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Effect of the Preparation of Aluminium, Magnesium and Titanium Alloys Surface on Properties of Adhesive Bonded Joints

Abstract: The article presents issues related to the joining of hard-to-join metals such as aluminium, magnesium and titanium alloys as well as discusses problems accompanying the joining of the above-named materials and indicates the possible use of adhesive bonding when joining them. The article pays particular attention to appropriate surface preparation for adhesive bonding as a factor determining the proper functionality of joints. In addition, the article presents metallographic tests and results of static shearing tests of adhesive bonded joints in relation to the manner of surface preparation, i.e. grinding, abrasive blasting and low-temperature plasma treatment and demonstrates the significant effect of surface preparation using low-temperature plasma on the strength of adhesive bonded joints.

Keywords: adhesive bonding, Aluminium, Magnesium, Titanium Alloys

DOI: *10.17729/ebis.2017.5/9*

Introduction

Adhesive bonding is a technological process involving the joining of two or more materials using an adhesive, i.e. a surface-active substance, the primary component of which is a synthetic or natural polymer in the form of a suspensoid. The application of an adhesive film between elements to be joined enables the obtainment of a stable connection, i.e. an adhesive bonded joint [1-3]. An undeniable advantage of adhesive bonding is the possibility of joining nearly all structural materials, e.g. aluminium, magnesium or titanium alloys characterised by good mechanical properties yet difficult to join using brazing, fusion welding or pressure welding. The quality and service life of adhesive bonded joints is affected by many technological and mechanical factors [5], including the type of an adhesive and a method of its application, the shape and dimensions of a joint, the type of a load affecting a joint and the manner of surface preparation before adhesive bonding. The

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aforementioned surface preparation is of great importance in adhesive bonding and significantly affects the properties of adhesive bonded joints. The appropriate preparation of surfaces to be joined should provide proper adhesion, i.e. adhesive force between an adhesive and a material being joined [1-4]. The obtainment of appropriately high adhesion in a joint requires the removal of all impurities from surface layers of elements to be joined. In addition, it is necessary to ensure appropriate surface expansion and activation. In production conditions, the above-named effects are obtained using various chemical and mechanical treatment methods. Grinding and abrasive blasting belong to mechanical methods providing the appropriate expansion and roughness of surfaces enabling the obtainment of appropriately high mechanical adhesion [1-4].

The use of cold atmospheric plasma, also known as low-temperature plasma, is an example of treatment making it possible to increase surface energy and polarity, i.e. factors determining the obtainment of high specific adhesion, and, consequently, adhesive bonded joints characterised by good mechanical properties. Plasma is generated at temperature, where the mean kinetic energy of molecules exceeds the value of an ionisation potential. Usually, plasma is a slightly ionised gas characterised by the high or very high content of neutral particles including atom nuclei, elementary particles as well as atoms, their ions etc. The use of low-temperature plasma makes it possible to increase the surface energy of elements being joined and, as a result, to obtain the good quality of adhesive bonded welds [6]. Presently, the above-named method is one of the most effective technologies enabling the cleaning, coating and activation of plastic surfaces, particularly those of low surface energy such as polypropylene (PP), polyethylene (PE) or polytetrafluoroethylene (PTFE). In comparison with environmentally unfriendly and expensive chemical treatment, the use of low-temperature plasma is relatively easy to use and implement in industrial conditions as well as environmentally friendly. The popularity of the above-named method is particularly visible in the electronic industry (production of mobile phones) and in the automotive industry (cleaning and activation of polymer elements before adhesive bonding) [5, 6].

Test Materials

This article presents the effect of selected preparation methods of aluminium (5754), magnesium (AZ31B) and titanium (Grade 2) alloy surfaces on the shear strength of adhesive bonded joints. Because of the presence of a complex oxide layer on their surface, the above-named materials are difficult to join using traditional joining techniques such as brazing, pressure welding or fusion welding.

Aluminium is a silver-white metal from the third group of the periodic table, characterised by low density (2.71 kg/dm^3), high thermal and electric conductivity as well as high plasticity, and, consequently good treatability and castability. In addition, aluminium is characterised by high corrosion resistance and low specific gravity – a very desirable quality in the production of light structures. Aluminium also has a relatively low melting point (approximately 660°C) and high affinity for oxygen (leading to the formation of a high-melting film of Al_2O_3 oxides on the surface). Because of the foregoing, aluminium and its alloys are rated among materials difficult to join [7]. In its pure form, the metal is not characterised by particularly favourable mechanical properties. Fortunately, these aluminium properties can be significantly improved by adding small amounts of alloying agents, usually iron, silicon, copper, zinc and titanium, which, on one hand decrease aluminium plasticity, whereas on the other increase its mechanical strength and hardness. Certain aluminium alloys are characterised by mechanical properties similar or even more favourable than those of some steel grades. The foregoing combined with low kerb weight make aluminium alloys the most popular structural materials used in nearly every industrial sector. Aluminium alloys EN AW 5754 are characterised by medium tensile strength, high fatigue strength and very high corrosion resistance both in the industrial atmosphere and when exposed to seawater. In turn, magnesium, one of the lightest metals, comes from the second group of the periodic table and is characterised by even lower density than aluminium (1.74 kg/dm^3), high vibration damping ability, good castability and low specific gravity [8]. Because of the fact that, similar to aluminium, magnesium in the pure form is characterised by rather poor mechanical properties and low plasticity (including low toughness, low creep resistance and low tensile strength at higher temperature), its role in structural solutions is rather limited. Magnesium is primarily used in light metal alloys where it is combined with aluminium, manganese, zinc, lithium or rare-earth elements [9]. The low density combined with highly favourable mechanical properties of magnesium alloys make them widely used in the aviation and aerospace industries.

Magnesium alloy AZ31B, i.e. the most popular alloy in terms of workability, is characterised by high corrosion resistance (up to a temperature of 120°C). Usually, this alloy is used in light structures having appropriate rigidity and strength. Because of its very good casting properties, magnesium alloy AZ31B is widely used in the automotive and machine-building industries. Cast structural elements made of this alloy are characterised by high material homogeneity, shape accuracy and complexity as well as high strength and good plasticity [9].

Titanium is a chemical element belonging to the group of transition metals of the periodic table. Titanium is a relatively light metal having a density of 4.51 kg/dm^3. Its colour is grey and metallic, whereas its melting point amounts to 1649°C. In addition, titanium is known for its high corrosion resistance and a very high mechanical strength-specific gravity ratio. In its pure form, titanium is characterised by very good plastic properties, forgeability and treatability, yet even small amounts of impurities (oxygen, nitrogen, carbon, iron, hydrogen and silicon) may significantly reduce the above-presented favourable properties [10-12]. Titanium and its alloys are widely used in the production of sports equipment such as bicycle frames, tennis rackets, skis, golf clubs and helmets, i.e. object which must be characterised by high strength and light weight at the same time.

Titanium (alloy) Grade 2 is commercially pure titanium (containing small contents of oxygen and iron). It is characterised by favourable mechanical properties and good ductility. A thick oxide layer on the surface of titanium provides very high corrosion resistance in oxidising media. Because of this fact, titanium Grade 2 can be used in various environments (exposed to seawater, chlorine, chlorine dioxide, sulphides, nitric acid etc.). The chemical composition and primary mechanical properties of alloys selected for the tests, i.e.: aluminium EN AW 5754, magnesium AZ31B and titanium Grade 2, are presented in Table 1.

Table 1. Chemical composition and primary properties of aluminium 5754, magnesium AZ31B and titanium Grade 2 [13-16]

Designation	Chemical composition, % by weight					Melting point, °C	Density, kg/dm^3	Tensile strength *Rm*, MPa
	Al	Mg	Zn	Ti	others			
EN AW 5754	rest	2.6 ÷ 3.6	0.2	-	0.5 Mn; 0.4 Si; 0.3 Cr	595 ÷ 645	2.67	190 ÷ 200
AZ31B	3.0	rest	1.0	-	0.2 Mn	605 ÷ 630	1.77	193
Ti Grade 2	-	-	-	rest	0.3 Fe; 0.08 C; 0.25 O; 0.03 N	1660	4.51	390 ÷ 540

Testing Methodology

Sheets having a thickness of 2.0 mm, made of aluminium 5754, magnesium AZ31B and titanium Grade 2, were sampled for specimens (25 × 80 mm) used to make overlap adhesive bonded joints subsequently subjected to static shear tests. The dimensions of a single overlap adhesive bonded joint are presented in Figure 1. The thickness of the adhesive bonded weld was kept constant by using steel wires having a diameter of 0.2 mm and acting as spacers.

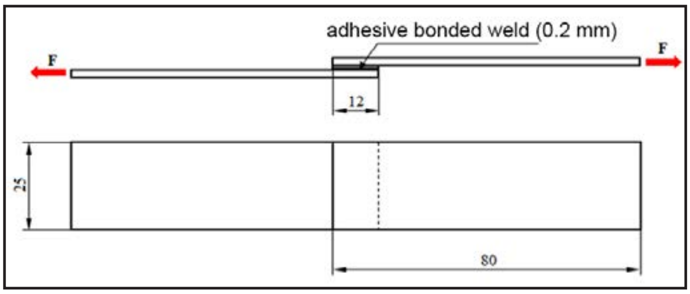

Fig. 1. Schematic overlap joint used in the static shear test

The surfaces of the test specimens were subjected to two types of mechanical treatment, i.e. grinding and abrasive blasting (used to obtain appropriate surface expansion). In addition, the surfaces were subjected to the effect of low-temperature plasma aimed to increase the surface energy of the materials. Each specimen, both before and after mechanical treatment, was cleaned using Loctite 7063, i.e. a single-component CFC-free dissolvent used to clean and degrease surfaces prior to adhesive bonding. Grinding was performed using a fine-grained (red) abrasive cloth, whereas abrasive blasting was performed using corundum having a granularity of 800 μm and fed under a pressure of 6 bars. Six sets of overlap joints (5 for strength tests and 1 for metallographic test) were prepared for each method of surface preparation. The tests also involved the preparation of the same number of overlap joints, yet additionally subjected to the effect of low-pressure plasma (made using argon of purity class 4.5, i.e. 99.995%). The parameters of surface modification performed using low-temperature plasma included a power of 300 W, an operating voltage of 18 kV, a plasma gas (argon) flow rate of 16 dm^3/min and a constant exposition time of 60 s. The principle of operation of the plasma-generating device is presented in Figure 2 [6]. In work [6] it was stated that the use of low-temperature plasma treatment makes it possible to significantly increase surface energy, (by 43%) and the polar variable (by 316%) in relation to the surface of aluminium alloy EN AW 7075.

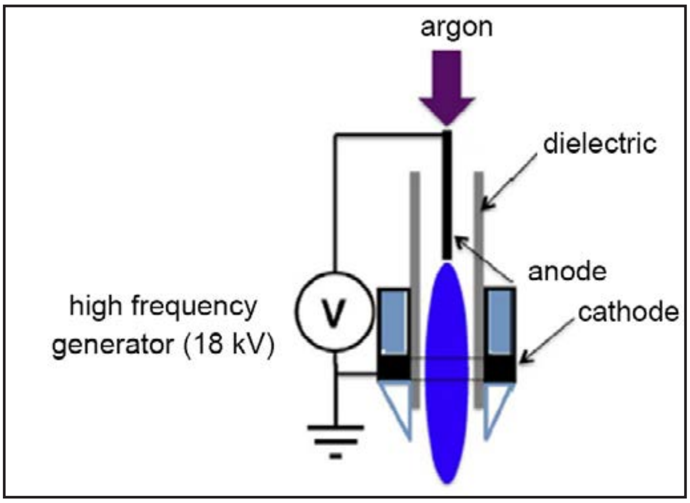

Fig. 2. Principle of operation of the low-temperature plasma generator [6]

The adhesive bonded joints were made using two types of adhesives, i.e. methacrylate resin-based industrial adhesive Agomet F330 and hybrid (cyanoacrylate-methacrylate) adhesive HY 4080 GY (Loctite). The first of the above-named adhesives enables the obtainment of the high tensile-shear strength and the high tear strength of adhesive bonded joints as well as high resistance to a temperature of up to +200°C. The ultimate strength is obtained after 24 hours following the making of a joint. Before application, the adhesive should be appropriately prepared, i.e. provided with an appropriate amount of a hardening agent (3 ÷ 5% by weight) and mixed with the resin. Next, the adhesive should be applied on related surfaces within 4 to 6 minutes ("life time"). The second of the adhesives selected for the tests is a so-called hybrid adhesive as it constitutes the combination of a methacrylate resin-based adhesive and a cyanoacrylate adhesive mixed in a 1-1 ratio 1 using an appropriate static mixer (simultaneously performing the role of a feeder). The combination of the two different adhesive substances aimed to obtain an adhesive providing joints with high mechanical strength, similar to that of joints made using methacrylate resins, and to reduce a time after which a joint obtains its ultimate mechanical strength (in cases of most metals this time is restricted within the range of 18 to 24 hours). Both adhesives are of general application and, among other things, are used to join metals, e.g. unalloyed steels, stainless steels, aluminium, copper, brass, plastics, e.g. hard PCW, ABS, polystyrene, polycarbonate, acrylic glass, parts of polyester moulds etc.

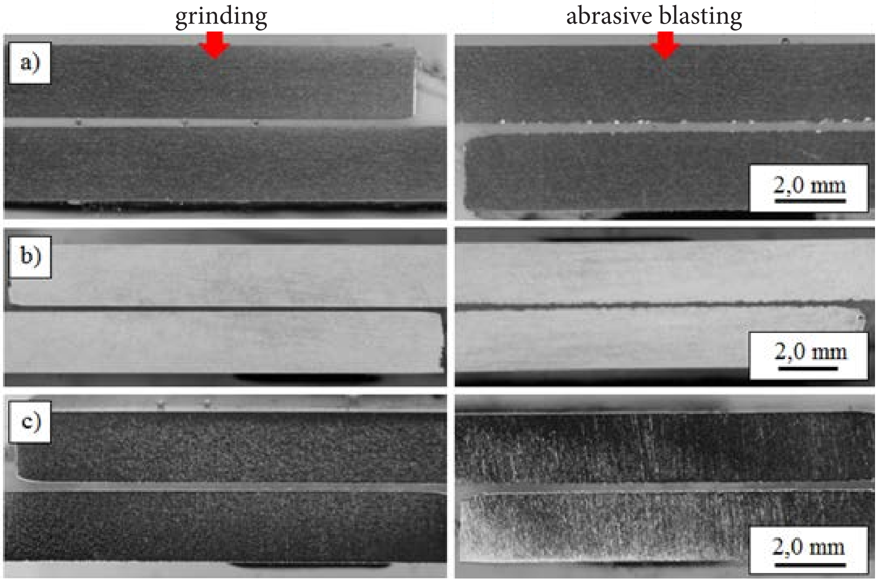

Fig. 3. Macrostructure of the adhesive bonded joints of aluminium alloy EN AW 5754 (a), magnesium alloy AZ31B (b) and titanium alloy Grade 2 (c) made using adhesive HY 4080 GY

The adhesive bonded joints were tested using a testing machine after 48 hours following the adhesive application. The static shear test of overlap joints was performed using a universal testing machine provided with a hydraulic drive (Louis Shopper), where the cross-beam travel rate amounted to 0.2 cm/min and the measurement range was up to 10 kN.

Metallographic Tests

During the metallographic tests the overlap adhesive bonded joints were cut at the half of the overlap and included in epoxy resin EPIDIAN 53. Afterwards, the metallographic specimens were subjected to grinding performed using abrasive paper having gradation restricted within the range of 400 to 2500 and to polishing performed using diamond slurry having 1 μm-sized grains. The above-presented metallographic specimens were subjected to observations performed using an SZX7 (macroscopic photographs) and a CK40M (microscopic photographs) light microscopes (Olympus) coupled with a Toup View camera and a software programme enabling the digital processing and archiving of photographs.

Figure 3 presents selected macrostructures of joints in relation to individual material groups, made using adhesive Loctite HY 4080 GY and subjected to mechanical treatment. Microscopic tests performed using light microscopy did not reveal the noticeable effect of treatment involving the use of low-temperature plasma. It is possible to analyse the effect of low-temperature plasma on the properties of the surface layers of metals, yet it requires the performance of complicated and expensive tests such as the XRD or EXPS analyses. The photographs present air bubbles formed when stirring individual components of the adhesive and during its application. The remaining joints, made using adhesive Agomet F330, were similar as to their appearance and were characterised by the presence of the similar number of single air bubbles in the adhesive bonded weld.

The photographs (Fig. 4) presenting the microstructure of adhesive bonded joints made using adhesive Agomet F330 revealed the effect of mechanical treatment on the expansion of surfaces subjected to adhesive bonding. The use of abrasive blasting led to the significantly greater surface expansion providing more favourable conditions for the obtainment of higher mechanical adhesion resulting in the higher strength of an adhesive bonded joint.

To better demonstrate the effect of individual mechanical treatment methods on surface expansion, the microscopic test results were confronted with results of surface roughness measurements. The surface roughness measurements were performed using a portable MarSurf PS10 profilometer (Mahr) equipped with a measurement probe provided with a diamond tip having the point radius of 2 µm and the measurement range of 350 µm. Table 2 presents measurement results concerning two primary surface roughness parameters R_a and R_z related to individual types of surface treatment. The table contains the average values of measurements performed along three measurement lines. As could be expected, the greatest surface roughness was obtained using abrasive blasting. The use of additional treatment, i.e. low-temperature plasma, resulted in slight surface smoothing. The roughness of surface also depended on the material subjected to treatment; the same treatment parameters were used in relation to all of the test materials. The greatest surface roughness was obtained in relation to magnesium alloy AZ31B, indicating the poorest mechanical properties among the materials subjected to the tests (see R_m in Table 1), whereas the lowest surface roughness was that of titanium Grade 2, i.e. the material characterised by the highest mechanical parameters.

The adhesive bonded joints presented in Figure 4 were free from imperfections.

Table 2. Surface roughness of aluminium alloy EN AW 5754, magnesium alloy AZ31B and titanium alloy Grade 2 in relation to the method of treatment

Material	Surface treatment	Surface roughness, µm	
		*Ra*1)	*Rz*2)
Aluminium EN AW 5754	grinding	0.53	4.50
	abrasive blasting	12.43	78.82
	grinding + plasma	0.46	4.06
	abrasive blasting + plasma	11.37	76.11
Magnesium AZ31B	grinding	0.64	7.07
	abrasive blasting	13.67	83.18
	grinding + plasma	0.59	6.86
	abrasive blasting + plasma	12.98	81.02
Titanium Grade 2	grinding	0.37	2.68
	abrasive blasting	8.50	62.19
	grinding + plasma	0.31	2.24
	abrasive blasting + plasma	7.97	61.05

1) *Ra* – average arithmetic profile deviation from the mean line
2) *Rz* – roughness value in relation to 10 profile points

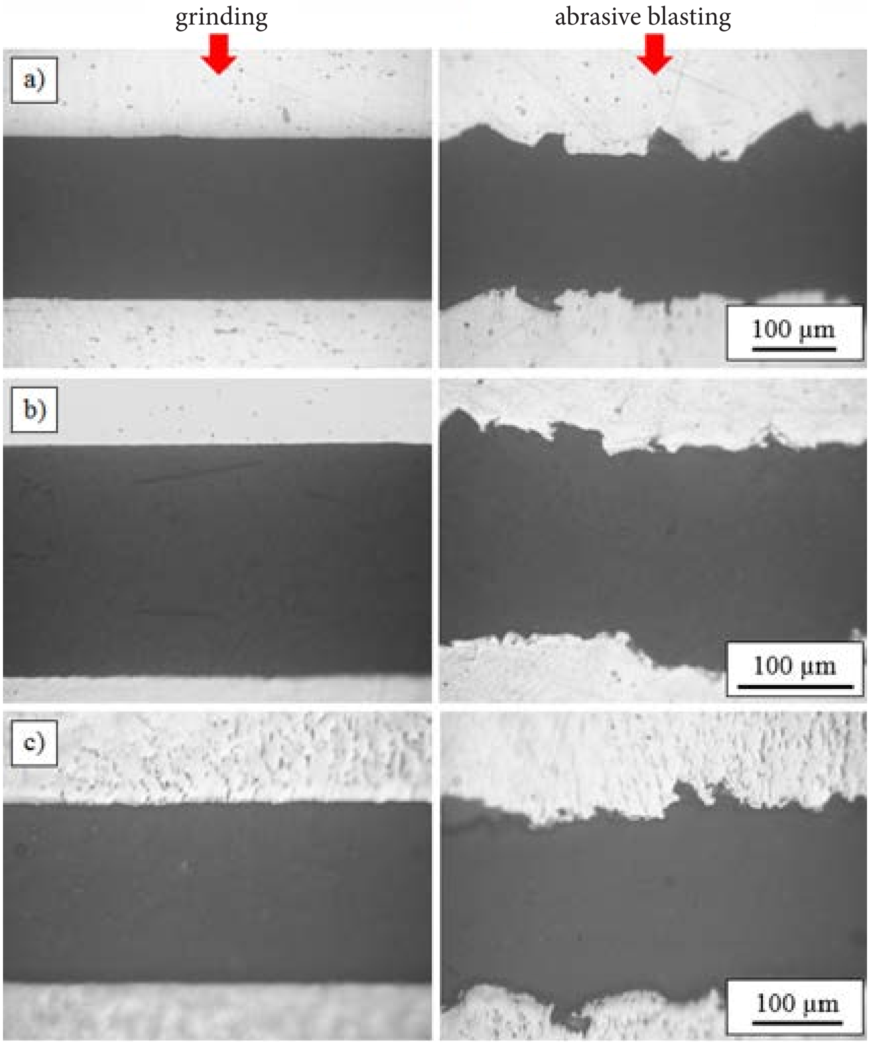

Fig. 4. Microstructure of the adhesive bonded joints of aluminium alloy 5754 (a), magnesium alloy AZ31B (b) and titanium ally Grade 2 (c), made using adhesive Agomet F330

Strength Tests

The results obtained in the static shear tests of the overlap adhesive bonded joints are presented in Table 3. The table contains average values of five measurements in relation to each manner of surface modification performed before the process of adhesive bonding and in relation to an adhesive used in a given test. The results reveal both the effect of mechanical treatment (grinding, abrasive blasting) providing appropriate surface expansion and that of low-temperature plasma making it possible to increase surface energy. In addition, the results of the static shear test in relation to individual materials, surface modification methods and adhesive types are presented in the form of a graph in Figure 5.

The analysis of the results obtained in the static shear test revealed the existence of certain dependences regardless of materials and adhesives used. It was possible to observe correlations between types of mechanical treatment and the strength of adhesive bonded joints. The mechanical strength of the specimens having surfaces prepared using abrasive blasting was by 2 to 4 MPa higher than that obtained using grinding, which, given relatively low strength of aluminium and magnesium alloys, resulted in an increase restricted within the range of 15 to 25%. In addition, the mechanical strength of adhesive bonded joints was increased through the combination of mechanical treatment and the use of low-temperature plasma. The application of low-temperature plasma increased

Table 3. Shear strength of adhesive bonded joints in relation to the surface preparation manner

Material	Surface treatment	Adhesive	Average shear strength Rt, MPa	Standard deviation, MPa
Magnesium AZ31B	grinding	Agomet F330	10.3	2.00
	abrasive blasting		13.2	1.61
	grinding + plasma		12.5	1.34
	abrasive blasting + plasma		14.6	0.07
Aluminium 5754	grinding		11.9	1.00
	abrasive blasting		14.5	1.51
	grinding + plasma		14.5	2.66
	abrasive blasting + plasma		18.1	0.06
Titanium Grade 2	grinding		25.5	8.95
	abrasive blasting		24.5	1.31
	grinding + plasma		28.2	2.44
	abrasive blasting + plasma		27.9	0.75
Magnesium AZ31B	grinding	Loctite HY4080 GY	8.8	0.88
	abrasive blasting		13.5	0.34
	grinding + plasma		10.9	0.42
	abrasive blasting + plasma		18.8	4.34
Aluminium 5754	grinding		11.9	1.65
	abrasive blasting		10.8	0.80
	grinding + plasma		16.5	5.79
	abrasive blasting + plasma		18.3	0.27
Titanium Grade 2	grinding		17.0	1.13
	abrasive blasting		19.1	0.43
	grinding + plasma		18.3	0.21
	abrasive blasting + plasma		20.8	0.52

mechanical strength by further 15 to 25% in comparison with the strength obtained using mechanical treatment only. The highest mechanical strength, regardless of a material, was observed in cases of joints, the surface of which was prepared using abrasive blasting combined with the treatment performed using low-temperature plasma. The foregoing led to the appropriate surface expansion (mechanical adhesion) and the proper cleaning of the surface and, consequently, resulted in the obtainment of higher surface energy (specific adhesion). Significant differences in terms of strength related to individual materials resulted primarily from different properties of surface layers of individual metal alloys, entailing various specific adhesion on each individual substrate.

Regardless of the adhesives, materials and surface treatments used, the failure of the adhesive bonded joints was similar. Exemplary fractures of adhesive bonded joints made using hybrid adhesive Loctite HY 4080 GY are presented in Figure 6, whereas the fractures of adhesive bonded joints made using adhesive Agomet F330 are presented in Figure 7.

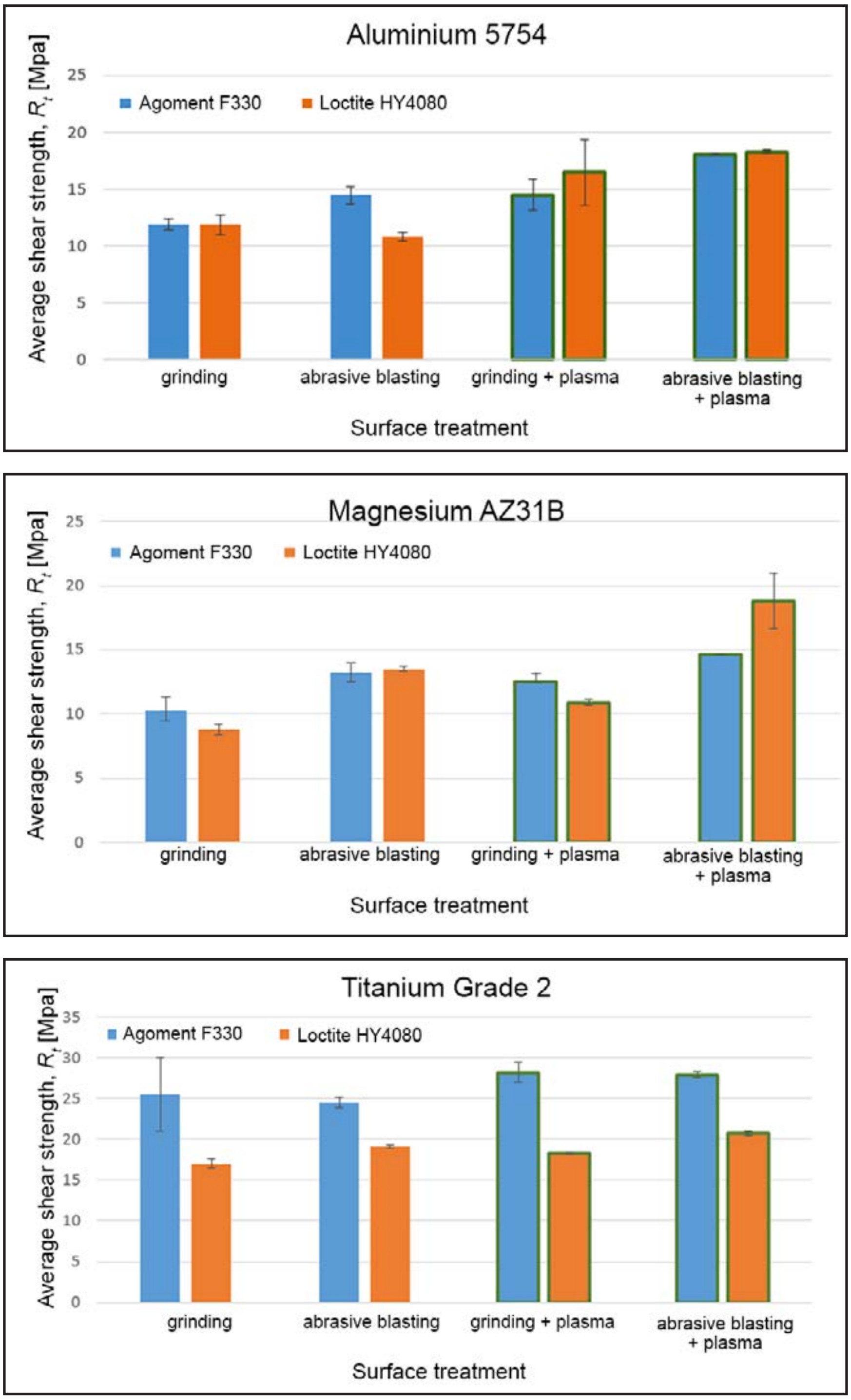

Fig. 5. Average shear strength of adhesive bonded joints in relation to the manner of surface preparation

Regardless of the adhesives, materials and surface treatments used in the tests, the failure of the adhesive bonded joints was similar. In each case, the failure was adhesive in nature. It can be concluded that shear strength differences of individual groups of materials were affected by specific adhesion, varying in relation to each material subjected to adhesive bonding. Depending on the base material, its surface energy and polarity, the adhesive formed bonds characterised by various strength values. A similar situation can be observed when performing the adhesive bonding of various plastics, where the use of the same adhesive and various grades of plastics results leads to significant differences in the strength of joints.

Conclusions

The performed tests justified the formulation of the following conclusions:

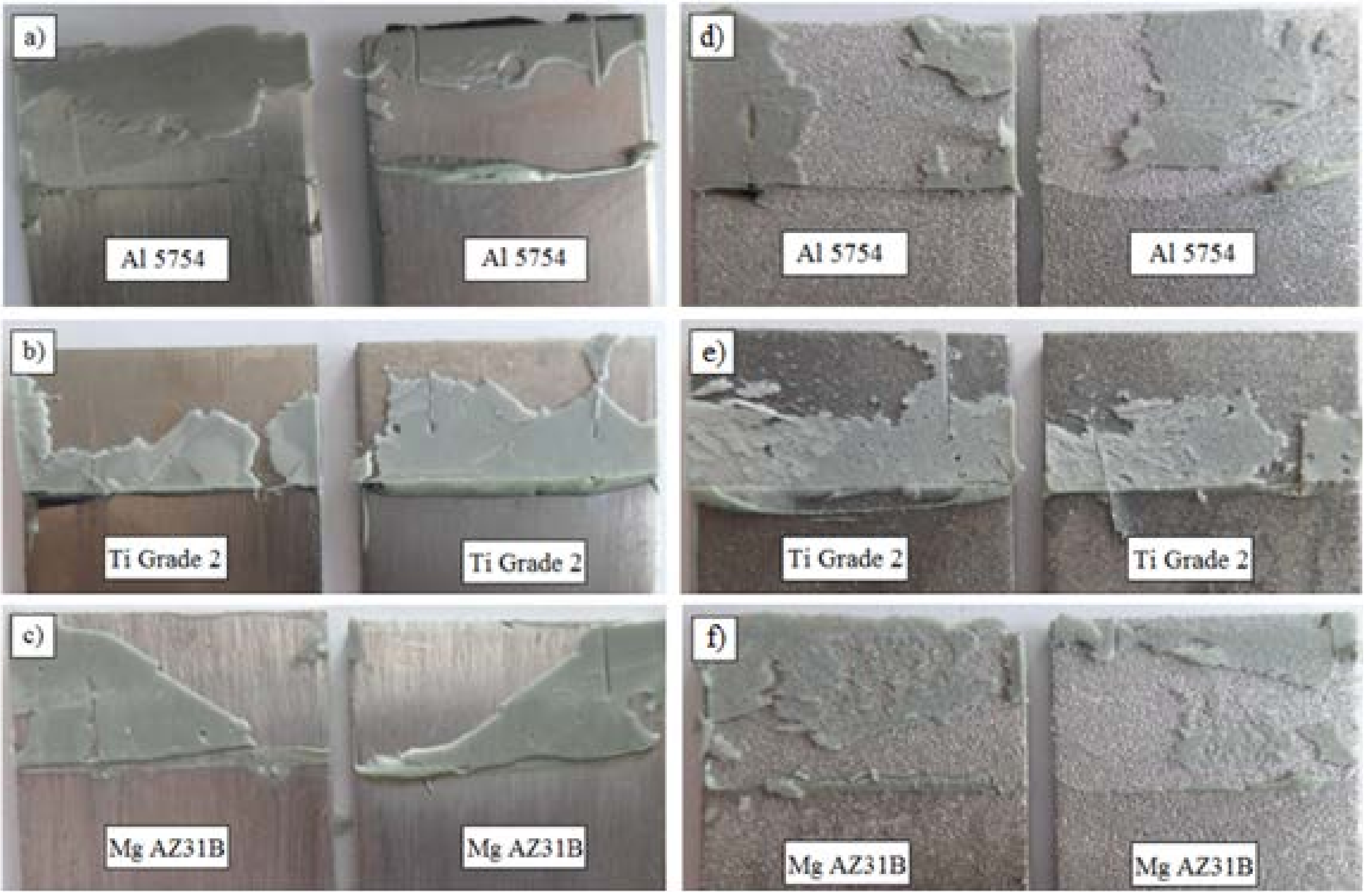

Fig. 6. Exemplary fractures of overlap joints made using adhesive HY 4080 GY, individual alloys and various types of treatment, i.e. grinding (a-c) and abrasive blasting (d-f)

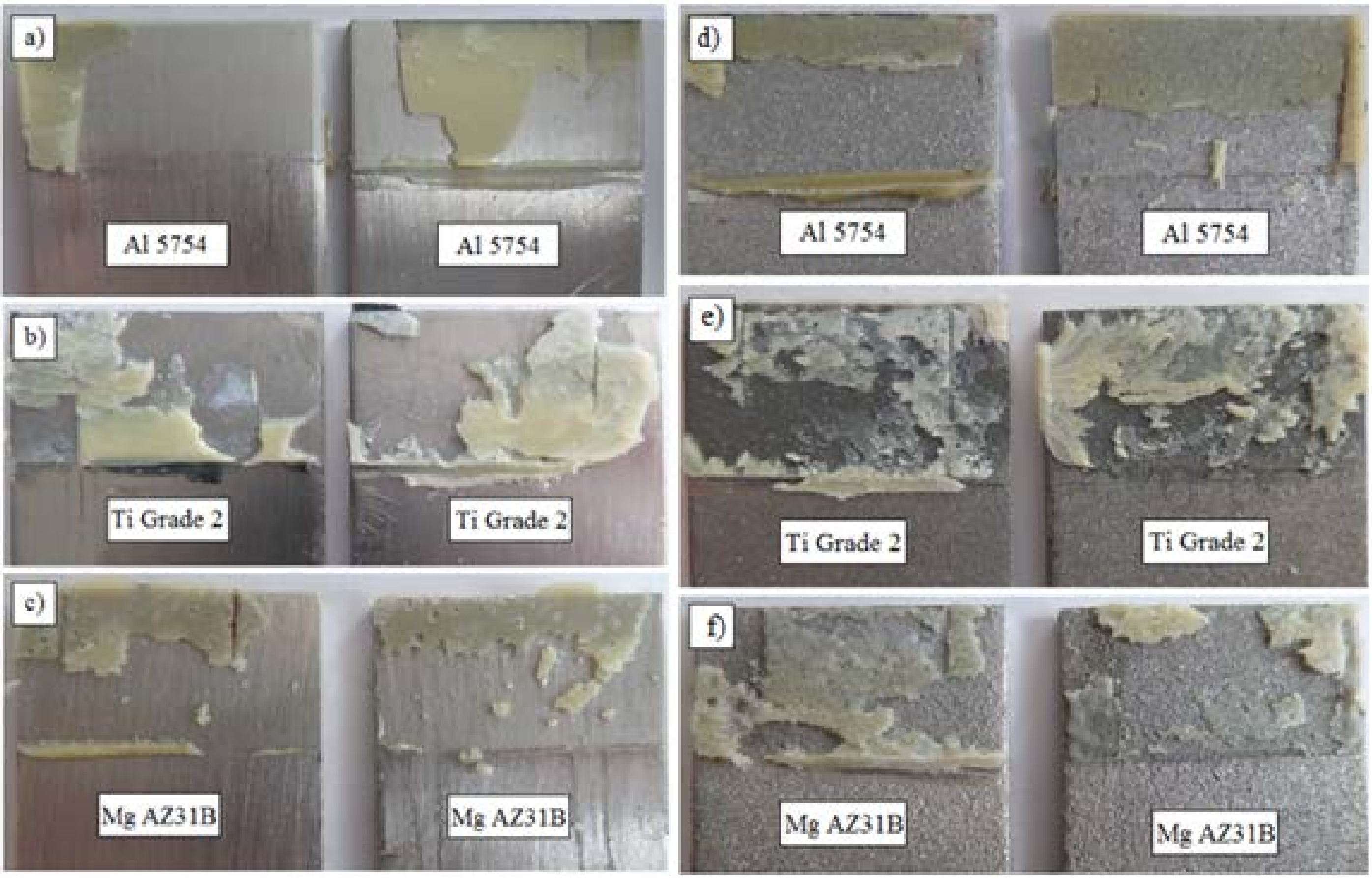

Fig. 7. Exemplary fractures of overlap joints made using adhesive Agomet F330, individual alloys and various types of treatment, i.e. grinding (a-c) and abrasive blasting (d-f)

- use of abrasive blasting enabling the appropriate preparation of surface has a favourable effect on the mechanical strength of adhesive bonded joints (obtained strength compared to that obtained using grinding was higher by up to 25%),
- by increasing surface energy leading to greater specific adhesion, additional treatment involving the use of low-temperature plasma increased mechanical strength to 28 MPa for the joints of titanium Grade 2,
- in addition to surface modification, the final strength of an adhesive bonded joint was also affected by the type of metal subjected to adhesive bonding and by metal surface properties. The highest shear strength was obtained in relation to the adhesive bonded joints of titanium Grade 2, whereas the lowest shear strength was characteristic of the joints of magnesium AZ31B,
- newly developed hybrid adhesives, characterised by significantly shorter initial bonding time, enable the obtainment of joints characterised by similar mechanical strength (particularly as regards tested aluminium and magnesium alloys) to that obtained using strong methacrylate resin-based structural adhesives.

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