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Shear Strength of Adhesive Bonding of Plastics Intended for High Temperature Plastic Radiators

Ilya Astrouski, Tereza Kudelova *, Josef Kalivoda and Miroslav Raudensky

Heat Transfer and Fluid Flow Laboratory, Faculty of Mechanical Engineering, Brno University of Technology, Technicka 2896/2, 616 69 Brno, Czech Republic; ilya.astrouski@vut.cz (I.A.); josef.kalivoda@vut.cz (J.K.); miroslav.raudensky@vut.cz (M.R.)
* Correspondence: tereza.kudelova@vut.cz

Abstract: The use of adhesive joints has increased in recent decades due to their competitive features in comparison with other joining methods. They can be used in specific applications where there is no possibility to use alternative connection techniques. Adhesive bonding was used to assemble the prototype of a high-temperature car radiator (operated up to 125 °C) with a total of 12,240 plastic tubes. This work aims to estimate the shear strength of different adhesives intended for bonding the plastics used to assemble the above-mentioned high-temperature radiator. Fourteen commercial adhesives were tested with one thermoset plastic (G11 glass fabric epoxy sheets) and two glass-reinforced thermoplastics (polyamide PA66-GF30 and polyphenylene sulfide PPS-GF40). Tests were conducted according EN 1465 to determine tensile lap-shear strength of bonding. Testing showed that only 4 of the 14 adhesives tested exhibit substantial bonding strength at temperatures above 120 °C and only one is resistant at 180 °C. The AS60/AW60 adhesive showed the best results for all three substrates: 1.6 MPa for epoxy sheets and PA66-GF, and 1.4 MPa for PPS-GF40. Additionally, the influence of the surface treatment with cold plasma was evaluated on a clean and activated bonding surface, causing a 30% increase in the shear strength.

Keywords: plastics; adhesion; gluing; shear strength; fracture analysis

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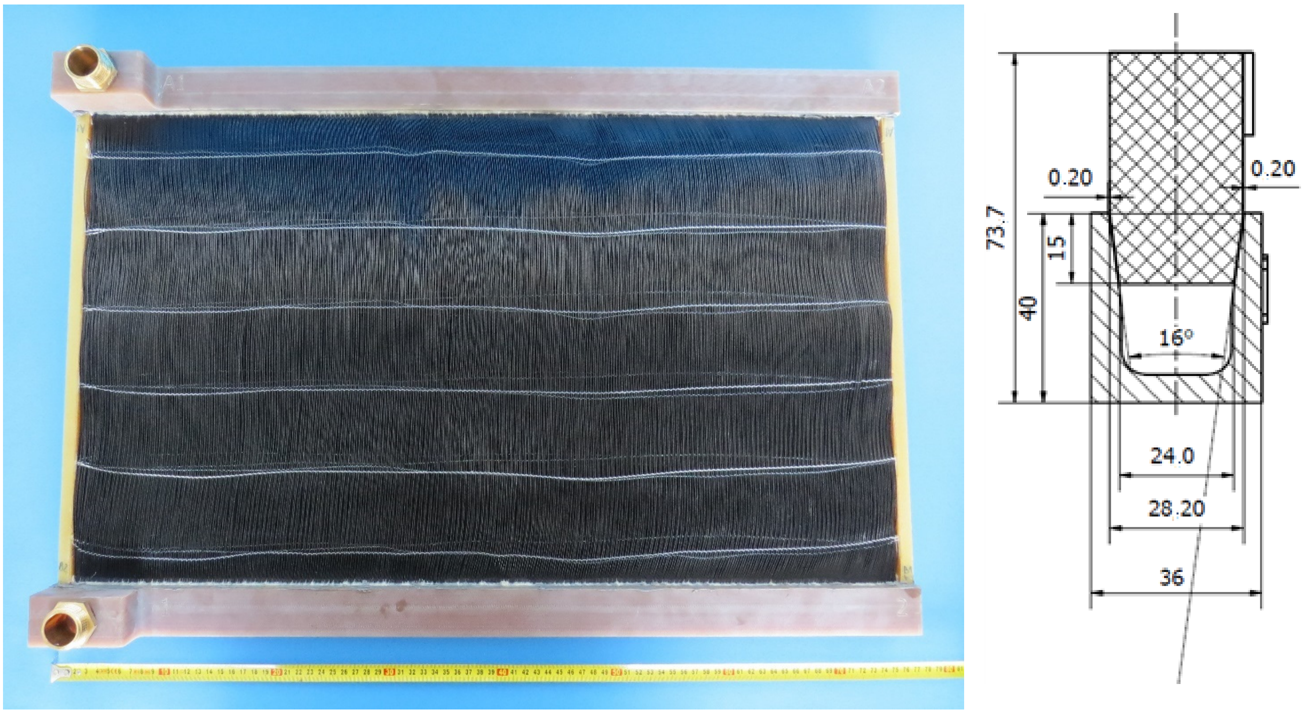
1. Introduction

In recent decades, engineering thermoplastics and fiber-enforced plastic composites have been used more and more for a variety of applications, such as automotive, aerospace, and electrotechnical equipment. These materials successfully compete with metals (mainly aluminum alloys) because of their lightness, good mechanical and processing properties, and their resistance to environmental conditions and corrosion. Polymeric hollow-fiber heat exchangers are a new type of heat exchanger which can compete with metal ones in applications where low weight and corrosion resistance are important. Currently, they are being tested as a heat exchanger in many applications including automotive [1,2] and Li-ion battery thermal management [3,4]. To achieve high thermal performance, liquid-to-gas plastic heat exchangers should be made of a large number of hollow fibers to ensure both low tube wall thickness and high air-side transfer area [5]. Figure 1 shows a polymeric fiber radiator made of a total of 12,240 tubes (34 layers of 360 tubes). Connection of such a number of microtubes using classical methods like thermoplastic overmoulding or welding is very complicated or impossible. That is the reason alternative thermoset materials, in our case epoxides, should be used. On the other hand, the epoxides cannot be connected by welding (because there is no melting, as there is for thermoplastics), so the associated connection should be made through bonding.

Adhesive bonding creates permanent inseparable joints by applying and solidifying the adhesive layer between the parts. Adhesive bonding is a very effective type of connection because it is very simple and has a very good strength to weight ratio. Also, this type of connection does not distort the components being joined and does not create stress. The benefits of adhesive bonding are very important for automotive and aerospace, where there is a strong trend to use lightweight materials and joints [6]. On the other hand, adhesive bonding has several limitations. One of the main disadvantages is that the properties of bonded joints are affected by varying temperature and environmental conditions. Also, the proper solidification of some adhesives requires heat curing and can be a complication for the assembly process. Moreover, adhesive joints are considered inherently weak under peel loading [7]. The influence of temperature on the bonded connection is also associated with potential differences in the thermal expansion of materials and the adhesive layer because it creates additional stress in the joint area when the temperature varies [8]. Moreover, the majority of adhesives are polymer-based and show some degradation when exposed to high temperatures. The other main defining property of adhesives for high-temperature applications is the glass transition temperature, Tg. At temperatures below Tg, the material is brittle with high modulus and strength. Above Tg, the material softens, and the adhesive is more elastic and tougher, but with limited mechanical strength. Therefore, a high Tg is strongly required for high-temperature adhesives [9].



(a) (b)

Figure 1. (**a**) Plastic heat exchanger with coolant manifolds made of glass fabric enforced epoxy G11 EPGC308, adopted from [1]; (**b**) diagram of the bonding connection applied to glue the heat exchanger core to the manifolds.

Bonding joints can fail adhesively or cohesively. Adhesive failure is interfacial failure on the surface between the adhesive and the substrate. Cohesive failure occurs when a fracture is inside of the adhesive layer allowing adhesive to remain on both parts of the substrate. In some cases, the substrate fails prior to the adhesive, and it is known as a cohesive failure of the substrate. Figure 2 shows the failure patterns of adhesive and cohesive joint failures due to EN ISO 10365:1992 [10]. In analyzing an adhesive joint that has been destruction tested, the mode of failure is often expressed as a percentage of cohesive or adhesive failures.

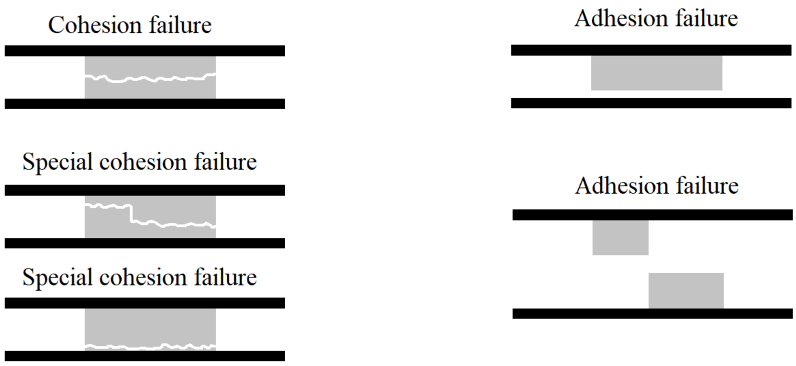


Figure 2. Adhesive and cohesive joint failures acc. to EN ISO 10365:1992. (The figure was modified according to [10]).

On the one hand, cohesive failure within the adhesive or in the substrate is the preferred type of failure because with this type, the maximum strength of the materials in the joint has been reached. On the other hand, the cohesive failure found by an elevated temperature test may indicate that the adhesive lost its strength and is not suitable for this temperature.

Vice versa, adhesive failure means that the adhesive inner strength is higher than the forces between the adhesive and the substrate. There are several different mechanisms of the adhesion, but the main ones are the following. The first is the flow of the adhesive into the microstructure of the substrate providing mechanical interlocking between the substrate and the adhesive. The second is the physical bonding between the substrate and the adhesive due to Van der Waals forces. Typically, the Van der Waals forces do not contribute much to the overall adhesion force. Finally, the third is the chemical bonding, which includes covalent, ionic and metallic connections, which are typically much stronger than the physical bonds. However, the chemical bonding between the different materials is more complicated and the number of the available bonding sites is limited.

Thus, due to its complex nature, the adhesion forces are very dependent on many factors: chemical composition of adhesive and substrate, substrate microstructure, roughness, surface energy, wettability and cleanness. The most effective way to improve the adhesive strength is the pre-treatment and preparation of the surfaces being bonded. One of the widely used approaches to improve the chemical bonding is use of plasma to clean and activate the surface for bonding by creating additional bonding sites.

Automotive high temperature radiators (see Figure 1a) undergo various temperatures and pressure conditions during their service lives, so appropriate strength of bonding must be ensured. Automotive companies typically require the radiator to withstand internal pressure of 2.5 bar at a maximal operation temperature of 125 °C. Considering the safety factor equal to 3 between the operating and burst pressure, we consider the required burst pressure as 7.5 bar (0.75 MPa). The shear strength required to withstand this pressure can be calculated as:

$$\sigma \approx 0.75\ \text{MPa} \times 24/15 = 1.2\ \text{MPa}$$

where 24 mm is the width of the surface under pressure and 15 mm is the height of the bonding overlap. Thus, shear stress at least of 1.2 MPa will be required to consider the adhesive able to withstand the forces due to pressure presence.

The strength of adhesive bonding is also very sensitive to the stress direction. Typically, adhesive connection resists the tensile and shear stress well. On the other hand, the bonding is very sensitive to piling as it causes stress concentration in a small area. In this case, the bonding is firstly broken on one side of the bonding area, and the failure then proceeds through the area destroying the whole bonding. To prevent piling the right design of the bonding connection should be applied. Figure 1b shows a drawing of the bonding connection applied to glue the heat exchanger core inside of the manifold. The bonding is designed in such a way that there is no adhesive piling, the tensile stresses are small, and the dominant stress is the shear stress. Therefore, we experimentally measured the shear strength as the main parameter to estimate the bonding resistance.

We can conclude that determination of the joint's ultimate strength at an elevated temperature is crucial to predicting joint durability even if the durability at an elevated temperature may be much lower than in its normal working conditions. The goals of this study are as follows:

- To experimentally study a wide range of adhesives and determine their resistance to high temperatures;
- For particular plastic substrates, to determine the best adhesive for high temperatures;
- To estimate the influence of the cold plasma surface treatment;
- To choose and recommend successful adhesives.

2. Experimental Section

2.1. Tested Plastic Materials

Glass epoxy sheet G11 180 °C, EPGC308 type according to EN-60893. Epoxy sheets are made from modified glass fabric and epoxy resin with the addition of filler. They are characterized by their excellent mechanical and electrical insulating properties, which are retained even at higher temperatures, and by their resistance to cracking. They are used in the production of components with good mechanical and electrical properties, for electrical equipment at higher temperatures or in humid environments, and for stressed electrical insulating components, such as chassis, body equipment, housing parts of distribution boards, and transformers [11].

ERTALON Polyamide 66 is filled with 30% glass fibers. Compared with virgin PA 66, this 30% glass fiber reinforced and heat stabilized nylon grade offers increased strength, stiffness, creep resistance and dimensional stability while retaining excellent wear resistance and high service temperatures [12].

TECATRON-GF40 black is a 40% glass fiber-reinforced PPS. Because of the high level of glass fiber reinforcement, TECATRON-GF40 black demonstrates excellent rigidity and very high strength at high temperatures compared to unmodified PPS (TECATRON natural). High thermal stability, high dimensional stability, very good chemical resistance, good electrical and thermal insulation and good resistance to sterilization round off the profile of this modified PPS. These material properties make glass-filled PPS suitable for use in parts that are exposed to high static loads over long periods in high temperatures. However, the inherent chemical structure along with the addition of glass fibers tends to create toughness issues with this material. As glass fibers tend in some cases to have a marked abrasive effect on mating surfaces, this PPS composite may be less suitable for sliding applications [13]. Table 1 reviews the mechanical properties of the described materials.

Table 1. Mechanical properties of the tested plastic materials.

Material Used	Glass Epoxy Sheet G11	PA66-GF30	PPS-GF40
Resin and reinforcement	epoxide + glass fabric	PA66 + glass fibers	PPS + glass fibers
Flexural modulus, MPa	26,000	-	4600
Tensile strength, MPa	400	85	83
Notch impact strength parallel to lamination (Charpy), KJ/m^2	100	6	24
Shearing strength parallel to laminations, MPa	50	-	-
Density, g/cm^3	1.9	1.29	1.63
Temperature index (TI) or Temperature of Deflection, °C	180	150	230
Water absorption	(3 mm thick) 9 mg	56 mg/96 h according to ISO 62	0.01%/96 h according to ISO 62

2.2. Tested Adhesives

An overview of the tested adhesives is shown in Table 2 together with a brief description of each. The adhesives tested were selected based on two main requirements, high glass transition temperature Tg and the flexibility to withstand dimensional change due to thermal expansion. All the tested adhesives were two-component epoxies intended for bonding or encapsulating.

Table 2. Review of tested adhesives.

Manufacturer	Brand Name	Type	Tg, °C	Operation Temp. Range, °C	Brief Description and Reference
Reltek	Bondit	B-46	-	−65–135	An adhesive for bonding dissimilar materials, such as plastics, including UHMW, HDPE, PP, PET, PEEK, PPS, PBT, Acetal, ETFE, PVC, PVCF, PVDF, ABS, ECTFE, polyamide, polyimide, rubber, and urethane compounds on metal, glass, composites, cement, wood, and cellulose [14].
		B-481	-	up to 170	
		B-536	-	-	
Elantas	Elan-tech	AS60/AW60	135	up to 150	Excellent thermal and chemical resistance, high modulus, rigid, very good for magnets.
		ASM 031			Ferrite, synthesized materials, magnets, thermally resistant materials [15].
	Epoxylite	E8628	127	up to 204	E 8628 Hi Temp-two-component, low viscosity compound for service temperature up to 200 °C. For potting of collector rings, slip rings and field coils where rigidity and dimensional stability are required [16].
GRM		LG700/HG700	-	up to 120	Laminating system for RIM, RTM, vacuum technology, and hand laminating. Extremely low viscosity enables processing of very light laminates. High reactivity. Working time from 25 min to 3 h. Very good heat resistance and high flexibility even when post-cured at room temperature [17].
Epoxies		20-3001 NC	-	−50 to +150	20-3001 NC is a low viscosity, unfilled epoxy potting and encapsulating system which forms a bubble-free glass-like finish when cured [18].
		20-3302 NC	-	−45 to +135	20-3302 is a low viscosity system designed for L.E.D. encapsulating, fiber optics, and any potting or adhesive application requiring optimum clarity. This epoxy system also provides a bubble-free and glass-smooth finish [19].
Girrbach	Gimapox	EL6	160	up to 160	Epoxy resin system has a glass transition temperature of up to 160 °C and very high thermal stability. The material has good chemical resistance [20].
Havel		LH 301	-	up to 160	Epoxy resin LH 301 is a resin which makes it possible to achieve a temperature resistance of about 160 °C with a hardener H 513. When curing amines or polyamines, it offers high mechanical properties, and good chemical and thermal resistance. Ratio LH 301:H 513 = 100:22 The product must be cured at temperatures of min. 50 °C for 4 h, and to achieve maximum temperature resistance, it is then appropriate to harden it by gradually heating it to 160 °C [21].
5 M	Letoxit	LH 190	-	up to 150	This two-component epoxy adhesive is composed of part A and part B and hardens at a higher temperature. The adhesive is intended for bonding of metals and applications with high temperature resistance [22].
Henkel	Loctite	EA 9483	61	-	EA 9483 is a 2-part, ultra-clear, flowing, general purpose epoxy adhesive suitable for bonding and potting where optical clarity and high strength are required [23].
		EA 9497	116	-	LOCTITE® (Henkel AG, Düsseldorf, Germany) EA 9497 is a 2-part, grey epoxy adhesive for structural bonding that offers high technical performance. The product provides high temperature resistance up to +180 °C for heat-resistant bonding and filling. It is thermally conductive and delivers high compression strength. It is ideal for bonding metal components [24].

2.3. Preparation of Samples

The samples were made by gluing the desks with a slip surface of 12.5 × 24.5 mm. For the glass epoxy sheet an additional 33 × 24.5 mm square was bonded in order to have the jaws of the tensile test machine on one plane (see Figure 3 for details). For PA and PPS, samples were made by CNC milling. Such shapes of samples were used to prevent torsion in the specimen and ensure one-dimension loading (shear stress) of the slip surface (see Figure 4b). Bonding surfaces were treated with care before the application of the adhesive. Epoxy sheet samples were cleaned and sandpapered slightly (size 200 paper) because the surface was very smooth. The milled samples (PA and PPS) were not sandpapered but were ungreased to remove possible oil impurities from the tools. For PA and PPS, an additional bunch of samples was prepared and treated with cold plasma because it represents an efficient, clean and economical alternative for activating the surfaces of thermoplastics [25]. The Relyon PZ2 hand plasma Piezo brush [26] was applied closely before the application of the adhesive (for up to 20 min) because the ageing of the plasma-treated surface reduces the plasma effect.

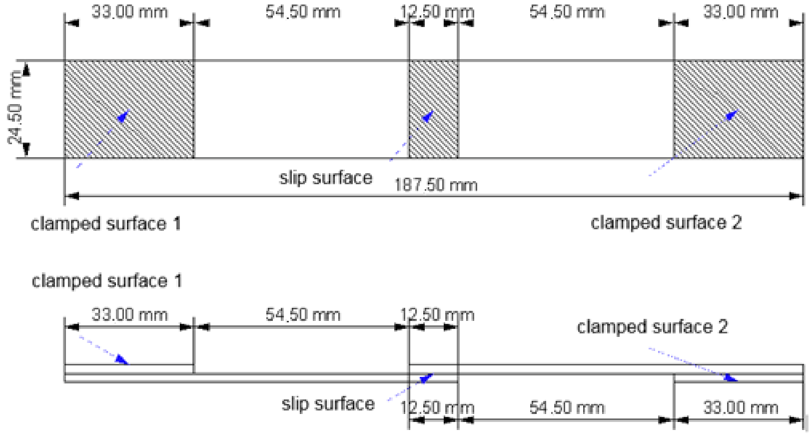
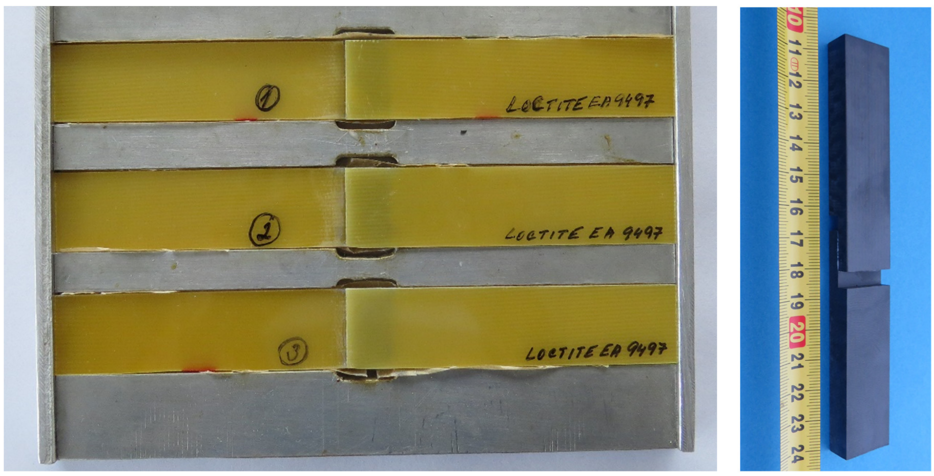


Figure 3. The size of the tested samples made of G11 glass epoxy sheets.



(**a**) (**b**)

Figure 4. (**a**) Aluminum frame with glass epoxy samples; (**b**) milled PA66-GF30 sample.

The adhesives were prepared according to the manufacturer's manuals and applied with the recommended thicknesses. The application methods varied depending on the viscosity of the particular adhesive. The low-viscosity adhesives were applied with a dropper so that the entire shear surface was covered with a continuous layer of adhesive. The overflows of the adhesive in the joints were wiped off with a cotton swab. The samples were then fixed in the cells in the aluminum plate (see Figure 4a), and the bonded joints were loaded with weights. The samples were fixed and loaded (up to 1.5 kg, 50 kPa pressure) this way in order for the adhesive to be cured according to the manufacturers' recommendations. After adhesive curing, the end faces of the joints were roughened with sandpaper (size 200) to prevent them slipping in the clamping jaws of the tensile machine. Three samples were prepared and tested for each pair of the substrate-adhesive.

2.4. Testing Method

After curing, the samples were tested for tensile shear strength in accordance with ČSN EN 1465 (Czech version of BS EN 1465: 2009-Adhesives. Determination of tensile lap-shear strength of bonded assemblies). ISO 4587 provides a standard method to determine the lap-shear strength of a bond between two rigid test specimens. Each specimen should be 100 × 25 × 1.6 mm. These are bonded with an overlap of 12.5 mm and clamped over a 37.5 mm length at each end. The force required to pull the specimens apart is then recorded. ISO 4587 is used for quality control in many industries, including aerospace, automotive, and consumer goods manufacturing. Each sample was clamped in the jaws of the tensile machine (Z010 TE, Zwick & Roell, Ulm, Germany) in a closed heated temperature chamber (see Figure 5) [27]. The chamber was heated to the required temperature and the sample was tempered for 5 min prior to testing. After tempering, the machine was started and the shear stress was measured at a speed of 10 mm/min. The slip surfaces of the broken samples were photodocumented to evaluate the type of failure.

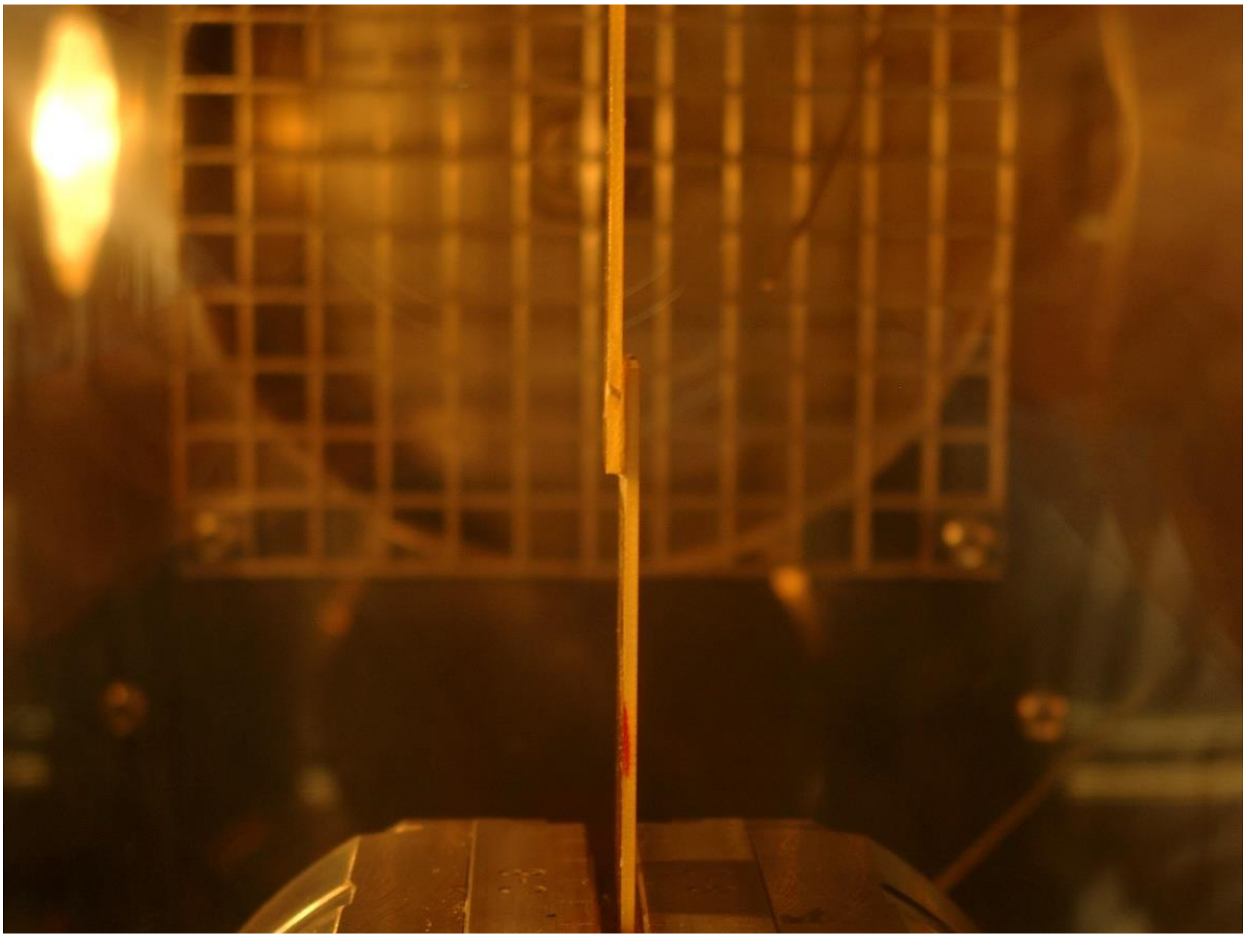


Figure 5. Epoxy sheet sample fixed in the clamping jaws of the heated chamber of the tensile machine Z010 TE, Zwick & Roell.

3. Results and Discussion

3.1. Glass Reinforced Epoxy Sheets G11, 130 °C and 160 °C

Epoxy sheet material is a good base material to test the gluing properties as it has high mechanical strength and thermal resistance, good adhesion properties, and dimensional stability (low thermal expansion due to reinforcement with glass fabric). The first round of testing at 130 °C showed that many adhesives do not withstand the temperature (cohesive failures), so testing at higher temperatures was continued only with successful ones.

At 130 °C, the LH 301 adhesive joint had the highest strength, while the strength of the Bondit B-46 adhesive was the lowest (see Table 3 for results). For the Bondit B-46 adhesive, two of the three bonded joints were broken before starting the test during conditioning. The same was true for the joint bonded with 20-3302 NC. One of the reasons could be the effect of a warm air stream causing some vibration of the sample. LH 301 and AS60/AW60, the two strongest joints, showed a slight slip of the specimens in the jaws during the test. Here, it seems that the joint will not come apart spontaneously unless it is mechanically stressed. For the LH 301 adhesive sample, it is clear (from the pealed-off shear surfaces) that the adhesive joint only came loose with the destruction of the fiberglass board material (casting resin). The casting resin was literally torn down to a layer of glass fabric. The difference in strength between the best and worst adhesive is one order of magnitude. The four best adhesives showed a strength in the range of 2.9–2.6 MPa, with another adhesive (LG700/HG700) experiencing a decrease in strength of almost half.

Table 3. Overview of tested adhesives, curing temperatures, and shear strength tested with glass epoxy sheet G11.

Adhesive	Adhesive Amount (mg)	Curing Conditions (h)	(°C)	Shear Strength at 130 °C (MPa)	Shear Strength at 160 °C	Failure Type at 160 °C
B-46	5.3	4	93	0.29 ± 0		
B-481	6.7	3	93	1.27 ± 0.13	1.18 ± 0.21	A */100%
B-536	13.9	1.5	93	1.45 ± 0.22		
AS60/AW60	48.4	15 3	23 130	2.83 ± 0.18	1.59 ± 0.23	A */100%
ASM 031	10.3	2 4	120 140	0.73 ± 0.13		
E8628	–	4	177	0.53 ± 0.23		
LG700/HG700	5.6	2	120	1.52 ± 0.06		
20-3001 NC	9.6	16	25	0.94 ± 0.05		
20-3302 NC	9.9	2	52	0.64 ± 0		
EL6	3.9	24 2	25 120	1.00 ± 0.43		
LH 301	27.3	–	–	2.94 ± 0.05	1.21 ± 0.17	A */90%
LH 190	67.5	24 0.5 1 3	22 60 100 120	2.75 ± 0.18	1.63 ± 0.18	A */100%
EA 9483	16.0	168	22	1.36 ± 0.1		
EA 9497	54.8	24 0.25	22 120	2.59 ± 0.04	1.42 ± 0.34	A */90%

* adhesive fracture.–failure at the adhesive-bonded material interface.

In addition to the shear strength values, the way the joints fracture also provides important information about the joint quality. In the specimens examined, four typical fracture types were observed: cohesive fracture, adhesive fracture, fracture at the material layer and mixed fracture. The material layer failure is characterized by damage to the base material-fibers or particles are torn from the surface. Adhesive failure occurs on the border between the base material and the adhesive (all the adhesive remains on one of the adherents). Cohesive failure occurs when the adhesive loses its strength and breaks (particles of the adhesive remain on both adherents).

Tests at a temperature of 130 °C show that only 5 adhesives withstand the increased temperature without significant softening and loss of strength, thus showing that it is an adhesive failure. The other adhesives did not withstand the temperature and the bonded connections were broken cohesively. The best adhesives were also tested at 160 °C and the best result was provided by LH 190, while AS60/AW60 also displayed very good strength. LH 109 and AS60/AW60 can be recommended for gluing glass-laminate. Figure 6 shows the slip surfaces of the samples with evident type of the failure, adhesive type. Out of all tested adhesives the best-performing ones were two-component epoxy resins applied on epoxy glass sheet substrate and cured at elevated temperate. Due to this fact, some chemical bonding can occur regardless of the fact that the epoxy of substrate was fully cured long before the application of adhesive epoxies. Nevertheless, with temperature increasing to 160 °C, the forces of chemical bonding were reduced, and internal stresses due to different thermal expansion occurred (substrate epoxy sheets have lower thermal expansion coefficient due to presence of glass fabric). These factors together with reduction of strength of adhesives themselves have caused the adhesive failures of the tested samples. Results of shear strength at elevated temperatures are significantly lower than those at normal conditions, showing that the temperature factor is crucial. For example, the Henkel Loctite bonding guide [28] refers to lap shear strength of epoxy to epoxy bonding of up to 21 MPa (for Loctite 4307 adhesive). In this case, the bonding strength is higher than the substrate strength. Nevertheless, the effect of elevated temperature is not reflected as tests were performed for normal conditions. There were no data available for epoxy-to-epoxy bonding tested at elevated temperature of 130 and 160 °C.

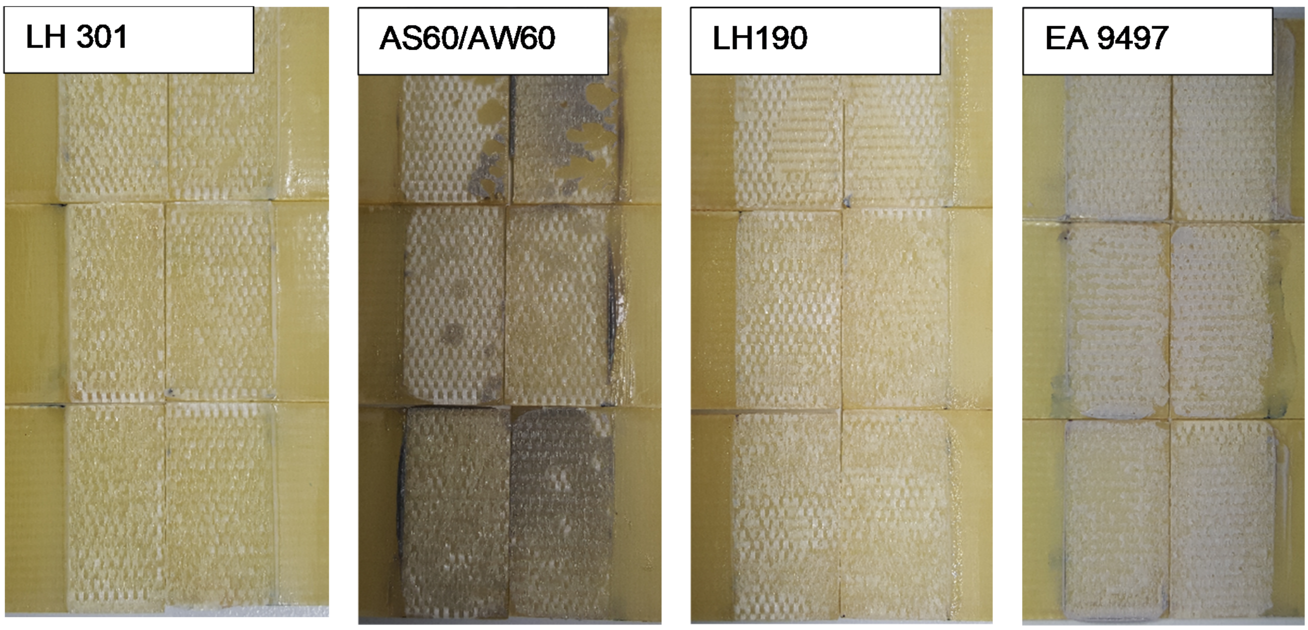


Figure 6. Fracture surfaces of the 4 best-performing adhesives—130 °C test. The slip surfaces of all the adhesives show the presence of the adhesive on both sides of the samples which can be interpreted as cohesive failure. However, in all cases, the majority of the adhesive layers were on one side, showing the nature of the adhesive failure nature.

3.2. *PA66-GF30, 110 °C and 160 °C*

Unlike epoxy glass-enforced sheets, the polyamides are difficult to bond because they are hydrophobic, chemically inert and possess poor surface wettability. All these factors make proper pre-treatment very important, including cleaning, removing hydrocarbon contaminates, roughening with sandpaper and plasma surface treatment. All of those were used to ensure reasonable adhesion for elevated temperatures.

The strength of the bonded joints is given in Table 4, together with the evaluation of the type of failure. The letter A indicates an adhesive failure (failure at the adhesive-bonded material interface), and the letter C indicates a significant cohesive failure in the adhesive (adhesive remained on both surfaces of the bonded joint). An image of the three broken joints after the test at 110 °C is shown in Figure 7. However, as the surface of the joint was textured and the thickness of the adhesive was different over the overlap area, it was not easy to assess exactly whether A or C was broken, as a thin layer of adhesive appeared to remain on many surfaces of both bonded joints, although most tore off the surface.

Table 4. Shear strength of joints of PA66-GF30 at 110 and 160 °C and evaluation of the type of failure (A denotes adhesive failure, and C cohesive failure in the adhesive).

Adhesive	EA 9497			LH 190			AS60/AW60			B-481		
Surface treatment	no	plasma		no	plasma		no	plasma		no	plasma	
Temp. °C	110		160	110		160	110		160	110		160
Shear strength [MPa]	0.58 ± 0.27	1.76 ± 0.21	0.37 ± 0.20	1.66 ± 0.15	1.91 ± 0.41	0.76 ± 0.33	1.69 ± 0.46	2.15 ± 0.34	1.64 ± 0.32	0.94 ± 0.11	1.00 ± 0.12	0.43 ± 0.10
Failure type	A 100%	A 95%	A 85%	A 95%	A 90%	C 20%	A 95%	A 95%	A 70%	A 90%	A 95%	C 50%

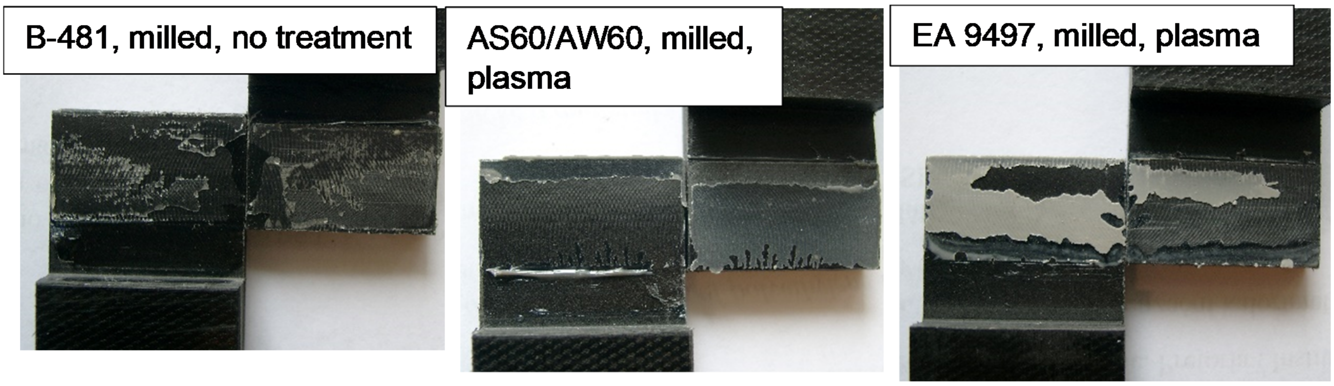


Figure 7. Photos of PA66-GF30 samples fracture, tests at 110 °C.

Plasma surface treatment increased the bond strength of all adhesives except B-481; by almost three times for EA 9497 and by 25% for AS60/AW60. This joint also showed the highest strength of all tested.

B-481 adhesive appears to be completely unsatisfactory for PA66GF30 because plasma surface activation did not increase the reactivity of the adhesive. Loctite EA 9497 is also unsatisfactory without plasma surface activation. The strongest bond was given by LH 190 and AS60/AW60 adhesives after plasma activation of the bonded surface, but the strengths without plasma activation are also high.

For gluing the PA66-GF30, LH 109 and AS60/AW90 can be recommended, preferably after activation of the connecting surface by plasma.

Regarding testing at an elevated temperature of 160 °C, the results are different. Only adhesive AS60/AW60 shows a strength higher than 1 MPa, specifically 1.64 MPa. EA 9497 and AS60/AW60 adhesives were broken adhesively, which is clearly seen in Figure 8 in the milky color of the adhesive (Table 4 shows the percentage of the adhesive remaining on one side of the bonded joint). For adhesives B-481 and LH 190, the failure was partly cohesive and partly adhesive (see Figure 8).

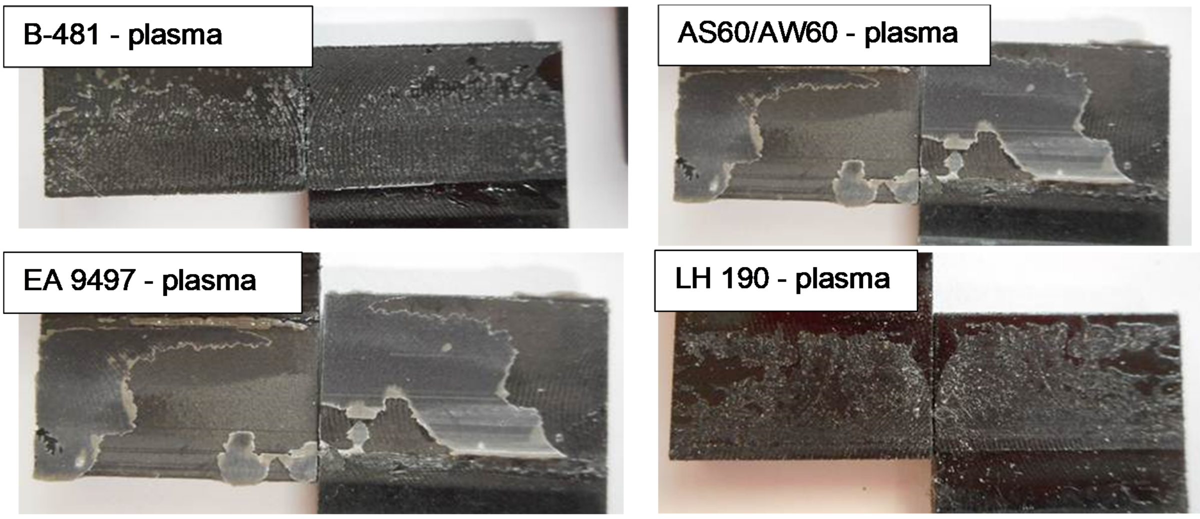


Figure 8. Photos of PA66-GF30 samples fracture, tests at 160 °C.

Thus, at 160 °C, AS60/AW60 showed the highest strength, 1.64 MPa, and the second-best LH 190 showed half the strength—0.76 MPa; the joints bonded with B-481 and Loctite EA 9497 have even lower strength.

The achieved values of shear strength can be seen as low but considering the temperature factor and compared to the results of shear strength at normal temperature, we can declare them as reasonable. According to the Masterbond company [29], lap shear strength of polyamides bonded with 7 different epoxies (cleaning, degreasing and mechanical abrasion treatment) were 2.19–4.14 MPa (317–600 psi) at room temperatures.

Lutey et al. [30] have tested the polyamide 66 bonding with two-component polyurethane Terosol PU 9225. Testing at room temperatures showed average shear strength of 2.68 MPa for untreated polyamide, 4.60 MPa for mechanical abrasion treatment and 8.12 MPa for cold plasma treatment (Plasma Beam by Diener). Plasma treatment of polyamide prior to bonding was also studied by Karoly et al. [31]. Three commercial adhesives were tested: cyanoacrylate (Loctite 406), acrylic based adhesive (Loctite 3035, two-component glue), and two-component epoxy adhesive (Loctite 9466), all from the Henkel AG, Germany. Bonding with epoxy adhesive Loctite 9466 showed the shear strength of 0.8 MPa and 1.4 MPa for untreated and plasma-treated samples, respectively. The most successful cyanoacrylate adhesive Loctite 406 showed 3.8 MPa and 3.9 MPa for untreated and plasma-treated samples, respectively. These studies show similar results of the lap shear strength and confirms importance of plasma pre-treatment.

3.3. PPS-GF40, 130 °C and 180 °C

Polyphenylene sulfide (PPS) is a high-temperature, semi-crystalline engineering thermoplastic. It is chemically inert and has low surface energy, offering a wide resistance to chemicals. While these characteristics are ideal for material performance, its poor surface wettability is the bonding challenge. For products made of PPS, surface cleanliness and plasma pretreatment are critical requirements to achieving high values of bond strength.

Results of the shear strength test for PPS material are given in Table 5. It can be said that PPS joints were broken mainly adhesively. Only two tests showed cohesive failure, namely adhesives B-481 and LH-190 in test with 130 °C (see Figure 9) A very thin layer of adhesive with the same pattern can be observed on both torn surfaces of the samples. Table 5 shows the percentage of slip surface which was torn off/remained on one of the parts of the bonded joint. Figure 9 shows the slip surface of the broken samples; it can be seen that some samples have areas which do not have enough adhesive (marked as A). This area was used to correct the values of the slip surface used to calculate the shear strength.

Table 5. Shear strength of joints subjected to tensile stress at 130 °C and 180 °C and evaluation of the type of failure (A denotes adhesive failure, and C cohesive failure in the adhesive).

Adhesive	**AS60/AW60**			**LH 190**		**EA 9497**		**B-481**	
Surface treatment	no	plasma		plasma		plasma		plasma	
Temperature [°C]	130		180	130	180	130	180	130	180
Shear strength [MPa]	1.77 ± 0.06	2.39 ± 0.15	1.38 ± 0.09	1.79 ± 0.15	0.01	1.01 ± 0.25	0.22 ± 0.09	0.83 ± 0.20	0.40
Torn off adhesive area on one side of the joint	99%	70%	95%	90%	70%	70%	100%	70%	100%
Failure type	A	A	A	C	A	A	A	C	A

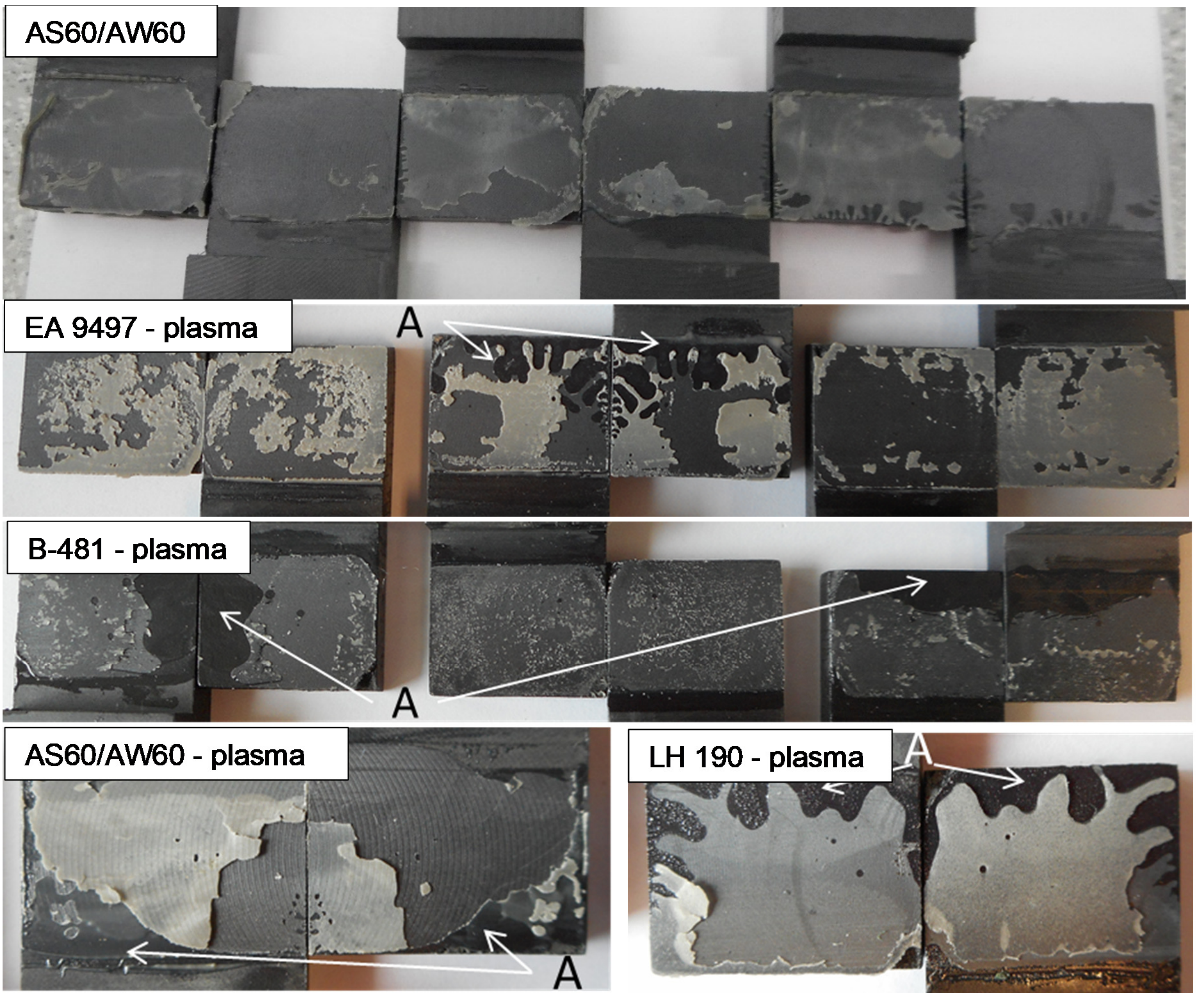


Figure 9. Images of broken joints tested at 130 °C for each type of adhesive. Areas which do not have enough adhesive are marked as A.

At 130 °C, AS60/AW60 adhesive had the highest strength, LH 190 adhesive showed 25% lower strength, EA 9497 adhesive connections had almost 60% lower strength and B-481 adhesive connections showed the lowest strength (65% lower strength in comparison with AS60/AW60).

At 130 °C, the average strength of the joints is given in Table 5 together with the percentage of the adhesive failure. For the well-prepared adhesive joint, the adhesive breakage occurred at the adhesive-composite part interface. The plasma surface treatment has a fundamentally positive effect on the shear strength of the adhesive joint, whilst the increasing of the temperature from 130 to 180 °C had a negative effect.

On the PPS-40GF at 180 °C, the adhesive AS60/AW60 is the only one that showed a reasonably high strength of 1.38 MPa. The other adhesives did not withstand the temperature of 180 °C and were broken at a very low loading. Letoxit LH190 adhesive is completely unsuitable for bonding PPS-40GF composite and use at 180 °C, despite the plasma activation of the surfaces of the bonded sheets (see Figure 10 for failure details).

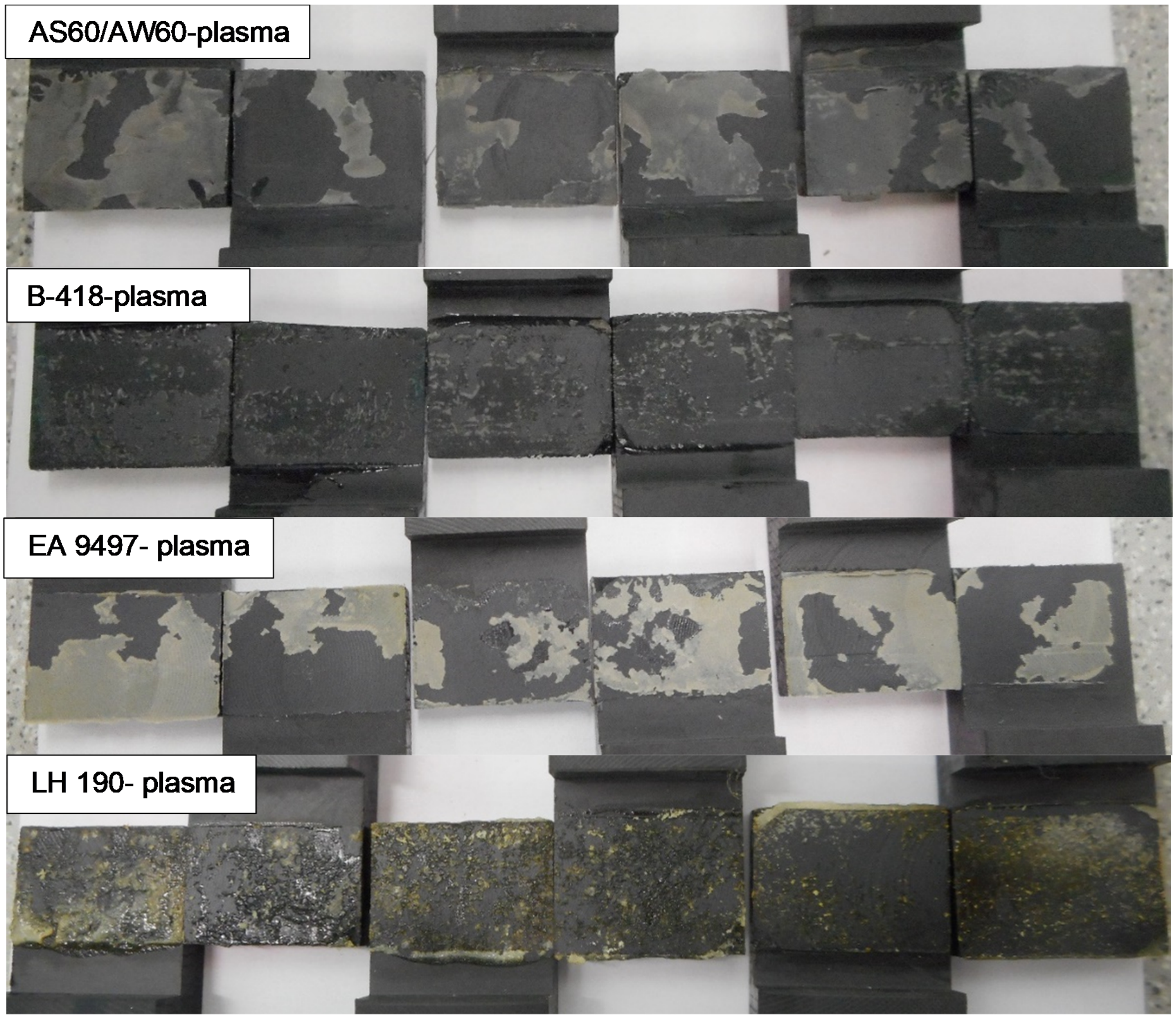


Figure 10. Images of broken adhesive joints of PPS-40GF composites tested at 180 °C.

There are little data available on PPS bonding with epoxies. Henkel Design guide declares the shear strength of PPS bonded by different epoxies in range of 1.0–7.2 MPa at normal conditions [28]. Considering the results achieved (max. strength 1.38 MPa) at the testing temperatures 180 °C, we can conclude that temperature factor is crucial in strength reduction.

4. Conclusions

14 commercially available adhesives intended for elevated temperatures were tested for gluing glass fabric epoxy composite G11, polyamide PA66-GF30 and polyphenylene sulfide PPS-GF40. Bonded connections were tested with a tensile machine with a calorimetric chamber to determine the shear strength for a particular temperature. Testing showed that only 4 of the 14 adhesives tested show substantial bonding strength at temperatures above 120 °C and only one resists at 180 °C.

For the G11 epoxy glass sheets, two adhesives can be recommended: AS60/AW60 and LH 190. Both adhesives show similar shear strength values of about 2.8 MPa and 1.6 MPa for 130 and 160 °C respectively.

For the PA66-GF30, there is a very significant difference between the results at 110 and 160 °C. While at 110 °C, three adhesives (AS60/AW60, LH 190 and EA 9497) show a shear strength of about 1.5–2.0 MPa, at 160 °C only AS60/AW60 can be used (1.6 MPa shear strength). Use of cold plasma is strongly recommended for PA66-GF30, because it increases the bonding strength by about 30%.

For the PPS-GF40, similar behavior as for PA66-GF30 was found. While at 130 °C, three adhesives achieve reasonable strength (AS60/AW60, LH 190 and EA 9497), at 180 °C only AS60/AW60 can be used (1.4 MPa shear strength). Use of the cold plasma is also very strongly recommended to activate the surface since it improves the resulting strength by about 35%. In general, we can conclude that the adhesive AS60/AW60 from Elan-tech showed the best results for the high-temperature gluing as the only adhesive which resists a temperature of 180 °C.

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