Influence of laser ablation and plasma surface treatment on the joint strength of laser welded aluminum-polyamide assemblies.

A. Al-Sayyad*[a], J. Bardon[b], P. Hirchenhahn[c], G. Mertz[b], C. Haouari[bd], L. Houssiau[c], P. Plapper[a]

[a]University of Luxembourg, 6 rue Coudenhove-Kalergi, 1359 Luxembourg, Luxembourg;
[b]Luxembourg Institute of Science and Technology, 5 rue Bommel, 4940 Luxembourg, Luxembourg;
[c]University of Namur, 61 Rue de Bruxelles, 5000 Namur, Belgium
[d]Polytech' Lille, Avenue Paul Langevin, 59655 Villeneuve d'Ascq, France

ABSTRACT

Laser assembly of a metal with a polymer is an innovative process for the development of hybrid lightweight structures.

It was already demonstrated that surface treatment of aluminum prior to laser joining has a critical influence on joint strength of laser assembly with polyamide. In this work, further investigation of the influence of surface treatment prior to laser assembly is carried out. In particular, two kind of surface modification pretreatments of aluminum, laser ablation and plasma surface modification, in combination with plasma surface pretreatment of polyamide, were investigated. Surface properties of aluminum and polyamide after pretreatment are compared to their untreated state. More precisely, surface chemistry, surface energy and roughness characteristics are evaluated by X-ray photoelectron spectroscopy (XPS), sessile drop tests and 3D profilometry, respectively. Joint strength of laser assembly of treated aluminum and polyamide is reported. The more influential surface characteristics for the improvement of joint strength are determined, paving the way to significant advances in metal-polymer laser assembly technology.

Keywords: Laser welding, polymer, metal, surface treatment, hybrid structures, plasma surface modification

1. INTRODUCTION

Metals and polymers are widely used in the fabrication of many goods and components due to their appealing properties. Metallic materials are used mainly for their high mechanical properties and distinctive thermal and electrical properties. Polymeric materials are industrially appealing for their low density, low cost, high corrosion resistance, and high deformability. Metal to polymer joining is beneficial in combining distinctive properties of both materials, leading to an overall reduction in weight, and cost, of the joined component with no significant impact on its mechanical properties. This would serve many industries including automotive, ship manufacturing, and aerospace.

Mostly, Hybrid metal-polymer assembly is conventionally achieved by means of mechanical joining or adhesive bonding [1]. However, those joining techniques have several limitations. Mechanical joining introduces additional weight and stress concentration points to the joined component, while adhesive bonding produces harmful environmental emissions and requires relatively long curing time [2]. To address problems related to conventional metal-polymer joining techniques, thermal joining techniques, such as laser welding, have been developed. However, thermal joining of polymers to metals is challenging due to differences in melting temperature of both materials. Moreover, using a guided laser beam as a heat source in the thermal joining of metals to polymers is very much appealing; it possesses the ability to finely control the heat density at the joint interface to avoid polymers degradation, and it is applicable to various welding geometries. However, deeper understanding of the process parameters and adhesion phenomena is crucial for the industrialization of the process.

In a recent review [3], Tamrin and coworkers reported that, among other factors, surface treatment of materials prior to laser assembly has a large influence on the joint strength. This research work contributes to give further understanding of the influence of surface characteristics on the laser joint strength. To achieve this objective, aluminum and polyamide 6.6 are surface treated before they are assembled by laser. Their surface characteristics such as their topography and chemistry are evaluated and a relationship between these properties and joint strength is discussed.

2. EXPERIMENTAL WORK

2.1 Materials

In these experiments, 0.5 mm thick EN-AW1050A aluminum (Al) in half-hard state, together with 4 mm thick polyamide 6.6 (PA), purchased from Dutec, were used. The samples had a geometry of 30 mm × 60 mm and 25 mm × 75 mm for Al and PA, respectively. Prior to any surface treatment, PA was cleaned by a 10 min immersion in an ultrasonic (US) bath in isopropanol, and Al sheets by a 5 min immersion in an US bath in ethanol. Both were left to dry overnight. The cleaned samples are referred to as "reference" in this report.

2.2 Plasma setup and deposition parameters

Plasma treatment of Al and PA was performed in an open reactor operated at atmospheric pressure [4]. Working gas was a mixture of 80% vol. nitrogen with 20% vol. oxygen gas. Samples were set on the bottom electrode and exposed to plasma when the high voltage top electrodes was moved back and forth over the samples. The number of movement (passes) was set to determine the total treatment time. More precisely, total treatment time was 96 s for Al sample and 60 s for PA sample.

2.3 Laser processing

2.3.1 Laser ablation

Al surface was ablated by means of short-pulsed laser beam using TruMark 6130 laser from TRUMPF, forming a naturally oxidized layer at the top of the Al surface.
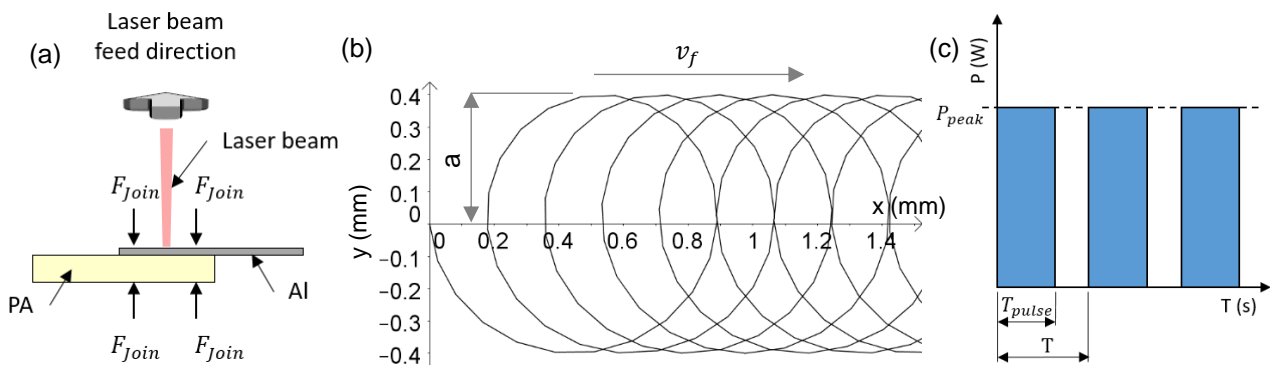
2.3.2 Laser welding

Figure 1. (a) schematic diagram of direct laser beam welding process, (b) spatial modulation, (c) temporal modulation.

Laser assembly is performed as described in fig. 1.a. To achieve a precise control of heat input and to avoid polymer degradation at the joint interface, a fiber laser with a wave length of 1070 nm and spot diameter of 31 μm was used to perform the welding process as described in [5]. In order to enlarge the interface width, a circular spatial power modulation is superposed to the feed direction as shown in fig. 1.b. Several laser manufacturers refer to the superposed trajectory as "wobble". In the reported experiments, wobble amplitude of a=0.4 mm, circular oscillation frequency of f_{osc}=500 Hz, and a corresponding feed velocity of the center of the oscillated laser beam $v_f = 88.8$ mm/s were used.

Moreover, to prevent polymer degradation due to excessive heat input, laser power was adapted to the welding process by temporal power modulation as illustrated in fig. 1.c. A temporal frequency 1/T=25 KHz, peak pulse power of P=400 W, and a pulse duration of T=35 µs, was used in the reported experiments.

2.4 Analytical techniques

2.4.1 Design of Experiments (DoE)

Full-factorial experimental design was set to determine the effect of subjecting both aluminum and polyamide to plasma surface treatment on the welded joint shear strength. Two levels of treatment solvent cleaning (reference), and plasma treatment, on both metal and polymer side, were investigated. In addition, the effect of laser ablating aluminum on the shear strength of the welded joint was reported. Nomenclature of samples according to their preliminary surface treatment is summarized in table 1 below.

Shear strength was quantified by means of a single-lap shear test. To make sure that failure is solely due to subjected shear load, care was taken during clamping by using a specially designed clamping fixture to avoid bending of the welded sheets, i.e. peeling of joint interface was avoided.

Table 1. Sample nomenclature according to several treatment conditions

	Full factorial DoE		
	Al ref (solvent cleaning)	Plasma treated Al (96s)	Laser Ablation (LA)
PA ref (solvent cleaning)	(0,0)	(96,0)	(LA,0)
Plasma treated PA (60s)	(0,60)	(96,60)	-

2.4.2 Surface energy

The wettability of materials surface to water and diodomethane was measured from static contact angle measurements performed with a goniometer (OCA15+ from Dataphysics) using a sessile drop method. A measurement with two liquids was performed for every surface, treated or not. An average and standard deviation value was calculated by performing 3 sessile drop experiments. Surface energy was then calculated from Owens–Wendt–Rabel–Kaelble (OWRK) method, where the surface energy is split into two components, namely a polar and a dispersive part.

2.4.3 XPS

The atomic composition of Al surface and its chemical bonding states were investigated using a XPS equipment described elsewhere [6].

2.4.4 Surface Topography

Surface topography was measured by a Tencor P10 3D profilometer by scanning a 100 x 200 µm² area, with a pixel size of 1 x 1 µm². Three measurements performed at different locations of the sample surface were carried out on each sample to be able to assess the homogeneity of the topography. 3D unfiltered parameter Sa (arithmetical mean surface height) values were then calculated for each sample

3. RESULTS AND DISCUSSION

3.1 Shear strength and DoE analysis

Results of shear strength were analyzed using "Minitab" software. Plasma treatment of both Al and PA, together with laser ablation of Al, resulted in a significant improvement of the shear strength as shown in the interval plot below (fig. 2). Table 2 summarizes the corresponding p-value of non-overlapped error bars of the shown interval plot. It compares different treatment states and determine significant differences in their corresponding shear strength. P-value was calculated using two samples T-test assuming equal variances.

Table 2. P-values based on t-test assuming equal variance, highlighted p-values indicating statistically significant difference between mean values.

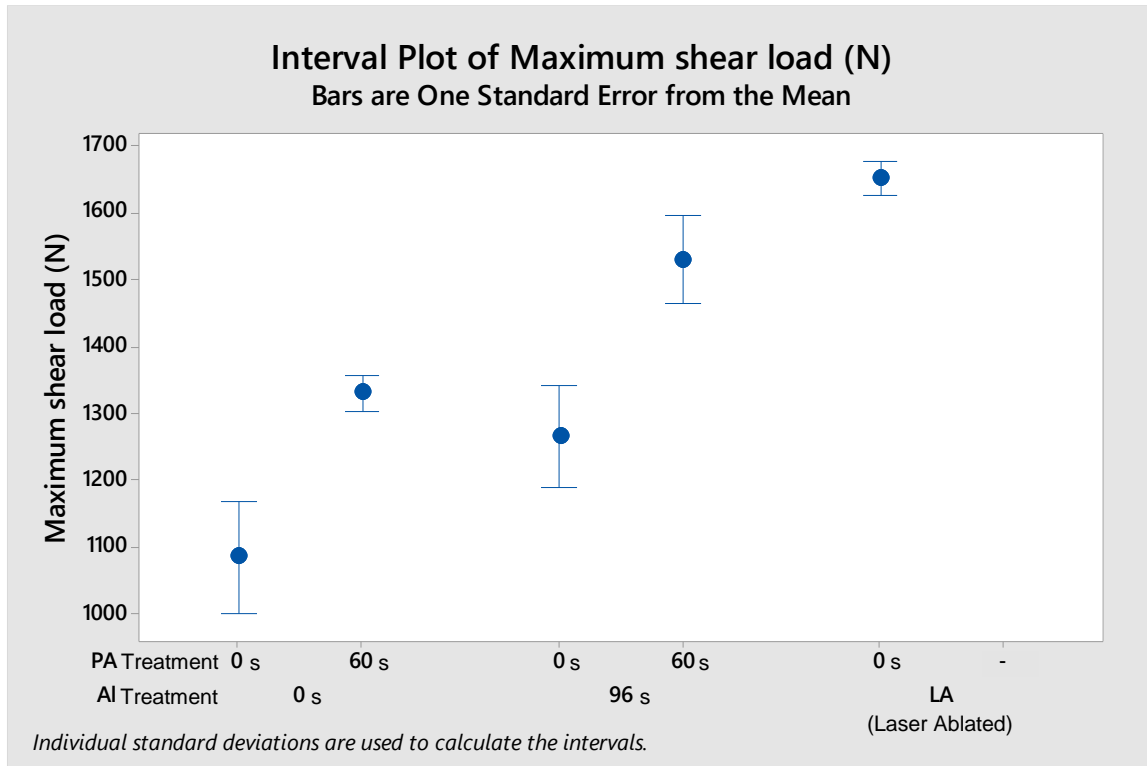|	(0,0)	(0,60)	(96,0)	(96,60)	(LA,0)
(0,0)	-	P=0.02	P=0.15	P=0.00	P=0.00
(0,60)	P=0.02	-	Overlapped	P=0.15	P=0.00
(96,0)	P=0.15	Overlapped	-	P=0.02	P=0.00
(96,60)	P=0.00	P=0.15	P=0.02	-	P=0.1
(LA,0)	P=0.00	P=0.00	P=0.00	P=0.1	-

Figure 2. Interval plot of maximum bearable shear load.

Moreover, analyzing the generated DoE model which describes the effects of plasma treatment on the corresponding shear strength, it can be interpreted from the Analysis of Variance (ANOVA) results that treatment of both aluminum and polyamide are effective in improving the shear strength, with a corresponding p-value of 0.04 and 0.00 respectively. It is noticed, from the p-values, that plasma treatment on the polyamide surface has a higher significant effect, on the shear strength, compared to that of aluminum. Moreover, interaction between plasma treatments on both material surfaces has no significant effect on the outcome (p-value=0.79). To add on, contour plot of the effect of plasma treatment (not shown here), indicates an overall increase in the mean shear load by plasma treatment on both material's surfaces.

3.2 Surface energy

Plasma treatment of Al and PA increases their water wettability, which indicates an increase the surface energy, in particular the polar part.

The same is observed for Al after laser ablation, where water and diiodomethane perfectly wets the surface, indicating an even greater increase in surface energy compared to plasma treatment.

Contact angle measurements and surface energy calculation results are provided in table 3.

Table 3. Contact angle measurements with water and diiodomethane and the corresponding surface energy (SE) calculations using the OWRK method and the Ström & Al surface tension for both liquids. St. dev. stands for standard deviation.

	Contact angle (°)				Surface Energy (mN/m)			
	water		diiodomethane		total SE		polar part of SE	
	average	st. dev.	average	st. dev.	average	st. dev.	average	st. dev.
Pa (-,0)	70.4	2.1	38.2	2.1	47.5	3.3	7.0	1.3
Pa (-,60)	50.1	2.8	48.7	0.1	55.2	1.3	20.2	5.4
Al (0,-)	68.8	2.4	47.4	0.8	44.9	2.9	9.2	2.1
Al (96,-)	40.6	3.0	42.2	1.4	62.6	3.9	24.1	2.8
Al (LA,-)	0.0	0.0	0.0	0.0	>81.4	0.0	30.6	0.0

3.3 XPS

For the purpose of this work, an XPS analysis was performed only on the aluminum sheets before and after treatment. The results are presented in table 4. In addition to the surface composition of each sample, a mean value for the oxide thickness is calculated as described by Strohmeier [7]. This value must be considered cautiously because (i) the exact nature of oxide (or hydroxide) is not cross-checked by another technique and (ii) it does completely neglect the roughness in the calculation.

Table 4. Elemental composition, amount of aluminum that is metallic, and mean value of oxide thickness [7], obtained by XPS measurements for aluminum sheets before and after different treatments.

Sample	% Al	%O	%C	% N	% F	% Al metal	Oxide thickness (nm)
Al (0,-)	29.88	46.89	17.15	1.29	4.79	25.98	4.5
Al (LA,-)	31.59	58.07	9.98	0.36	0.00	1.97	11.9
Al (96,-)	25.75	55.04	12.31	1.47	5.43	23.73	4.8

Fluorine is certainly a contamination from the analysis chamber and will no longer be discussed.

Surface treatments (plasma and laser ablation) both leads to a surface which is richer in oxygen and poorer in carbon. However, this change is more pronounced for laser ablated Al where a much larger oxide thickness is calculated. The latter is certainly assigned to the laser ablation process which oxidizes the surface, so that more oxygen is present, and at the same time "cleans" it, thereby decreasing the presence of adventitious carbon.

As for nitrogen (N) element, the increase in N content is low for plasma treated Al and is certainly assigned to the contact with activated species in the nitrogen-rich plasma. N content of laser ablated sample is very low, probably due to the presence of a large oxide layer on the surface.

3.4 Topography

Sa values are provided in table 5.

Table 5. Sa values for treated and untreated PA and Al.

	PA (-,0)	**PA (-,60)**	**Al (0,-)**	**Al (96,-)**	**Al (LA,-)**
Sa, nm	44.4	42.0	319	304	1083

The 3D profilometry measurements do not show any significant changes in the roughness amplitude of Al and PA surfaces after plasma treatment. On the contrary, the roughness amplitude of laser ablated Al surfaces is greatly increased, by a factor of 35.

Moreover (information not shown here), untreated and plasma treated Al show a series of parallel grooves, certainly because Al is processed by rolling, whereas these grooves completely disappear after laser ablation and are replaced by series of aligned "pyramid-like" features.

4. CONCLUSION

A significant increase of shear strength of aluminum-polyamide assemblies is recorded after plasma treatment of aluminum and seems to be related only to chemical modification of aluminum surface. When aluminum surface topography and chemistry is changed by laser ablation, the increase in shear strength is larger, and is assigned to additional strengthening effects related to the very high increase in roughness amplitude.

Plasma surface treatment of polyamide is also effective in improving the shear strength of the laser assembly. However, its interaction with plasma treatment of aluminum (i.e. when both surface are treated before assembly) is not significant, which requires further investigations.

The increase in shear resistance of aluminum for both treatments and polyamide for plasma treatment follows the increase of total surface energy of both surface after treatment, and more particularly the increase of the polar part of surface energy.

This work has confirmed that an improvement of the assembly's shear strength can certainly be achieved without an increase of the interfacial interlocking, as observed in case of plasma treated Al samples. Such improvement can be correlated to changes in the surface chemistry of the assembled parts as a result of surface pretreatment.

REFERENCES

[1]   S. Amancio-Filho and J. dos Santos, "Joining of polymers and polymer–metal hybrid structures: Recent developments and trends," Polymer engineering & science, vol. 49, no. 8, p. 1461–1476, 2009.

[2]   P. Kah, R. Suoranta, J. Martikainen and C. Magnus, "TECHNIQUES FOR JOINING DISSIMILAR MATERIALS:," Rev. Adv. Mater. Sci, vol. 36, pp. 152-164, 2014.

[3]   K. F. Tamrin, . Y. Nukman and S. S. Zakariyah , "Laser Lap Joining of Dissimilar Materials: A Review of Factors Affecting Joint Strength," Materials and Manufacturing Processes, vol. 28, p. 857–871, 2013.

[4]   J. Bour, J. Bardon, H. Aubriet and D. Ruch, "Different Ways to Plasma-Polymerize HMDSO in DBD Configuration at Atmospheric Pressure for Corrosion Protection," Plasma Processes and Polymers, vol. 5, no. 8, pp. 788 - 796, 2008.

[5]   C. Lamberti, T. Solchenbach, P. Plapper and W. Possart, "Laser Assisted Joining of Hybrid Polyamide-Aluminum Structures," Physics Procedia, vol. 56, pp. 845-853, 2014.

[6]   C. Sperandio, J. Bardon, A. Laachachi, H. Aubriet and D. Ruch, "Influence of plasma surface treatment on bond strength behaviour of an adhesively bonded aluminium-epoxy system," International Journal of Adhesion and Adhesives, vol. 30, no. 8, pp. 720-728, 2010.

[7]   B. R. Strohmeier, "An ESCA method for determining the oxide thickness on aluminum alloys," Surface and Interface Analysis, vol. 15, no. 1, pp. 51-56, 1990.