviour [1] and the required sliding in-between separate prepreg plies during out of the plane deformation.

The prepreg-prepreg friction has previously been reported to influence the quality of the formed composite component; the friction coefficient between layers can cause inbuilt residual stresses [2], core crushing during autoclave moulding of honeycomb sandwich components [3] and wrinkles due to resistance of forming. Considering the latter, previous studies have shown large differences in the in-plane draping behaviour between different generations of prepreg materials [4-7]. In particular, it seems like the thermoplastic, microscopic particles used as tougheners (craze stoppers) greatly influence the degree of sliding within and between prepreg layers during forming.
Existing models describing interply friction often considers a combination of Coulomb (friction between dry surfaces) and hydrodynamics (friction between two layers completely separated by a thin layer of fluid). Further, the hydrodynamic models often include rheological models predicting the relationship between shear rate and fluid viscosity. Process parameters as sliding rate (i.e. matrix shear rate) and matrix temperature has experimentally been shown to significantly influence the interply friction coefficient [2]. Martin et al. [3] suggest that the interlaminar friction between woven prepregs is dependent on both the distribution of surface resin and resin viscosity. However, measurements on the interply friction coefficient between two layers of thermoset prepreg has also revealed [2] that as the temperature increases, the influence from Coulomb friction increases, indicating increased mechanical interaction between the reinforcing layers.

The Strubeck theory [8], initially developed to explain various types of friction in relation to the sliding rate, lubricant viscosity and bearing pressure in tribology, represents another approach to describe the prepreg-prepreg interply friction [9]. The Strubeck curve shows the Coulomb friction coefficient as function of the Hersey number given as:

\[
H = \frac{\eta \nu}{p}
\]  

where \( \eta \) is the (dynamic) viscosity of the matrix, \( \nu \) is the velocity at the contact surface and \( p \) is the applied pressure. The example curve in Figure 1 illustrates three different areas according to the type of friction taking place. The first region is governed by boundary lubrication similar to the Coulomb friction. The second part is a mixed mode friction, which gradually translates into the third hydrodynamic region with increasing thickness of the lubrication layer. The theory predicts that in the hydrodynamic region, the friction coefficient increases with thickness of lubricating layer as more liquid needs to undergo shearing for the thicker liquid layers. Please note that in Figure 1, the normal force is used to calculate the Hersey number instead of the pressure, as in Equation 1, wherefore the dimension differ. The feasibility of the Strubeck curve is that it enables a translation of the different tribological phenomena into one phenomenological model easily implemented into e.g. FE software.
The aim of this study is to measure and evaluate the prepreg-prepreg interply friction for two different generations of aeronautical carbon/epoxy prepreg systems: one including particle tougheners in the matrix and the other more traditional system without particles. Earlier studies have shown [6, 7] that these tougheners provide flow-control significantly influencing the in-plane deformation and it is therefore expected to find large differences also in the friction coefficient. Based on the theory presented above the following parameters falls out as important for the interply prepreg friction: contact pressure, matrix viscosity, thickness of matrix film on the surface and fibre roughness. The aim is to (directly or indirectly) investigate these parameters to find the governing friction mechanisms and if possible develop simple models based on the Stribeck curve.

2 MATERIALS

The two different prepreg materials tested were HexPly® T700/M21 (referred to as M21) from Hexel and Cycom® HTA/977-2 (referred to as 977-2) from Cytec. Both are aerospace graded carbon/epoxy systems that are widely used by the aerospace industry. The systems are toughened by thermoplastics to prevent cracks from growing; in the case with M21 the state is in form of particles and with 977-2 it is liquid. Figure 2 shows the microstructure of the different systems when consolidated and cured; where the lightest grey is the fibre, the darker grey is the matrix and the darkest grey in M21 is the thermoplastic particles. Both M21 and 977-2 have similar volume fraction of fibres (57%), but due to the particles working as distances in-between the fibre layers in the M21 material, the local fibre volume fraction in M21’s fibre layer is much higher than in 977-2. This can be a part of the fact that, although having a lower matrix viscosity, M21 is more difficult to consolidate [10].

Two different versions of the materials were tested: virgin material and pre-consolidated material. The latter material was bagged onto a metal plate and consolidated at 80ºC and 8 bar during 30 min in order to reach the cured ply thickness (CPT), i.e. the requested degree of consolidation. The purpose of this treatment is twofold; to reduce the thickness of the initially resin rich surface by reaching full impregnation and to compact (smoothen) the fibre reinforcement. Further, the consolidation is expected to influence the behaviour of the toughening particles: Prior to consolidation the particles are free to move and roll. However, after consolidation the
particles are compressed towards and into the reinforcement, partly locking them into position and consequently increasing the surface roughness of the prepreg.

In order to ensure that the material remained at CPT after consolidation and prior to testing, the consolidated material was held under vacuum until used.

![Image of microstructure of consolidated prepreg laminates](image)

Figure 2: Microstructure of consolidated prepreg laminates a) M21 (×500) b) 977-2 (×1000) (note the different scales). Courtesy of Per Hallander, Saab AB, Linköping, Sweden.

### 3 EXPERIMENTAL

To measure the interlaminar friction between prepreg plies a test rig was developed. The apparatus is shown in Figure 3 and consists of one pneumatic cylinder and three plates holding the prepreg: one large (150 x 200 mm²) fixed to the lower beam of the Instron 4505 testing machine and two smaller plates (100 x 90 mm²) moving upwards as the test starts. To obtain isothermal conditions at elevated temperatures the apparatus can be mounted inside a heating chamber, specially designed to fit the Instron 4505.

To start a test, sufficiently large pieces of prepreg are fixed onto the three plates using silicon rubber bags and vacuum (-0.9 bar and 5 min). The outer cover film is kept on in order to secure a clean surface. The plates are thereafter mounted onto the test rig, the cover film removed, the oven closed and heating initiated. When the set temperature is reached, the rig is closed activating the pneumatic cylinder by increasing the air pressure to a level providing the desired normal force. The test is started 2 minutes after activation of the cylinder.

The load and displacement is measured using the Instron load cell and machine position, respectively. The friction coefficient, \( \mu \), is thereafter calculated from:

\[
\mu = \frac{F}{N}
\]  

where \( F \) is the measured maximum load before slippage and \( N \) is the normal force applied to the test area.
In this first series of tests the aim was to see the influence of temperature, deformation rate and pressure. All test configurations can be seen in Table 1. The different temperatures were selected based on initial experiments at SAAB Aerostructures and with the aim to test both materials at temperatures where their matrix viscosities were similar. However, it should be noted that the matrix viscosity given includes the toughening particles (for M21). Since the volume fraction of particles at the prepreg surface is approximately 35%, it is expected that the actual matrix viscosity (without particles) is approximately one magnitude lower [11]. The cross-head rate was selected based on realistic forming rates during sheet forming (vacuum forming) of pre-stacked prepreg material. The vacuum during this process set the maximum normal pressure used in the tests.

Table 1: Test matrix.

<table>
<thead>
<tr>
<th>Material</th>
<th>Temperature [°C]</th>
<th>Pressure [MPa]</th>
<th>Cross-head rate [mm/min]</th>
<th>Viscosity [Pa·s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>977-2</td>
<td>70</td>
<td>0.51/0.76/1.2</td>
<td>0.1</td>
<td>1000</td>
</tr>
<tr>
<td></td>
<td>70</td>
<td>0.51</td>
<td>0.05/0.1/1</td>
<td>1000</td>
</tr>
<tr>
<td></td>
<td>80</td>
<td>0.51</td>
<td>1</td>
<td>400</td>
</tr>
<tr>
<td>977-2, consolidated</td>
<td>80</td>
<td>0.76</td>
<td>0.1</td>
<td>400</td>
</tr>
<tr>
<td>M21</td>
<td>60</td>
<td>0.76</td>
<td>0.1</td>
<td>1000</td>
</tr>
<tr>
<td></td>
<td>70</td>
<td>0.76</td>
<td>0.1</td>
<td>400</td>
</tr>
<tr>
<td></td>
<td>85</td>
<td>0.51/0.76/1.2</td>
<td>0.1</td>
<td>100</td>
</tr>
<tr>
<td></td>
<td>85</td>
<td>0.76</td>
<td>0.05/0.1/1</td>
<td>100</td>
</tr>
<tr>
<td>M21, consolidated</td>
<td>70</td>
<td>0.76</td>
<td>0.1</td>
<td>400</td>
</tr>
</tbody>
</table>
RESULTS AND DISCUSSION

Figure 4 illustrates load-displacement curves for M21 and 977-2, respectively. As can be seen, for the given test conditions, the measured load (and friction coefficient) is approximately 10 times higher for M21 than for 977-2. At the considered temperatures, the given matrix viscosity (with particles) is significantly lower for M21 than for 977-2. This would, from a purely hydrodynamic perspective, result in an opposite trend. Further, the Striebeck curve shows that that in the hydrodynamic region, the friction coefficient increases with thickness of lubricating layer (i.e. thickness of matrix surface film). The virgin M21 has a less tacky and dryer surface than 977-2, indicating a thinner matrix layer. (However, for the consolidated material shown in Figure 2 the film thickness is larger.) This indicates that at least one of the friction coefficients for the tested materials are not governed by hydrodynamic friction.

As can be seen in Figure 4, the experimental data do not show a significant load peak as expected for Coulomb friction. Instead, the sliding starts at different loads depending on the test settings and the load increases rapidly up to (close to) a plateau. In fact the curve never really flattens out or drops, which seems to be reasonable considering that as the test continues, the lubricating matrix layer is worn off and the fibre roughness remains or increases with the abrasion. In discussions with the industry, the resistance to initial sliding was selected as most interesting from a processing point of view, wherefore the friction values representing each set of tested parameters in Figure 5-7 are taken at slippage initiation. It should however be noted that although at a slightly different level, the same trends can be reported when consequently choosing values closer to (or at) the “plateau”.

Figure 5 presents the influence on temperature on the prepreg-prepreg friction. The figures display that a lower viscosity (i.e. higher temperature) leads to a higher friction coefficient for both materials. However, the difference in friction coefficient is small compared to the reduction in viscosity: reducing the viscosity 10 times increases the friction coefficient with 12% (for M21). Nevertheless, the observed trend is in contradictions to predictions from pure hydrodynamic considerations. It should be
remembered that the prepreg is a soft material and using a lower matrix viscosity, all fibres are held more loosely together, also promoting fibre pop-up and increased surface roughness. Here, it seems like this effect is larger than that from reduced hydrodynamic friction. It can be noted that for M21, the difference between the friction coefficients at the two highest viscosities tested (lowest temperatures) is small. It would be interesting to compare to tests performed at room temperature in order to find if the friction coefficient is converging with respect to temperature.

Figure 5 reveals that the consolidated M21 shows an increased friction coefficient compared to the virgin material tested at the same viscosity. This is reasonable considering that the consolidation increases the surface roughness by compressing and locking the particles into the surface of the fibre reinforcement. The opposite was seen for the consolidated 977-2, which gave a significantly lower friction coefficient than the virgin material; probably due to the smoothened prepreg surface following consolidation. These results show that for the considered materials, the influence of surface roughness seem to be larger than that of all other parameters tested, especially for M21.

Figure 5: Viscosity vs. friction for M21 and 977-2, tested at 0.76 MPa and 0.1 mm/min. Note that 977-2 is multiplied by 10.

Figure 6 shows that the friction coefficient decreases with increasing pressure for both M21 and 977-2. The Coulomb friction coefficient is by definition constant independent on normal pressure, see Equation 2. If the prepreg-prepreg friction is purely hydrodynamic, the decrease in friction coefficient would be linear with increasing pressure. For the two tested materials, the decrease in friction coefficient is somewhat larger (for M21) or significantly larger (for 977-2) than for a purely hydrodynamic friction considering the two lowest pressures tested. Thereafter the friction coefficient reduces less upon further reduction in pressure. Please note that it is expected that the increased pressure also compacts the fibre reinforcement and therefore reduces the roughness of the surfaces. These two phenomena consequently add, which might be the reason for the initial dramatic reduction in friction coefficient for 977-2. Considering the large difference between the M21 friction coefficient at the highest pressure and its friction coefficient for the consolidated material (see Figure 5), it is obvious that different mechanisms are active: “rolling” particles under high pressure compared to
significantly increased surface roughness. However, for 977-2 the friction coefficient at
the highest pressure is still slightly higher than that of the consolidated material,
indicating that higher pressure is required to obtain the same reduction in surface
roughness as the consolidated material.

Figure 6: Friction vs. applied pressure, a) M21 tested at 85°C and 0.1 mm/min and b)
977-2 tested at 70°C and 0.1 mm/min.

Figure 7 shows the Striebeck curves for the M21 and 977-2, respectively. For M21 it
can be seen that the Striebeck curve does not follow a clear trend. This can be an effect of
the more complex matrix-particle-surface interaction present, with a complexity beyond
that predicted by the Striebeck curve. It can also be so that all measurements actually are
in the mixed zone (Coulomb and hydrodynamic). Please note that the difference in
investigated Hersey numbers is one magnitude smaller for M21 than for 977-2 (due to a
smaller difference in matrix viscosities at the selected temperatures). In fact, the
measured differences in friction coefficients are lower than for the 977-2 material and
for the considered Hersey numbers the friction coefficients for M21 can only be
modelled as one average value with fairly high errors. The 977-2 material on the other
hand, seems to follow the trend predicted by the Striebeck curve. However, it should be
noted that this theory does not take into account that the surface roughness changes
depending on the settings of test parameters, which has previously shown to influence
the friction coefficient.

Figure 7: Experimental Striebeck curve for a) M21 and b) 977-2.
4 CONCLUSIONS
This study measures and compares the prepreg-prepreg interply friction coefficient of two different carbon/epoxy prepreg material systems: 977-2 and M21. The experimental experience of a higher resistance to deformation for M21 compared to 977-2 is thereby confirmed: M21 requires approximately 10 times higher load than 977-2 to allow for interply slippage, despite that the matrix viscosity of M21 is significantly lower.

The results further show that at elevated temperatures, the interply prepreg-prepreg friction is composed of a combination of hydrodynamic friction and Coulomb friction and thus depending both on matrix viscosity and surface roughness. The latter is significantly influenced by the processing conditions. For pre-consolidated 977-2, the interply friction is smaller than for the virgin material due to the reduction in surface roughness. Pre-consolidated M21, on the other hand, shows increasing resistance to motion due to the fixation of the toughening thermoplastic particles onto the surface and the corresponding increase in surface roughness.

Considering the non consolidated material, the friction coefficient is not constant with increasing pressure as predicted by the Coulomb theory. Instead, the friction coefficient reduces as a result of reduced surface roughness and increasing pressure, indicating that the friction still is partly hydrodynamic in its nature.

The prepreg material is soft and the surface roughness is influenced not only by pressure, but also by increasing temperature, as the softening matrix promotes increasing roughness. The friction coefficient consequently increases with increasing temperature.

For the considered test parameters, the friction coefficient of the 977-2 can be modelled using the Stribeck curve. However, for M21 the range of investigated parameters is too small and the combination of the soft surface, fluid matrix and hard thermoplastic particles too complex to enable drawing of the Stribeck curve. Instead, for the considered range of test parameters the friction coefficient of the (non consolidated) M21 can be considered as constant independent of processing conditions if allowing for fairly large errors.

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