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Evaluation of adhesive binders for the development of yarn bonding for new stitch-free non-crimp fabrics

Md Abdulla Al-Monsur, Georg Bardl and Chokri Cherif

Abstract
Non-crimp fabrics (NCFs), especially multi-axial warp-knitted fabrics, are used as reinforcement materials for fiber-reinforced composites. The manufacturing of multi-axial warp-knitted fabrics by a conventional stitch bonding process to produce NCF has several disadvantages, such as filament damage, low production speed, yarn disorientation, etc. In order to overcome the existing limitations, the idea of using an adhesive binder to attach the fabric layers is a promising approach, so that the use of stitching yarns can be eliminated. The fundamental investigations presented in this paper show that the selection of the binder material has a major influence on the parameters of the textile products. Whereas the tested hotmelt adhesives offer a short curing time and a small but nevertheless sufficient bonding strength between bonded yarns, the tested reactive adhesives show a bonding strength up to 10 times higher, but at a considerably longer curing time. The reason for the different bonding strength is identified in the different penetration into the yarns. The experiments also show a significant influence of the fiber type and sizing, which needs to be taken into account when selecting fabric binders.

Keywords
non-crimp fabric, adhesive, stitch free, fiber-reinforced composite, binder

Introduction
Traditional construction materials, such as aluminum and high-strength steels, are being replaced by textile-reinforced composites because of their prominent lightweight potential in many specific application areas, like the production of different component parts of aircraft, automobiles and sports, where both the strength and stiffness are crucial. The application of the textile composite reduces the weight of the compound to a great extent. Therefore, textile-reinforced composites have emerged as a leading trend in lightweight structure design. Non-crimp fabrics (NCFs) are structures made of one or several layers of straight yarns laid upon each other and transformed to a fabric normally by a stitching process, through which they remain straight and free of any substantial crimp. The produced fabrics are easier to cut and handle as the stitching holds the material together. Stitch-bonded NCFs have become the material of choice for the production of textile-reinforced composites because of the possibility to include several layers of variably oriented yarns (e.g. $+45^\circ$, $-45^\circ$, $0^\circ$, $90^\circ$) in one fabric, therefore allowing one to specify yarn orientation according to loading requirements. Also NCFs show better mechanical properties (strength, Young's modulus, degree of drape), flexibility in design and low production cost compared with the most widely used woven fabrics. They can be used for the composite production process (VARI, RTM, etc.). Multi-axial warp-knitted fabrics are currently used in a wide variety of application areas, such as construction of automobiles, aerospace components, geo textiles, vessel body parts and pneumatic materials.

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The mechanical properties of the composites made of NCF are affected by the quality of the textile reinforcement. For example, Airbus Industry Material Specifications (AIMS) define and quantify different defects of NCF, for example crease or wrinkle, cut or tear (adjacent yarn is cut or broken), yarn splice (broken or severed yarn, which is rejoined), fuzz ball (accumulation of loose or frayed fibers within fabric or on the surface), gap (open space between parallel fibers or between the filaments), missing knitting loop, incorrect fiber orientation and missing reinforcement yarn. In addition to the possibility of the introduction of these defects, the current production process for NCFs through stitching results in several major drawbacks. Stitching causes distortions of the fiber orientation in the fibrous mat. Also, it leads to a displacement of parallel fibers, resulting in gaps or openings. The tension of the warp-knitting yarn may lead to the local compaction of the reinforcement fiber bundles, resulting in gaps and so-called “fish eyes” in the fabric that in turn lead to matrix-rich parts in the composite, also resulting in a degradation of mechanical properties. Furthermore, the yarns face damages by means of the stitching needles, which in most cases pierce through the individual reinforcement yarns. The mechanical characteristics of the composites are affected negatively in both cases, which results in a decrease of strength of the composite materials by up to 40%. In order to overcome these problems, it is necessary to develop an alternative process, which can improve the performance of NCF. In this regard, the aim of the latest investigation is to remove the stitching yarns and replace the stitch bonding of the yarns by a bonding through adhesive binders. Binders are currently used in pre-forming in several ways. Textile preforms have been produced through a process termed “chemical stitching”, in which binder is applied. In this process, adhesive binder is applied between the textile layers using infusion needles. Different energy sources are used for the fast curing of the binder. Afterwards the textile is proceeded to the next position. A method to produce NCF with fusible thread is developed, where the aim is to maintain preferably 1–3% fusible thread by mass. A process to bond the glass fiber using water soluble or water dispersible and curable polyester resins is realized, in which curable polyester resin binders can be used for a number of applications, especially for glass fiber bonding in the production of fiberglass insulation products. The wettability of the adhesives to the fiber surface, surface topography, the functionality and the feasibility of the binder application is already investigated. In this regard, a surface modification of the glass and carbon fibers is investigated for improving the adhesion of chemical substances to the fibers, which also aims at the realization of adhesive-bonded NCFs. However, a direct application is not yet reported to produce the NCF using binder. Therefore, the investigation on the fabrication of adhesive-bonded NCF is highly demanded.

For the binder application, the following general requirements can be derived:

- the amount of binder should be small in order to avoid an increase of the stiffness of the produced fabric and to minimize possible imperfections in the matrix, which could be starting points for cracks;
- the bonding strength should be sufficient during fabrication, so that the produced fabrics can be handled and transported to the further process;
- the binder points should in general be homogeneously distributed;
- the binder should be compatible with the matrix of the composites;
- the produced fabric should be drapable after the applied force.

For the execution of the alternative production process of NCF, the warp-knitting unit is to be replaced by a suitable binder application unit. Primarily two possible approaches can be advised in this regard as follows.

1. **Surface application**: application of adhesive binder to each weft yarn layer in order to bond it with the next layer.
2. **Injection**: application of the adhesive binder with a nozzle system to bond all the layers of the fabrics together with a needle, in which the needle is placed at the nozzle end.

Figure 1 shows the possible production processes.

In this work, fundamental investigations are done on the minimum drop mass of different adhesive binders and the strength of the bonding joints between the yarns with a microscopic view of the bonding cross-sections. Furthermore, the curing time of the adhesives is also analyzed, since it is an important factor affecting the processing time during the fabrication.

**Materials**

**Adhesives**

In this investigation reactive and hotmelt adhesives are used. The reactive adhesives are liquid at room temperature and are cured by a reaction based on polymerization, polycondensation or polyaddition. As reactive adhesives, cyanoacrylates are selected because of their short curing time (usually 3–10 s). Different reactive adhesives from different manufacturers with different viscosities are used in order to analyze the effect of
the viscosity on the binder penetration and thus on the bonding strength.

The hotmelt adhesives, on the other hand, are solid in room temperature and need to be heated for application. Normally they have high viscosity. Regarding the base materials, there are different types of polymeric or co-polymeric materials. In this work, Bühnen Avenia B42042.1 is selected as a standard polyolefin adhesive from the packaging industry and Planatol HM Ultimate 1 because of its low viscosity. They are supplied as solid grains with a diameter of about 4–5 mm (cf. Figure 2).

The properties of hotmelt and reactive adhesives used in this work can be seen in Table 1.

**Fiber materials**

In order to evaluate the compatibility of the binders, different types of multifilament yarns are used in this study. As can be seen in Table 2, two types of glass and two types of carbon fiber yarns are used, with the different sizing being the only difference between the two types of fiber material.
Experimental tests

Application of adhesives

For the application of the reactive adhesives, an industrial application device Delomat 400 (Delo, Munich, Germany) is used, which can be seen in Figure 3. The important parameters that are determinant for the application of reactive adhesives are nozzle pressure, nozzle opening time and nozzle diameter. Nozzle pressure and nozzle opening time can be controlled by the machine settings. The adhesive is dispensed with pressure through the output nozzle. Needles with specific diameter are used at the nozzle end to control the amount of adhesive. The diameter of the needle used in this work is 0.4 mm. Nozzles of smaller diameter have been shown to not yield a reproducible drop formation (i.e. the drops do not separate from the nozzle).

The yarns are placed perpendicular in a spring holder, which ensures a straight fiber orientation and the yarns being positioned directly under the adhesive nozzle. The spring holder with crossed carbon fiber yarns can be seen in Figure 3 on the left, directly under the adhesive dispensing unit. The upper yarn is slightly deflected to the side by hand, then the nozzle is activated and the binder drop is applied. The upper yarn is then placed back over the binder drop and the lower yarn. A load is applied immediately to keep the joint under pressure for the better attachment. After the curing time the joined yarns are removed from the device.

For the application of the hotmelt adhesives, an industrial application device Bühnen 6040 (Bühnen Adhesive System, Germany) is used (c.f. Figure 4). The important parameters for the application of hotmelt adhesives are the temperatures of the tank, tube and nozzle, the nozzle pressure, nozzle diameter and nozzle opening time. The opening impulse is triggered manually by an optical sensor and thus the nozzle opened according to the adjusted opening time. The nozzle diameter used in this work is 0.3 mm. The bonding procedure is the same as that described above for the reactive adhesive.

Drop mass and curing time analysis

The adhesive drop mass is analyzed in order to select the machine settings for the binder to maintain the
minimum binder content with a specific processing parameter. The average drop mass of the adhesives is investigated by weighing a set of 10 consecutive drops on a precision scale. There is no specific norm for the determination of the curing time. The curing time is determined as the time after which the yarns could not be separated by the force of gravity alone, and the bonding is sufficient to carry the weight of the yarn.

**Bonding strength analysis.** In order to determine the bonding strength at the minimum achievable binder content, the setting for the minimum drop mass was used for the analysis. The test is performed with the tensile testing
There is no specific norm available for this test. The testing principle is developed by ITM using a special sample holder (Figure 5). The experiments are carried out following the parameters below:

- **Test speed (mm/min)** - 10
- **Clamping distance (mm)** - 2
- **Load sensor (kN)** - 2.5
- **Travel sensor** - Traverse
- **Pre load** - 0

The testing samples are made with the sample holders, in which two perpendicularly bonded yarns are placed. Before preparing the samples, the upper sample holder is attached to the upper clamp of the testing machine. After that, one yarn is attached to the lower sample holder and this holder is placed in the lower clamp of the testing machine. Finally, the other yarn is attached to the upper sample holder (which has been fixed to the upper clamp). The bonding point is maintained at the middle of both holders for the even application of the force. Then the force is applied until the complete debonding of the joint has occurred. The sample fitting can be seen in Figure 5. Figure 6 shows the bonded sample yarn, with fibers attached perpendicular to each other, that can be considered as a unit cell of a biaxial fabric.

**Microscopic analysis.** In order to examine the penetration of the binder into the yarns at the bonded crossing...
joint, the joints are analyzed under a microscope (see Figure 7). The cross-sections of the bonded joints are embedded in an epoxy resin matrix followed by curing at ambient temperature. Then the samples are prepared for testing after polishing properly several times. The microscopic analysis is performed with Microscope Axiotech 100 (Carl Zeiss AG, Germany).

Results and discussion

Adhesive drop mass and curing time

The drop mass is increased with the increasing nozzle opening time, nozzle pressure for processing both types of adhesives and with increasing temperature (in the case of hotmelt adhesives). For a shorter opening time or smaller needle diameter no drop formation at the nozzle tip is observed. Different pressure settings are required for different adhesives depending on the viscosity of the material, that is, reactive adhesives of higher viscosity require higher pressures and in some cases longer opening times in order to obtain a drop formation at the nozzle tip. The same applies for hotmelt adhesives, although it should be noted that due to the different application systems the pressure and opening time values for reactive adhesives are not comparable to those of hotmelt adhesives. Table 3 shows the parameter determined for the minimum drop mass for different adhesives processed in this work. The minimum drop mass is found to be 1.3 mg for hotmelt

Figure 6. Bonded yarn in rest (a) and bonded yarn fitted in the testing apparatus during strength analysis (b).

Figure 7. Cross-sectional view of a bonded joint.
and 6.7 mg for reactive adhesives, where the drop formation is regular and continuous.

In the case of the reactive adhesives, Delo 2153 requires the highest pressure, whereas Loctite 406 can be processed with the lowest pressure due to its lowest viscosity. The minimum nozzle opening time remains almost the same for all the reactive adhesives, except Delo 2153, because of its higher viscosity. In the case of the hotmelt adhesives, Bühnen Avenia B42042.1 requires a pressure twice as high as that required for Planatol HM Ultimate 1 because of the higher viscosity (almost double). The nozzle opening time and the temperatures for minimum drop mass are the same for both. Figure 8 shows the minimum drop mass for different adhesives studied in this work. The error bars show the standard deviation.

The reactive adhesives require a considerable time for curing, which is presented in Figure 9. The curing time required by hotmelt adhesives is very small (<2 s), due to the small drop mass, and is not further investigated.

### Bonding strength

The minimum dispensable drop mass of the different adhesives is used to prepare the samples for the bonding tests. Figure 10 shows the bonding strength of reactive adhesives with different types of yarns. Each test

<p>| Table 3. Determined process parameters for minimum drop mass of reactive and hotmelt adhesives |</p>
<table>
<thead>
<tr>
<th>Parameter</th>
<th>Delo 2207</th>
<th>Delo 2219</th>
<th>Delo 2153</th>
<th>Loctite 406</th>
<th>Loctite 4850</th>
<th>Bühnen Avenia B42042.1</th>
<th>Planatol HM Ultimate 1</th>
</tr>
</thead>
<tbody>
<tr>
<td>Viscosity of adhesive (mPas)</td>
<td>100</td>
<td>240</td>
<td>2153</td>
<td>20</td>
<td>400</td>
<td>2350</td>
<td>1300</td>
</tr>
<tr>
<td>Nozzle diameter (mm)</td>
<td>0.4</td>
<td>0.4</td>
<td>0.4</td>
<td>0.4</td>
<td>0.4</td>
<td>0.4</td>
<td>0.4</td>
</tr>
<tr>
<td>Nozzle opening time (s)</td>
<td>0.1</td>
<td>0.1</td>
<td>0.15</td>
<td>0.1</td>
<td>0.1</td>
<td>0.01</td>
<td>0.01</td>
</tr>
<tr>
<td>Pressure (bar)</td>
<td>0.5</td>
<td>1</td>
<td>5</td>
<td>0.25</td>
<td>2.15</td>
<td>2</td>
<td>1</td>
</tr>
<tr>
<td>Tank temperature (°C)</td>
<td>N/A</td>
<td>170</td>
<td>N/A</td>
<td>175</td>
<td>N/A</td>
<td>180</td>
<td>N/A</td>
</tr>
<tr>
<td>Tube temperature (°C)</td>
<td>N/A</td>
<td>170</td>
<td>N/A</td>
<td>175</td>
<td>N/A</td>
<td>180</td>
<td>N/A</td>
</tr>
<tr>
<td>Nozzle temperature (°C)</td>
<td>N/A</td>
<td>180</td>
<td>N/A</td>
<td>180</td>
<td>N/A</td>
<td>180</td>
<td>N/A</td>
</tr>
<tr>
<td>Minimum drop mass (mg)</td>
<td>8.9</td>
<td>7.0</td>
<td>6.7</td>
<td>7.1</td>
<td>7.2</td>
<td>3.0</td>
<td>1.3</td>
</tr>
</tbody>
</table>

**Figure 8. Minimum drop mass for different adhesives.**
result is a mean value of seven repetitive experiments. In general, all reactive adhesives attach better with carbon fiber than with glass fiber. The sizing of the carbon or glass fibers, which is the only difference between the two types of carbon/glass fibers tested, has a strong influence. Therefore, CF HTS 45 E23 is attached better than CF HTS 40 F13, except with Delo CA 2207 and with Loctite 4850. On the other hand, GF EC 600 350 is attached better than GF EC 600 354 with all types of reactive adhesives. Typical force-displacement curves of Delo 2219 with CF HTS 45 and GF EC 600 350 can be seen in Figures 11 and 12, respectively. Each curve marks an individual test with the stated yarn and adhesive parameters.

In comparison with the results from the reactive adhesives, hotmelt adhesives show significantly weaker strength at the bonded joints of yarns. The maximum forces required to break the bonded joints by hotmelt adhesives can be seen in Figure 13. In this case, it can be clearly observed that the attachment of both the hotmelt adhesives is almost the same with both types of carbon fiber. However, the adhesives attach better with GF EC 600 354 than with the other types of yarns. It is remarkable that the bonding strength of...
the hotmelt adhesives with GF EC 600 350 is comparatively lower because of the different sizing material. Overall, a higher bonding strength of Bühnen Avenia B42042.1 can be seen than that of Planatol HM Ultimate 1; however, this might also be due to the higher minimum drop mass used. Typical force–displacement curves of Bühnen Avenia B42042.1 with CF HTS 40 and GF EC 600 354 can be seen in Figures 14 and 15, respectively.

It is noted during the experiments, that the adhesive-bonded joints debond completely without the rupture of filaments in the case of hotmelt adhesives. On the other hand, the adhesive-bonded joints do not debond completely in the case of reactive adhesives. During the
experiments performed with the reactive adhesives, filament breakage occurred in some cases. Thus, it can be assumed that the strength of the bonded joints is even higher than the measured results.

**Microscopic analysis of bonding points**

The microscopic images taken from cross-sections of the bonded joints show that the reactive adhesives penetrate inside the filaments in a yarn and migrate up to a certain distance along the yarn. The reactive adhesive’s penetration in carbon fiber can be seen in Figure 16. The adhesive can be distinguished as slightly filling the inside of the yarns and covering them. The penetration of reactive adhesives in glass fiber (Figure 17) is similar. As can be seen, the reactive adhesive fills the gaps between individual filaments, also bridging the space between the upper and lower yarn. This results in a stiffening of the yarns in the region of the joints. Therefore, any tension applied to the joints is
transmitted to many individual filaments and the effective area of the bond is higher, which results in a high bonding strength.

Hotmelt adhesives, on the other hand, remain between the yarns on their respective surfaces and do not penetrate into the yarns due to their high viscosity (Figure 18). This means that only the filaments on the surface are attached to the binder, which form a solid connection lying between the yarns. Therefore, any force applied to the joints is transferred to only a few fibers, which results in a rather small effective bonding area, compared with that of reactive adhesives. The missing penetration into the yarns therefore explains the relative weakness of the hotmelt adhesive bonding compared to the reactive adhesives.
Conclusions

The goal of this study is to evaluate the processability and bonding behavior of hotmelt and reactive adhesives for bonding of glass and carbon fibers. Due to the smaller drop mass attainable with industrial standard nozzles and shorter curing time, hotmelt adhesives show a better processability. The bonding strength of the adhesives is examined through transverse tensile testing of the adhesive-bonded yarns. It is noteworthy that the bonding strength varies greatly for the different adhesives. From the results, it can be seen that no general recommendation can be given as to which reactive adhesive gives the highest strength. Indeed, just changing the sizing of the fibers can, in combination with certain binders, have a huge impact on the bonding strength. Each adhesive displays a different bonding strength for glass and carbon fibers, also depending on the fibers’ sizing. Nevertheless, a clear distinction can be made regarding the adhesive type: whereas hotmelt adhesives result in rather small bonding strengths (which are nevertheless sufficient to provide a bonding for handling the yarns), the tested reactive adhesives lead to bonding strengths up to 10 times higher. The reason can be found in the different penetration behavior of hotmelt and reactive adhesives due to their different viscosities (with the reactive adhesives’ viscosity being one or two orders of magnitude lower). The hotmelt adhesives do not penetrate through the filaments and also do not migrate along the yarn length. On the contrary, the reactive adhesives penetrate inside the yarns and migrate a certain length along the yarn length.

Regarding processability, the minimum drop mass can be obtained by the hotmelt adhesive. It also provides some benefits, such as better processability, easier processing, no risk of nozzle jamming, shorter curing time and smaller dosing. On the contrary, the highest bonding strength can be achieved by the reactive adhesive. It also shows a better penetration into the yarns. However, the difficult processing proves to be a drawback. The results presented here are intended as a first step in the development of a novel production process for NCFs that replaces the stitching yarns by adhesive binders. In order to develop a textile machine that integrates the adhesive application, the advantages and disadvantages of the two adhesive types will need to be taken into account. Since the bonding strength depends to a large extend on the fibers used, any machine integration will need to provide for different selections of binders depending on the yarn type processed.

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References


