INFLUENCE OF BINDERS IN THE LIQUID COMPOSITE MOULDING PROCESS ON THE FATIGUE PROPERTIES OF COMPOSITE STRUCTURES

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SUMMARY: Static and fatigue tests were conducted with binder coated glass fibre fabrics to determine the influence of a binder on the material properties of a laminate. Different resin systems are used for the main matrix and the binder system in order to investigate a typical situation from the manufacturing of preforms for the RTM process. The static tests showed a negligible influence of the binder on the relevant material properties. Oscillatory torsion tests on tubes for the determination of the fatigue properties do not indicate a negative influence of the binder system used here in combination with the epoxy matrix. The influence of the binder is far less important than the effect of the fibre fraction in the laminate. A significant decrease in the fatigue strength of the fibre matrix interface is detected in the fatigue tests of flat specimens.

KEYWORDS: LCM, RTM, binders, fatigue, material properties, composite structures

INTRODUCTION

Key issues for the choice of a manufacturing method are its costs, the achievable quality including reproducibility and material strength. Recently physiological aspects became also more important. A cost comparison for different manufacturing methods for fibre composite structures is strongly dependent on the number of parts to be produced. The demands on the quality and the material strength of such a structure has also a strong influence on the costs. Reproducibility is commonly regarded as very important part of the quality. Thus, the key issues for composite manufacturing have to be evaluated in an integrated approach. This also includes the design process [1]. Due to the increasing interest in the Liquid Composite Moulding (LCM) methods they obviously offer a very good compromise between the most important requirements in composite manufacturing.

LCM methods and in particular the Resin Transfer Moulding (RTM) processes are subject of strong interest for the manufacturing of high performance composite structures in the last
years. They are already used in an industrial scope for secondary structures or primary structures with lower demands for high specific strength since the sixties. Meanwhile the LCM techniques have improved significantly and high fibre fractions can be achieved in composite structures with these more sophisticated methods. Consequently LCM methods become an interesting alternative to other manufacturing methods like wet lay-ups or the pre-preg and autoclave route. In fact, the process is about to make its way in the aerospace industry to complement or to replace the common pre-preg autoclave route [2].

A significant amount of labour time is spent during the dry lay-up prior to the resin injection in LCM. Thus, saving labour time in this part of the process may be considered as being one of the most efficient approaches to save costs. Frequently pre-formed fibre reinforcements are used to facilitate the dry lay-up [3]. In order to produce pre-forms, one way is to apply epoxy powder on the fabrics. The epoxy powder here is commonly referred to as binder. However, if the binder powder is made of the same epoxy basis as the latter used injection resin no problems are to be expected for the material strength as the epoxy binder usually fuses with the injection resin during a RTM process at increased temperatures.

It is advantageous to have one epoxy system as binder and injection system [4]. Due to various reasons – usually costs - this is not possible for many applications and two different systems are used in a component. Consequently, there is a vital interest to investigate the influence of binders on the material properties if different systems are in use. That has been done previously for some representative static material properties for different binders and tackifier agents [5].

In this work a glass fibre reinforced plastics (GFRP) material incorporating a binder resin system different to the injection system is investigated. Apart from static material properties the fatigue strength is of particular interest. The static tests are conducted according to common test standards. For the determination of the fatigue strength tubes are tested in torsion with the fibres aligned in the main load directions. A second test method is introduced to determine the fatigue strength of the compound with emphasis on the interface between fibres and matrix. In this test method flat specimens are loaded under ±45° to the main fibre directions. A comparison of the results of laminates manufactured of fabrics with and without a binder is presented. The influence of the manufacturing method on the material properties is discussed.

MATERIAL TESTS

Prior to fatigue tests static material tests are carried out to detect effects on the static material strength due to the incorporation of a typical epoxy based binder on a glass fibre fabric. Different epoxy resin systems are chosen for binding of the fabric and for the use as matrix material. Thus, a typical problem evolved from the use of different resin systems as described in the introduction is investigated.

Despite the title of this paper most of the test specimens are manufactured by using hand lay-up methods. Only the tubular test specimens for the fatigue tests are manufactured using a vacuum infusion method. However, the aim of this work is the investigation of the influence of binders on the composite’s material properties. Binders are commonly used for LCM methods only. Because the specimens which are compared are manufactured in the same way no influences of the manufacturing method are to be expected. The differences in the results originate from the binder only.
Commonly the binders are melted in order to manufacture a preform. During the manufacturing of the flat fatigue test specimens the binder is melted prior to the application of the main matrix system. The tubular test specimens are manufactured without melting the binder. This way both main features of binder treated fabrics - the preform capability and the easier handling - are investigated separately.

**Static tests**

The tests are conducted according to different standards. Interlaminar shear strength is determined according to DIN EN 2377. Bending strength is measured according to DIN EN 2746 and the tensile tests are carried out using DIN EN 2747. Compression strength is measured with the Celanese test method DIN EN 2850.

**Fatigue tests**

There are no common fatigue test standards to determine the fatigue strength of a fibre composite specimen or component. Therefor, the fatigue tests carried out within the scope of this research project are done on the basis of a certification standard of the German aviation authority Luftfahrtbundesamt (LBA) for sailplanes and powered sailplanes [6]. There are two different test methods described in this certification standard. A tubular specimen which is loaded under torsion and a flat test specimen which is tested in tension under ±45° to the main fibre directions. Tests are carried out with both test methods.

*Tubular specimen*

The geometry of the tubular test specimens can be seen in Fig. 1. Its lay-up consists of four layers of glass fibre fabric and additional layers at the ends for the load introduction. In the test area the wall thickness is about 1mm. For a pure torsion load an investigation of the stresses in the different components -fibres and matrix- using utilisation factors according to [7] shows that the matrix is substantially higher used than the fibres. However, for the investigation of the influence of a binder on the fatigue properties of a fibre composite component it is more interesting to see a matrix or interface failure as the binder is not expected to decrease fibre properties.

The oscillatory torsion tests are carried out using a test frequency of 5.6 Hz. The upper load level is 10 times higher than the lower load level and the oscillating moment is always positive in the same direction (R=0.1).

![Fig. 1: Geometry of the tubular test specimen for fatigue tests (dimensions in mm).](image-url)
Flat specimen

The flat test specimen is a rectangular strip 210mm long, 32mm wide and 2mm thick with a test length of 130mm. As the specimens comprise of a symmetric stacking sequence with all fibres under $\pm 45^\circ$ the failure mode is again dominated by the matrix properties. This test method is very simple and it also provides information about the fibre-matrix interface very quickly.

Materials

The glass fibre fabric which is investigated is manufactured by CS-Interglas. This style 92125 fabric has a twill 2/2 weave pattern and the area weight without binder is 280 g/m². The binder is applied single sided on the fabric with a weight fraction of 4.5 to 5%. It is produced by Wacker Polymer Systems under the trade name VINNEX®.

Static tests are carried out with an unsaturated polyester resin (UP)(Alpolit UP-303/Interox 3P50), epoxy resin (EP) (Bakelite L20/SL), a vinyl ester resin (VE)(Derakane 411-45/LPT) and a polyamide film. Test specimens for the fatigue tests are manufactured with an epoxy resin system only (MGS L285/H286). However, this investigation concentrates on epoxy resin systems. The static results of the other matrix systems show that there is no significant decrease in material properties due to the use of the VINNEX® binder. For the static test specimens and the flat fatigue specimens the binder is melted before the main matrix system is applied on the fabric. No heat treatment of the binder was done prior to the infusion of the main matrix system of the tubular fatigue specimens.

TEST RESULTS

Static tests

A survey of the results of the different static tests is given in Table 1. The tested fabric is available with two different finish for the combination with epoxy resins. Finish FK144 is still on stock but will be replaced entirely by finish FK800 due to environmental reasons in the near future.

<table>
<thead>
<tr>
<th>Interglas 92125</th>
<th>FK800 dry/wet</th>
<th>FK800 B dry/wet</th>
<th>FK144 dry/wet</th>
<th>FK144 B dry/wet</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bend. Strength [N/mm²]</td>
<td>631 / 507</td>
<td>625 / 550</td>
<td>613 / 550</td>
<td>550 / 526</td>
</tr>
<tr>
<td>Bend. E-Mod. [GPa]</td>
<td>21 / 19</td>
<td>19 / 19</td>
<td>19 / 19</td>
<td>20 / 21</td>
</tr>
<tr>
<td>ILS [N/mm²]</td>
<td>72 / 63</td>
<td>53 / 48</td>
<td>57 / 53</td>
<td>51 / 46</td>
</tr>
<tr>
<td>Tensile strength [N/mm²]</td>
<td>386 / -</td>
<td>423 / -</td>
<td>430 / -</td>
<td>341 / -</td>
</tr>
<tr>
<td>E-Modulus [GPa]</td>
<td>21 / -</td>
<td>21 / -</td>
<td>19 / -</td>
<td>21 / -</td>
</tr>
</tbody>
</table>

Table 1: Survey of static test results (EP Bakelite L20/SL). The specimens are conditioned in boiling water for 2 hours to achieve the wet values. B stands for binder coated fabric. Tests are conducted at room temperature. The fibre volume fraction is 43%.
There are differences in the results of the differently treated fabrics. They often originate from statistical reasons rather than from the binder. The results of the fabrics with the FK144 finish cannot be compared directly. The properties of the fabric without binder are mean values of the measurements conducted over two years (231 measurements). Even if the test results of the binder coated fabric are a bit worse than the average, they are still in the range of other test results for fabric without binder.

The tests conducted with the FK800 fabric can be compared directly. For both laminates the resin system and the fabric itself come from the same batch. The only difference is the binder. If one evaluates the results of the FK800 finish, it is obvious that the binder does not have a significant influence on fibre dominated properties like bending strength and tensile strength. The interlaminar shear strength of the binder coated fabric is reduced but still in the statistical range of other test series with the FK144 finish.

**Fatigue tests**

*Tubular specimen*

The results of the fatigue tests with the tubular test specimens can be seen in Figs 2 to 4. It can be seen that apart from the absolute results there is less scatter in the measured values of the coated fabric compared to the standard fabric. The failure shear stresses of binder powdered and pure fabric tubes are matching pretty well. A comparison of the absolute torsion moments and the failure strains indicates a better performance of the binder coated fabric.

Even if the manufacturing method itself guaranties reproducible test specimens with respect to the fibre content there is a significant difference between the powder coated fabric and the fabric without binder. The fibre volume fraction in the specimens made of the standard fabric is 52\%, whereas the specimens with binder coated fabric have a fibre volume fraction of 41\%. Obviously the binder powder on top of the fabric surface - which is not melted prior to the dry lay-up and the resin infusion - is responsible for a little gap between the fabric layers. This gap is filled with resin during the infusion resulting in a lower fibre volume fraction.

An increasing fibre fraction leads to higher stress concentrations in the matrix around the fibres. Thus, the utilisation factor of the matrix further increases with higher fibre fractions. As described above the matrix is the determining component of the tubular test specimen. Thus, the lower fibre volume fraction of the tubes made of binder coated fabric leads to an improved fatigue strength. The influence of the fibre volume fraction on the fatigue strength of GFRP laminates has also been investigated in [8].

The binder coated test tubes have slightly thicker walls. For the same amount of fibres, the static failure moment has to be bigger for the binder powdered specimens as the polar momentum of inertia increases with the wall thickness of the tubes. This can be explained using Eqn 1,

\[
\tau = \frac{M}{I_p} \cdot r
\]
where \( \tau \) represents the shear stress, \( M \) the torsion moment, \( r \) the radius of the tube and \( I_p \) the polar momentum of inertia. For tubes with small wall thickness \( t \) the polar momentum of inertia can be expressed as

\[
I_p = 2\pi \cdot R_m^3 \cdot t
\]

Eqns (1) and (2) yield

\[
\tau = \frac{M}{2 \cdot \pi \cdot R_m^2 \cdot t} = \frac{M}{2 \cdot A_m \cdot t}
\]

where \( A_m \) represents the average cross sectional area and \( R_m \) is the average of the inner and outer radius of the tube.

The wall thickness of the tube is strongly dependent on the fibre volume fraction. As could be shown in the formulas above, the polar momentum of inertia also increases and it is to be expected that tubes with lower fibre volume fractions subsequently withstand a higher failure load. If one looks at the failure shear stress the influence of the fibre volume fraction is considered in the formula through the thickness.

Commonly, the strains in a fibre composite component are taken as a measure of performance. Strains are more or less independent from the fibre volume fraction as long as the fibres carry the load. Therefore, by using an evaluation with strains one can objectively determine the performance of the fibre reinforcement. According to Fig. 4 the binder coated fibre reinforcement performs better than the fabric without binder. This is quite remarkable. Obviously the fibre volume fraction is not only responsible for a difference in wall thickness and thus a different polar momentum of inertia but also for an absolute difference in the composite strength. This is not only in fatigue as described above. It can be concluded from the fatigue tests with tubes in this work that the influence of the fibre volume fraction is far more important than the influence of the binder.

As the binder material is different to the injection resin a small decrease in the mechanical material properties is expected. This could be shown in the static tests and in other test series [5]. However, no significant decrease in mechanical strength due to the use of this binder in combination with the epoxy resin system used for this investigation has to be expected on the basis of the results from the tube specimens.
Fig. 2: Results of the fatigue tests with tubular specimens. R=0.1. Evaluation using the absolute torsion moment M. The number of load cycles is shown logarithmically in the x-axis.

Fig. 3: Results of the fatigue tests with tubular specimens. R=0.1. Evaluation using shear stress. Measured results of binder coated fabric are shown in squares, non coated fabric results are shown in full triangles.
Fatigue tests conducted with flat test specimens according to [6] are usually not evaluated using diagrams. A discrete load level is defined and the specimens have to withstand a defined number of load cycles. If this number of load cycles is achieved by the specimens of a series the material can be certified. If not, the tested combination of fibre reinforcement and matrix system can not be used in sailplanes and motorgliders.

In order to get the required information quicker and without the intention to make a material certification the load level was set to 3.5 kN, which is a bit higher than required according to the latest standard. Formerly specimens were also tested at the chosen load level for these tests and it could be shown that the required information can be obtained, too. The accuracy is not as high as with the load level for a certification but it is sufficient for the purpose here.

The tests showed a significant decrease in fatigue strength of the interface between fibres and resin due to the binder. Test specimens without binder withstand about 70000 load cycles whereas binder coated specimens failed after 15000 load cycles. However, this test is not a realistic loading of a fibre composite structure as one usually directs the fibres in the main load directions and not under ±45°. This test is only used to compare the interface of different fibre reinforcement and resin combinations. Thus, the results can neither be used for design purposes nor do they exclude a fibre-resin combination for a real composite structure.
The fatigue strength might also be influenced when the binder is melted. Possibly the binder powder which is not melted reacts differently with the main matrix system compared to the melted binder. This phenomenon has to be investigated separately and is not subject of this paper.

**CONCLUSIONS**

The influence of the tested binder system together with an epoxy main matrix system on a glass fibre fabric is small or negligible for the static material strength. It could also been demonstrated with tubular specimens with fibres under ±45° and an oscillating torsion load that there is no significant influence of the binder on the fatigue properties. Here, the binder is not melted prior to the infusion of the main matrix system and the fibres are in the main load directions.

The fatigue tests on flat specimens clearly indicated a loss in fatigue strength due to the binder. There the fibres are under ±45° to the load direction and the binder is melted prior to the incorporation of the main matrix system. Thus, there is a significant decrease in the fatigue strength of the interface between fibres and matrix due to the binder. However, this does not necessarily have an influence on the fatigue properties of a real structure under realistic loading. This could be demonstrated with the tubular test specimens.

**ACKNOWLEDGEMENTS**

We would like to acknowledge the vital co-operation with the companies CS-Interglas and MGS. Without their voluntary contributions this research work could not have been carried out within the given budget.

**REFERENCES**


