EFFECT OF REPAIR DESIGN AND SURFACE TREATMENT ON ADHESIVELY BONDED COMPOSITE REPAIRS

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Abstract
The effect of surface preparation on the mechanical performance of adhesively bonded scarf repairs with varying scarf angles under quasi-static tensile loads will be investigated. To assure proper bonding of the repair-plies to the pre-cured laminate, two types of adhesive have been used. Additionally to sanding, a combination of corona treatment with a subsequent wet chemical functionalization by an organosilane has been applied. Potential changes in surface energy attributable to the chemical surface treatment were assessed by contact angle measurements. These measurements show an increase in surface energy for the functionalized surfaces. Tensile testing indicates that no significant changes in tensile strength arise from neither the type of adhesive nor from the surface functionalization. For shallow scarf angles, the fracture mode and tensile strength remain nearly constant. As the scarf angle is changed to a steeper angle, tensile strength is reduced and the failure mode switches from a tensile to a predominantly cohesive failure.

1. Introduction

As a consequence of the increasing amount of fiber-reinforced polymers being used in primary civil aircraft structures, a substantial risk of damages to such parts exists [1–3]. In order to avoid the costs for the replacement of damaged parts, various repair methods (e.g. bonded and bolted repair techniques) have been developed [1, 2, 4]. In particular, bonded patch repairs offer several advantages concerning e.g. aerodynamic smoothness and load transfer [2, 4, 5]. Nevertheless, they are challenging in terms of ensuring durability and in general continuing airworthiness of the repaired structure [5, 6].
Within this paper, the influence of a two-step surface functionalization, based on previous works [7, 8], on the surface energy of an epoxy based carbon fiber reinforced laminate will be investigated [9]. By using this surface treatment, consisting of a corona treatment followed by a wet chemical functionalization with an organosilane, it is intended to introduce chemical bonds between the functionalized surface and an adhesive. The corona treatment, a commonly used surface treatment method for polymers [10, 11], is used to increase the surface polarity as well as to generate bonding sites for the organosilane [7, 12]. The organosilane itself works as a coupling agent between the surface and the adhesive, therefore an organosilane with an epoxy group has been selected to match the adhesive used in the subsequent production of repair-specimens. Studies for similar applications have been conducted for e.g. aluminum

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substrates [13], but organosilanes containing methoxy groups are also known to form bonds to certain functional polymers [12]. Furthermore, the influence of this surface functionalization as well as of different adhesives and scarf angles on the tensile strength of specimens with tapered, adhesively bonded joints (further addressed as "repair-specimens") will be analyzed [9, 14]. Additionally, the corresponding fracture patterns will be categorized as well.

2. Experimental part

2.1. Materials

The commercially available Cycom® 977 - 2 prepreg material (Cytec Solvay Group, Tempe, US), consisting of an epoxy-based thermosetting resin reinforced with woven carbon fibers, was used in this study. Laminate plates were produced from this prepreg by following the stacking sequence $(+45, 0, -45, 90)_S$, resulting in a quasi-isotropic, symmetric laminate. All laminates were autoclave cured at 180 °C for 2 h under a pressure of 6.6 bar.
A supported film adhesive (Scotch-Weld™ AF 163-2L), supplied by 3M (Saint Paul, US), with a nominal thickness of 0.14 mm and an unsupported one (Scotch-Weld™ AF 163-2U) from the same supplier with a nominal thickness of 0.24 mm were chosen for bonding of the repair-joints. These film adhesives are epoxy based and offer the possibility to be cured under vacuum at 180 °C.
The surface functionalization was conducted using (3-(2,3-Epoxypropoxy)propyl)trimethoxysilane (subsequently addressed as "epoxysilane") provided by Wacker Chemie (München, DE) as well as 2-propanol and tetrahydrofuran purchased from Carl Roth (Karlsruhe, DE).

2.2. Specimen manufacturing

Repair-specimens according to AITM 1-0029 [14] with the two different adhesives as well as varying scarf angles represented by scarf ratios (for both adhesives 1:50, 1:40, 1:30 and 1:20 as well as 1:9 for the supported one) were produced (see Table 1 for a compilation of the amount of specimens).
Laminate plates with dimensions according to the respective scarf ratios were manufactured and checked for voids or micro-cracks via ultrasound. Subsequently, tapering was performed using an angular grinder (grit 100) with respect to the scarf ratios leading to taper areas with sanded surfaces. In addition, selected sanded surfaces were functionalized by the previously mentioned two-step procedure (see Table 1).
All tapered plates, either with sanded or with functionalized surfaces, were repaired using a soft patch approach, meaning that a layer of film adhesive was put in place on the taper area. Subsequently, slightly overlapping prepreg plies (according to the scarf ratio) were laid up on the film adhesive. The resulting repaired laminate plates were cured using the above mentioned cure cycle. It has to be noted, that for the functionalized surfaces, the pressure was slightly reduced to 6 bar. After curing, the repaired plates were checked via ultrasound as well. In order to assess the mechanical performance of the repair-specimens in comparison to the unrepaired ones, reference specimens were produced from unrepaired laminate plates as well.

Table 1. Amount of repair-specimens produced for varying scarf ratios, types of surface treatment and adhesive types.

Scarf ratio / surface treatment →	1:50		1:40	1:30	1:20		1:9
Adhesive type ↓	Sanded	Funct.	Sanded	Sanded	Sanded	Funct.	Sanded
AF163-2L	8	8	8	16	8	8	8
AF163-2U	8	8	8	16	8	8	0

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All specimens with dimensions depicted in Fig. 1 were cut from the corresponding laminate plates via a water cooled circular saw equipped with a diamond coated disk (Diadisc 5200, Mutronic Praezisionsgeraetebau, Rieden, DE). End tabs manufactured from glass fiber reinforced epoxy resin were bonded to the specimens by AF163-2L film adhesive. Curing of the film adhesive was achieved in a plate press (P300 E+, Dr. Collin, Ebersberg, DE) using the adhesive producer´s recommended cure cycle.

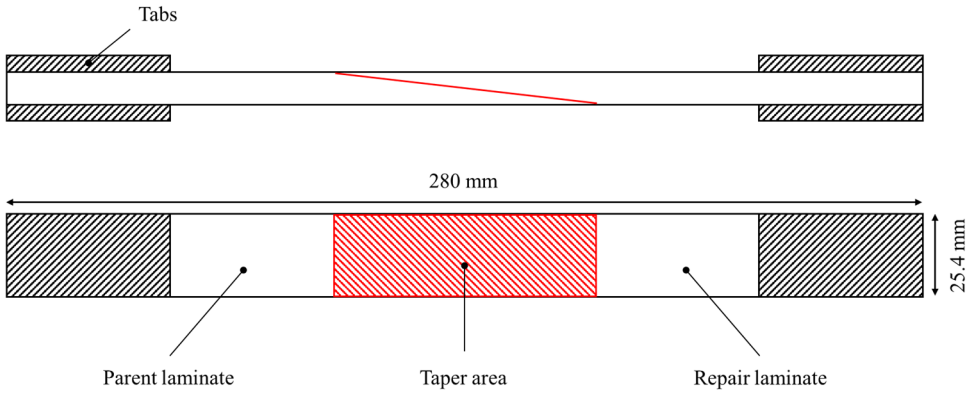

Figure 1. Schematic illustration of a repair-specimen.

Furthermore, specimens for the contact angle measurements with the dimensions of 40 x 10 mm (length x width), were cut from the unrepaired laminate by a water cooled disk saw (Diadisc 5200, Mutronic Praezisionsgeraetebau, Rieden, DE) and subsequently sanded and polished using a grinding/polishing machine (Phoenix Beta, Buehler, Lake Bluff, US) equipped with sanding paper (consecutively starting from grit P600 until P4000 was reached) as well as a polishing plate and diamond suspension containing particles with a diameter of 3 µm (MetaDi, Buehler, Lake Bluff, US). This was performed in order to minimize the influence of surface roughness on the contact angle measurement.

2.3. Surface functionalization

The surface functionalization was achieved by a two-step procedure. The respective areas were rinsed with 2-propanol in order to remove surface contaminations before being subject of an atmospheric-pressure plasma treatment (Laboratory corona station PG 3001, Ahlbrandt System, Lauterbach, DE). Subsequently, the epoxysilane was applied to the tapered surfaces with a brush or via immersion or a brush for the contact angle measurements. After ~24 h the excess epoxysilane was rinsed off with tetrahydrofuran, followed by a drying step (70 °C for ~20 min). Following each of the rinsing steps, the respective surfaces were dried with compressed carbon dioxide to remove excess solvents. It has to be noted, that the previously described repair process was started within ~2 days after the surface functionalization was carried out.

2.4. Contact angle measurement, tensile testing and optical analysis

Contact angle measurements were performed at standard ambient conditions on drops of de-ionized water and diiodomethane using a drop shape analyzer (DSA100, Kruess, Hamburg, DE). The calculation of the surface energy from the drop shapes was performed according to Owens, Wendt, Rabel and Kaelble [15]. Quasistatic tensile testing was conducted on at least 8 specimens using a universal tensile/compression testing machine (Z250, Zwick, Ulm, DE) equipped with a load cell (250 kN load bear-

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ing capacity) and wedge-screw grips designed for 250 kN maximum load. The tensile tests were performed according to AITM 1-0029 [14] with an initial grip separation of 180 mm and a test speed of 2 mm/min at room temperature. Tensile strength was calculated from the maximum load divided by the thickness of the parent laminate and the width of the specimen (each as a mean of three individual measurements per specimen). The failure modes were subsequently categorized according to AITM 1-0029 [14] as well. Corresponding pictures of the fracture patterns were taken by a digital single-lens reflex camera equipped with a standard lens (EOS 600D and EF-S 18-135mm f/3.5-5.6 IS, Canon Inc., Tokyo, JP).

3. Results and discussion

3.1. Surface energy

Contact angle measurements and the resulting surface energy depicted in Fig. 2 (a) show that through the surface functionalization by corona and subsequent wet chemical treatment with epoxysilane, the total surface energy (consisting of the polar and the dispersive part) can be raised in comparison to the untreated, polished samples. Especially the polar part of the surface energy is increased by the treatment, regardless whether the epoxysilane was applied by immersion or by a brush. This is important for an actual repair process, since not all parts can be immersed in a chemical agent. Figure 2 (b) shows the surface energy as a function of time for the functionalized surfaces. It can be observed that the total surface energy as well as the ratio of polar and dispersive parts remain relatively stable for at least one week. This is in accordance with previous findings [7] and with the timeframe from the end of the surface functionalization to the start of the repair process involved in the repair-specimen production.

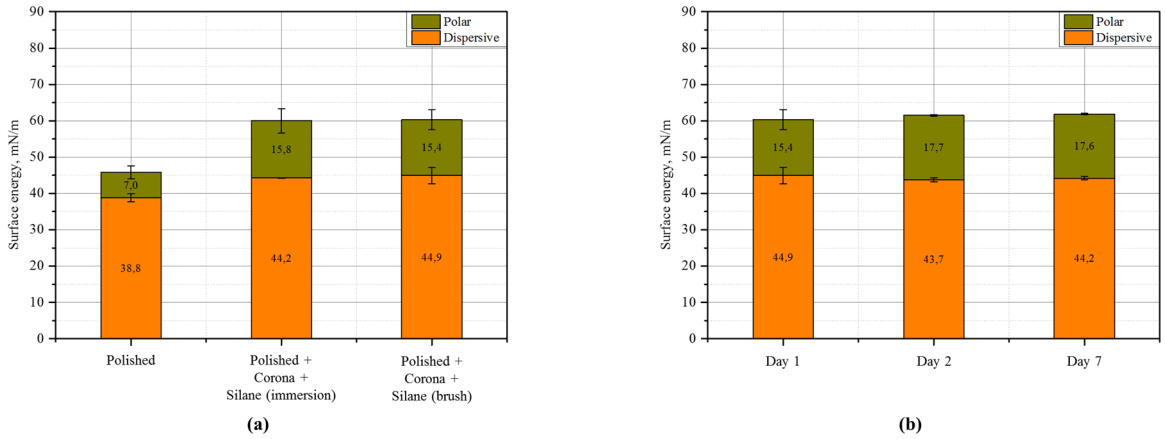

Figure 2. Surface energy for polished and functionalized surfaces (a) and surface energy of functionalized surfaces as a function of time (b).

3.2. Quasi static tensile tests

Figure 3 and Fig. 4, respectively, show the mean relative tensile strength (normalized to the reference´s tensile strength) of the repair specimens with sanded and chemically modified surfaces as a function of the scarf ratio for the supported and the unsupported adhesive. In terms of better visibility, the repair-specimens with functionalized surfaces as well as their corresponding counterparts with identical scarf ratios and sanded surfaces, and additionally the reference specimens´ tensile strength are highlighted.
Specimens with scarf ratios of 1:30 were produced from two different repaired plates for each type of adhesive, with the intention of investigating the reproducibility of the repair process itself (see also Table 1). As depicted in Fig. 3 and Fig. 4, no significant changes in relative tensile strength exist for

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these specimens, indicating a stable repair process. On the one hand, for scarf ratios of 1:20 to 1:50, no significant impact of the variation in scarf ratio on the relative tensile strength can be found in neither the supported nor the unsupported adhesive, when taking into account the standard deviations. On the other hand, no significant influence of the surface functionalization could be detected, which could be related to the shallow scarf angles resulting in tensile failure in the parent, the scarf or the repair laminate region rather than in the vicinity of the bonded surfaces (cohesive or adhesive failure).
For the scarf ratio of 1:9, a significant reduction in relative tensile strength to ~55 % as well as a failure mode transition to a cohesive failure can be observed, which could be attributed to the steeper scarf angle of approximately 6 °. A scarf angle dependent transition in fracture mode was also found by e.g. Twist et al. [16] for a quasi-isotropic laminate using a hard patch repair approach.

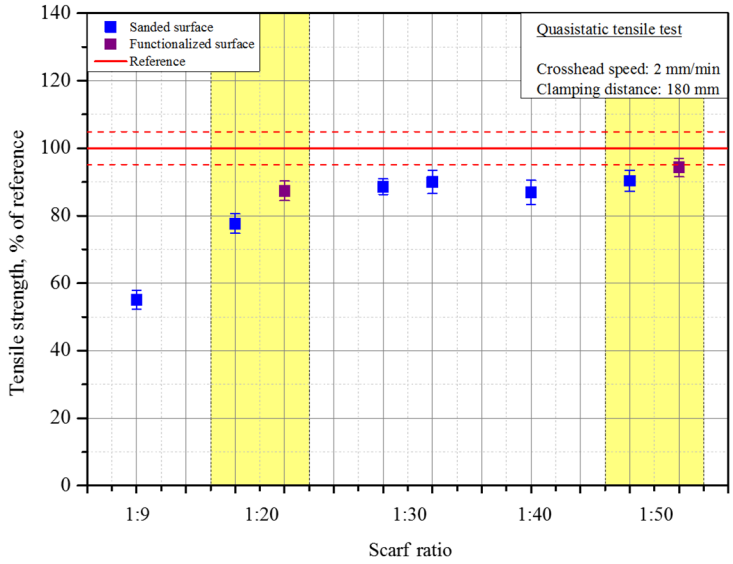

Figure 3. Repair-specimens´ relative tensile strength as a function of scarf ratios for the supported adhesive and both surface preparation methods (sanding and chemical functionalization) [9].

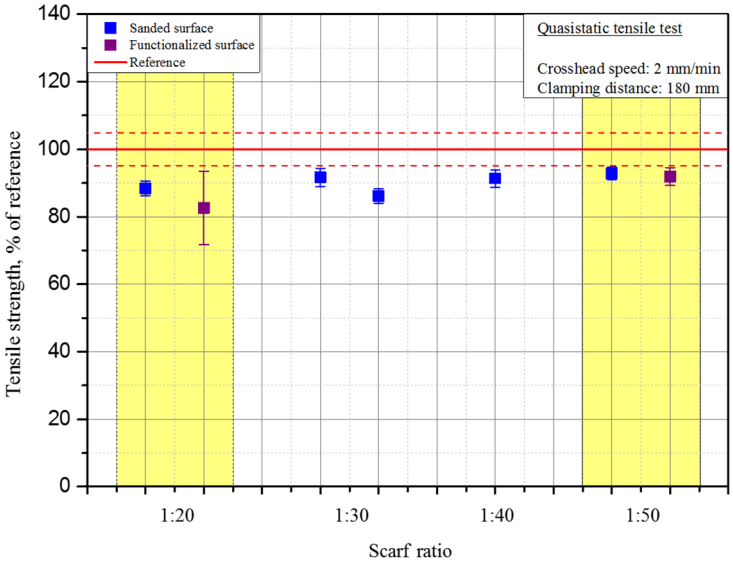

Figure 4. Repair-specimens´ relative tensile strength as a function of scarf ratios for the unsupported adhesive and both surface preparation methods (sanding and chemical functionalization).

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Figure 5 exemplarily shows the failure modes for specimens with functionalized as well as sanded surfaces with the supported adhesive. All specimens with scarf ratios of 1:20 to 1:50 show a tensile failure either in the parent, the tapered or the repair region depending on the scarf ratio. Comparable results were found for the unsupported adhesive as well. In contrast, the specimens with 1:9 scarf ratio exhibit a predominantly cohesive failure pattern. Failure hereby occurred either partially along the bondline as well as in the parent or the repair laminate or in the bonding region entirely.

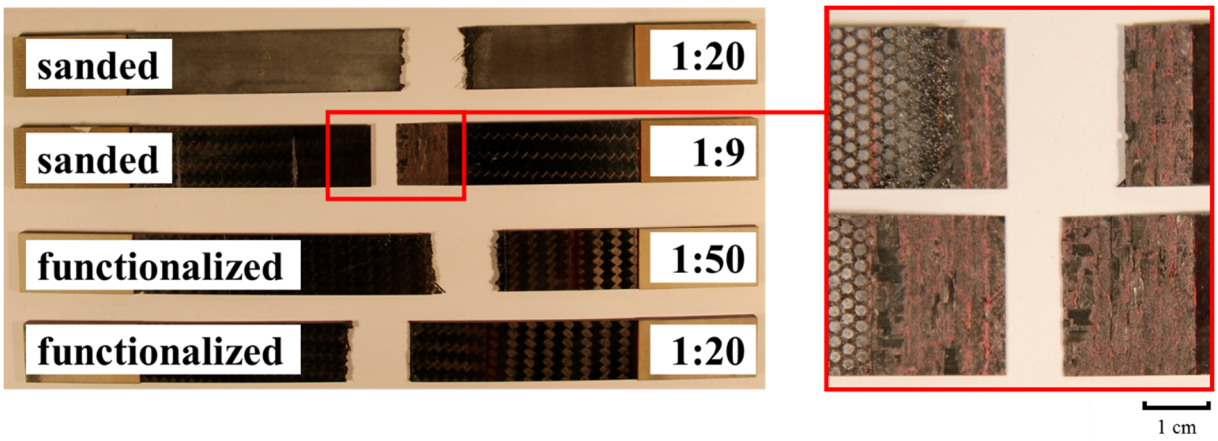

Figure 5. Exemplary failure modes of specimens for the supported adhesive with sanded and functionalized surfaces (left hand side) as well as detailed view of the failure mode for a scarf ratio of 1:9 (right hand side).

4. Conclusion

The contact angle measurements showed that on the one hand, the total surface energy (especially the polar part) was increased through the surface functionalization (a combination of corona and wet chemical treatment by an epoxysilane). On the other hand, this increase in surface energy was stable for at least one week, which was relevant for the repair process as conducted in the current study.
A significant increase in relative tensile strength as a result of the surface functionalization could not be detected, which could be related to the specimens´ failure patterns (tensile rather than cohesive or adhesive failure). Furthermore, no distinct influence of neither the type of adhesive (supported or unsupported) nor the scarf ratio (1:20 to 1:50) on the relative tensile strength could be demonstrated. For the 1:9 scarf geometry, a decisive change in both tensile strength as well as in failure mode could be observed. By cutting repair specimens with 1:30 scarf ratio from two repaired plates each for both adhesives, the reproducibility of the repair process could be confirmed.
Further work will be done concerning the influence of different, aviation industry relevant ambient conditions (e.g. hot / wet conditions or various immersion media). Hereby, the behavior under quasistatic, fatigue and impact loading on repair-, compression after impact and single lap shear specimens, respectively, will be investigated [9]. The aim of this work is to gain extended insight in bonding durability and damage tolerance of adhesively bonded repairs as well as in the influence of the surface preparation method.

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