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Adhesive joining of aluminium AA6082: The effects of resin and surface treatment

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Abstract

In this work the effects of both the substrate surface condition and the adhesive properties on single-lap aluminium joint resistance were analysed. The aluminium sheets were mechanically treated with two abrasive surfaces evaluating the induced roughness; four different resins were used in adhesion tests. Moreover, wettability tests were performed in order to evaluate the effect of the above-mentioned parameters on the substrate/adhesive interaction. A design of experiments was defined in order to quantify the effect of the considered factors and their correlation.

Keywords: Aluminum and alloys; Interfaces; Surface roughness/morphology; Surface treatment; Lap-shear

1. Introduction

The automotive industry develops lightweight and energy-efficient vehicles in order to accommodate the conflicting requirements coming from environmental legislation with the customer demands for greater performance, more luxury and safety features. With this aim, efforts are mainly directed towards the substitution of aluminium for steel in the body structure—corresponding to 20/30% of the total weight of the vehicle [1]. Aluminium structures are lighter than traditional steel ones and are already in use [2] to meet the requirements, in terms of both vehicle design and manufacture [3,4]. However, this substitution is not automatic, but it is important to study the material properties and the structural design in order to optimise car performance. As an example, it has been shown that a spaceframe construction (see Fig. 1), consisting of extruded components [5], can be comparable to a conventional steel monocoque in terms of strength and stiffness [1] even though aluminium has lower mechanical properties than steel. This result is due to the design of the spaceframe, together with the use of thicker material sections [6].

This combination between the material substitution and the new methods for basic body frame construction can be considered a significant challenge with respect to the methods of joining.

Welding—that is the typical technique of connection between steel components—is particularly difficult for aluminium due to the formation of a surface oxide layer, the result of aluminium reacting with oxygen in the atmosphere. This film protects the metal from corrosion, but has a considerably higher melting point than aluminium. Successful welding, therefore, depends in part on the technique applied for breaking down this oxide layer [7].

An alternative joining technology is adhesive bonding, which is important for the automotive industry, can be readily automated [8].

The benefits of adhesive bonding have been demonstrated by a number of car manufacturers with the production of concept cars and low-volume niche products, e.g. Jaguar's XJ220 [9], Ford's AIV [10], Rover's ECV3 [4], the Lotus Elise [11], and, to a limited extent, Honda's NSX [12]. Moreover, there are a number of

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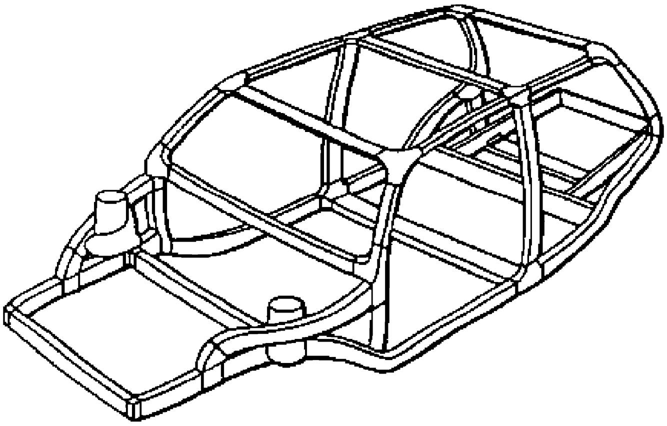

Fig. 1. Example of a spaceframe construction [1].

advantages in using adhesive bonding: it does not distort the joined components much as arc-welding does [7]; adhesive joints are characterised by a continuous bond, with a more uniform stress distribution over a larger area, but their stiffness is comparable with the use of mechanical fasteners or spot-welds, which have localised contact points [13]; adhesive bonding favours energy absorption reducing noise and vibrations [9,14,15]; the smooth joint reduces stress concentrations at the edges, thereby providing good fatigue resistance [9]; the adhesive seals the joint against moisture and debris; it is possible to join similar materials (i.e. metal to metal) and different materials (i.e. metal to polymer), and the adhesive layer—in case of metal substrates—can prevent corrosion [14,15].

But it is also possible to find a number of limitations in adhesive bonding: the joint properties depend on the adhesive choice; adhesives have to be employed with due safety measures (protective clothing, volatile emission control, fire protected storage and so on); structural adhesive can require heat curing, which increases the costs; they have a limited shelf life; materials control is necessary during supply, since their properties (i.e. viscosity) can vary, affecting the final joint characteristics; the joint strength depends on the pre-treatment [4,14,16]; what is more, the durability of such kind of bonding is difficult to assess and few studies are currently available in literature [17].

Pre-treatment is necessary in order not only to remove such contaminants as lubricants and oils, but also to provide the intimate contact needed for the adhesive to bond successfully with the adherent surface. The mere cleaning of the surface is not enough since aluminium passive oxide layer, which occurs naturally on exposure to air, is not optimal for bonding [10], even though it could provide excellent adhesion if atomically clean.

The aim of this work is to experimentally study the adhesive joint between aluminium substrates. Four different resins were employed and two different abrasive surfaces were used to pre-treat aluminium, analysing the effect of both the adhesive and the substrate roughness on the joint resistance.

Single-lap joint tests were utilised for the evaluation of the joint strengths, carrying out wettability tests in order to determine the interaction between adhesive and substrate. In order to evaluate the possibility of correlation among variables, the experimental tests were carried out according to a statistical design of experiment.

2. Experimental set-up

2.1. Materials

2.1.1. Adherents

Aluminium AA6082 sheets were used as a substrate. The behaviour of this material is defined by the tensile experimental tests and described by the flow curve $\sigma = 574\varepsilon^{0.155}$. The sheet thickness, according to the standard ASTM D1002, was 1.5 mm.

2.1.2. Adhesives

Four resins, mainly employed to bond structures bearing either high loads or critical conditions (aggressive environment), were chosen. In the following they will be identified as: "resin PE A, PE B, VE and EPO". Their characteristics, as supplied by the producers, are reported in Table 1. PE A and PE B are orthophtalic polyester resins VE is a vinylester resin and EPO is an epoxy system. This range will be useful to understand the effect of each kind of adhesive on the joint resistance.

Fig. 2 graphically compares some properties reported in Table 1 and shows the gel time versus temperature of the studied resins. Resin EPO has the highest tensile resistance, the resins PE A and VE have similar tensile resistance, while VE has very high elongation at break, and the resin PE B has the lowest mechanical resistance.

Resin EPO is characterised by high viscosity, but it has to be mixed with a high percentage of catalyst, which reduces it. The viscosity of the other resins is lower and their values remain constant during mixing.

Eventually, it is possible to observe that resin EPO shows the same gel time values of other resins only at temperatures greater than 60 °C (see Fig. 2).

2.2. Surface treatment

In the order to remove either weak adhering or contaminated outer layers on the substrate surface, thereby, exposing the freshly oxidised adherent bulk material directly to the adhesive, we performed mechanical abrasion, enabling the formation of a suitable surface layer. This treatment is also useful in order to evaluate the effect of roughness on the joint resistance. Roughening using abrasive surfaces is common in many industries. The surface was prepared with two different grinding papers, identified by P180 and P40. The 2.5% of the thickness was removed in order to obtain a regular surface. The residual particles remaining after mechanical abrasion were

Table 1
Resins properties

Resins	PE A	PE B	VE	EPO
Catalyst	Butanox M50	Butanox M50	Butanox M50	SD 8822
Catalyst quantity (%)	2	2	2	31
Appearance	Blue	Green	Yellow	Yellow
Viscosity (25 °C–37 s^{-1}) (P)	5.3	3.0	3.5	7.0
Stability (darkness −20 °C) (month)	6	3	3	24
Tensile stress (MPa)	57	50	60	70
Tensile modulus (MPa)	3700	3800	2700	3000
Elongation at break (%)	1.8	1.5	4.3	3.8

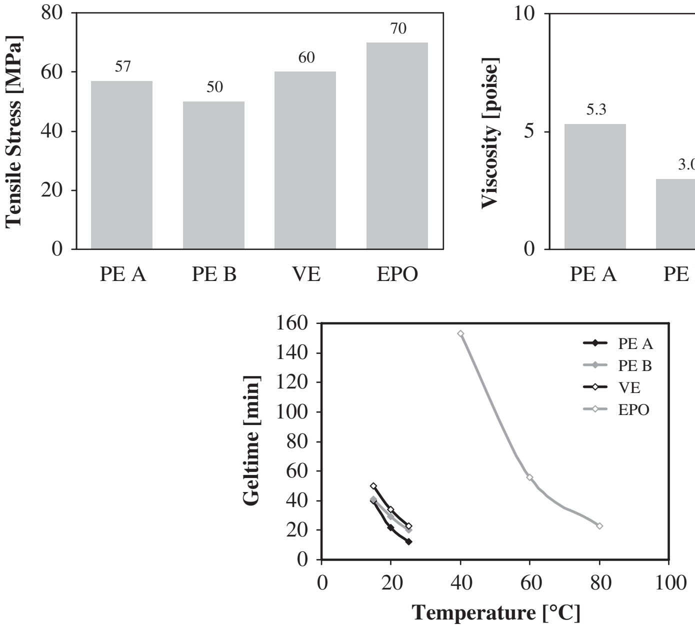

Fig. 2. Resin properties.

removed by cleaning the surface with a soft clean cloth and air in pressure.

2.3. Roughness measurement

A surface profiler, a RTH Form Talysurf, was used to determine the roughness profile, both before and after the surface treatment. The profiler moves along a single direction with a scanning length of 2.5 mm. Measurements were performed in different areas, along three different directions, of the same surface in order to verify the treatment uniformity and the average roughness values "R_a" were calculated. The experimental results showed small spreads, i.e. the variability in measured R_a values was always lower than 10%.

2.4. Wetting measurement

Wetting is a procedure that determines the diffusion of a liquid (adhesive) over a solid surface (substrate), creating an intimate contact between them. The air displacement caused by this physical attraction minimises the interfacial flaws. Good wettability of a surface is a prerequisite for a good adhesive bonding.

Simple wettability tests have been identified in order to assess surface energy/tension before bonding [18–20].

The surface energy is defined as the work necessary to separate two surfaces beyond the range of the forces holding them together and measured in terms of energy par unit area. The contact angle determination at the solid/liquid interface is one of the most sensitive methods for determining the surface energies of solid materials (BS EN 828:1998; ASTM D2578–99a).

Contact angles are closely related to wettability. A liquid (adhesive) will wet a solid (adherent) when its surface energy is lower than the solid surface energy. Balance of forces or equilibrium at the solid–liquid interface is given by Young's equation for contact angles greater than zero (see Fig. 3):

$$\gamma_{lv} \cos\theta = \gamma_{sv} - \gamma_{sl},$$

where θ is the contact angle, and γ_{lv}, γ_{sv} and γ_{sl} are the surface free energies of liquid–vapour, solid–vapour and solid–liquid interfaces, respectively [21].

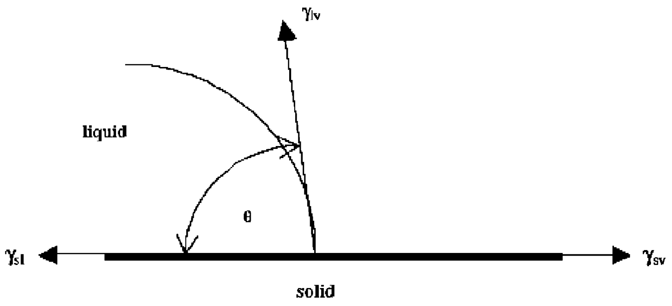

Fig. 3. Contact angle.

Contact angle measurements were realised according to the sessile drop technique and performed by a contact angle micrometer [22].

In order to measure the wetting angles, a 1 μl drop of each resin was accurately deposited on the substrate with a micro-syringe. The contact angle θ was measured by a CCD camera with an optical image (× 10 magnification) captured by a suitable PC images software. The complete instrument set-up is shown in Fig. 4.

The contact angle was measured with the following equation:

$$\theta = 2\,\arctan\left(\frac{H}{r}\right)$$

where H is the height of a droplet and r is the radius of the droplet's base.

The lower the contact angle, the greater the tendency of the liquid to wet the solid, until complete wetting occurs at an angle $\theta = 0$ ($\cos\theta = 1$). The surface tension of the liquid is then equal to the critical surface tension of the substrate. Large contact angles are associated with poor wettability.

2.5. Single lap joint

A wide variety of joint configurations are possible when bonding structures. The single-lap and double-lap configurations are the most commonly found in practice and are applicable for joining relatively thin adherents (i.e. #lt;1.5 mm); on the other hand, the more advanced stepped lap and scarf configurations are used to transfer high loads in joint with thicker adherents. For these the manufacturing is more complicated, requiring low machining tolerances [23].

Since the single-lap joint is generally the simplest and cheapest of all joints to manufacture due to its simple design and easy assembly, it was chosen for the test according to the standard ASTM D1002 for the determination of joint shear strength. The mean shear stress is evaluated by the formula: $\tau = (P/A)$, where P is the tensile load and A is the joint overlap area. In Fig. 5 the adherent and adhesive sizes are defined.

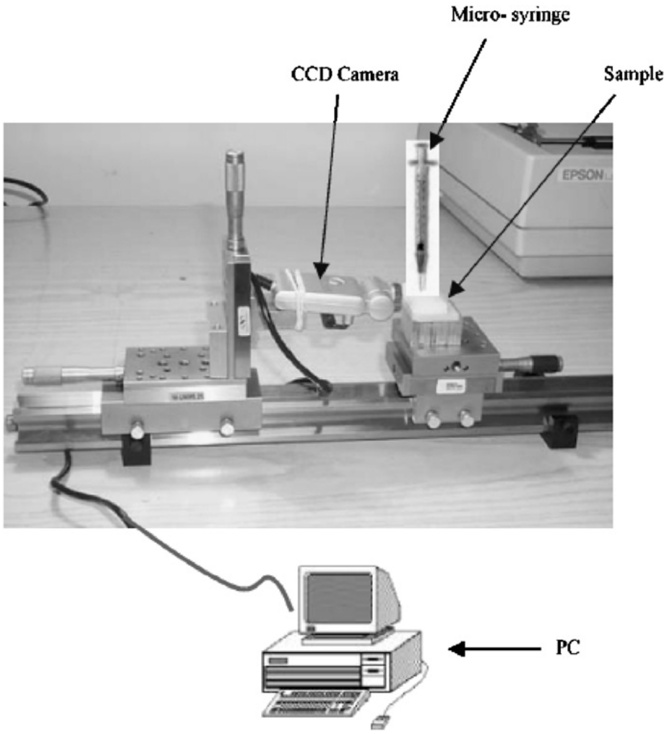

Fig. 4. Experimental apparatus for contact angle measurements.

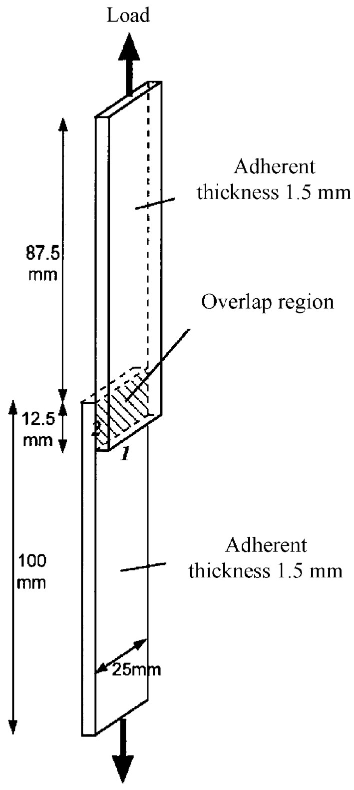

Fig. 5. Joint configuration.

The procedure for the samples preparation, after the mixing of each adhesive with its own catalyst, is the following: firstly, a thin layer of resin is applied to the surface of adherents using a brush, then the substrates are aligned and a pressure is applied with clips to squeeze out extra resin until the standardised thickness is obtained.

Moreover, the samples are realised under extractor fan, at constant and controlled temperature, and then tested after 10 days to allow the full cure of the resins. Universal Testing Machine model Galdabini Sun 5 equipped with a 50 kN load cell and with tensile clamps was employed.

2.6. Design of experiments (DOE)

In order to verify the influence of a number of parameters on both the joint resistance and the wettability, the DOE method was applied.

The safety measures, adopted in the phase of sample preparation, have allowed the elimination of such process factors as joint adhesion pressure, temperature and resin-curing time.

Only two parameters were considered: the surface roughness and the resin type, adopting, respectively, three and four variability levels (i.e. three values of R_a and four kind of resins). For each factor five replicates of the single lap joint test and thirty replicates of the wettability test have been carried out.

The experimental results were analysed through a two-way analysis of variance.

3. Results

3.1. Roughness measurement

The measured profiles on the aluminium sheet surfaces resulted in the following roughness values:

No treatment—$R_a = 0.33\,\mu m$.
Grinding paper P180—$R_a = 1.10\,\mu m$.
Grinding paper P40—$R_a = 2.35\,\mu m$.

3.2. Wetting measurement

Fig. 6 presents the trend of contact angle versus the roughness for every analysed resin. Increasing the roughness, the wettability angle of both polyester resins do not significantly vary, but it slightly increases, reaching 80°. These adhesives, then, similarly wet the substrate.

Epoxy resin has a different behaviour, with high contact angles (max 107°) showing low wettability.

Vinyl ester resin has a good wettability (minimum contact angle 50°), which unlike all the others, increases with the roughness.

It is possible to notice that the interaction between the resin and the aluminium substrate is influenced by the resin's chemical characteristics and that the higher differences are attained at higher roughness values. Moreover, at increasing R_a the contact angle trend decreases only for the resin VE.

Fig. 7 shows the contact angle trends of resin EPO and VE, describing also the different drop shapes at changing roughness.

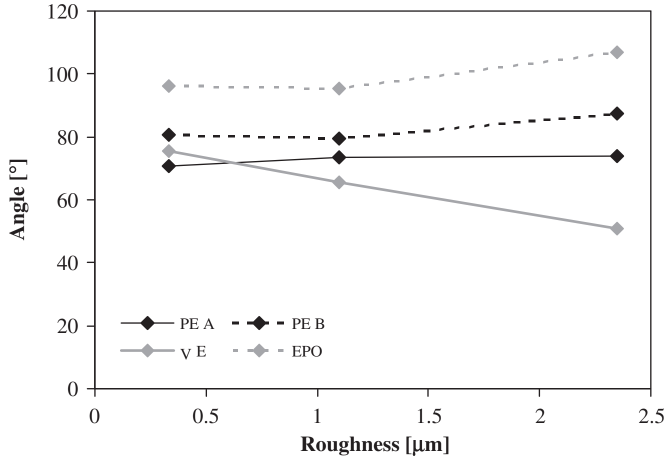

Fig. 6. Contact angle/roughness curves.

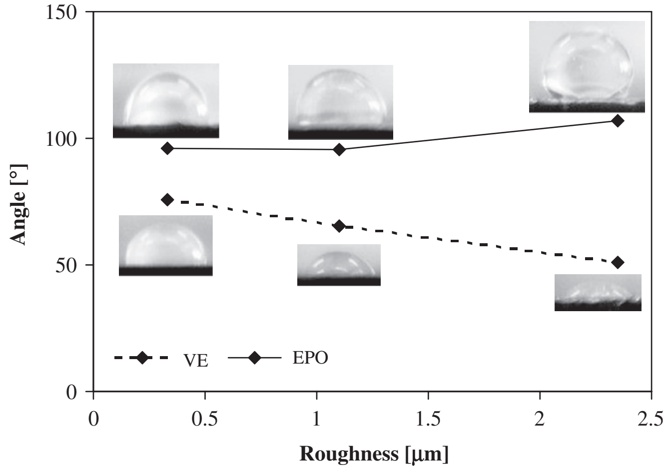

Fig. 7. Drop shape for EPO and VE.

The different behaviour of the two resin/surface systems is evident: the EPO resin tends to create a drop in equilibrium, while the resin VE creates a drop which spreads on the aluminium substrate. This resin shows a better wettability than the others. Also the resin drops show a different behaviour at increasing roughness. In one case (EPO resin), a high roughness favours the creation of a well-defined drop; in the other (VE resin) it favours the drop spreading. This phenomenon is due to the chemical interaction between the resins and the adherent. As far as the resins PE A and PE B are concerned, roughness do not induce a change in the bubble shape.

In Table 2 are reported the contact angles for each resin at varying roughness.

3.2.1. Analysis of variance

Since the first step for the analysis of variance is to look for a correlation between the variables (resin, roughness), the graph in Fig. 6 will actually show an interaction.

The results, reported in Fig. 8, show that the interaction is significant (P-value is smaller to 0.05) and it also shows that both the resin and the roughness are significant factors.

Interaction can be evaluated by the analysis of means: Fig. 9 shows that an interaction effect exists for all resins (their mean wettability values are found outside the confidence range).

Table 2
Contact angle values

Resin	Roughness	Contact angle (deg)			Std. dev. (deg)
		Average	Max	Min	
PE A	0.33	71	76	66	2
	1.10	73	79	68	3
	2.35	74	81	68	3
PE B	0.33	81	89	70	3
	1.10	80	89	70	4
	2.35	87	95	78	4
VE	0.33	76	82	69	3
	1.10	61	70	60	3
	2.35	51	58	45	3
EPO	0.33	96	99	91	2
	1.10	103	101	93	2
	2.35	107	110	102	2

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Source        DF        SS        MS        F      P
Resin          3   62078.6   20692.9  2263.62  0.000
Roughness      2     202.9     101.5    11.10  0.000
Interaction    6   12134.1    2022.4   221.23  0.000
Error        348    3181.2       9.1
Total        359   77596.9

S = 3.023      R-Sq = 95.90%      R-Sq(adj) = 95.77%
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Fig. 8. Statistical parameters.

The different effect of R_a on each kind of resin is clearly evidenced in Fig. 10. It affects in particular the VE and EPO resins wettability: for the first wettability it improves and for the second it worsens when roughness increases. For the other two resins (PEA and PEB) the roughness effect is less consistent.

3.3. Single lap joint test

Fig. 11 shows the mean stresses, determined from single-lap joint tests, at varying the roughness for each resin. The trend is characterised by a significant stress increase from the non-treated to the treated sheets. Thanks to both the elimination of contamination and the introduction of topography the mechanical treatment improves resin interaction with the aluminium surface. This is due to the creation of a physical bond during the spreading phase; a mechanical bond when the sheets are pressed; a chemical bond all along the curing phase. The presence of chemical bonds can be deduced because the increase of wettability does not always cause a consequent increase of the joint strength.

Moreover, the presence of an oxide layer on the surface influences in different ways the joint resistance [24]. The aluminium alloy is characterised by the presence of Mg, Si and Mn. An EDX analysis, performed by the authors, confirmed the presence of MgO (0.33%), this last decreases the bond properties. This happens in the case of the VE resin that, however, allows to realise a joint with similar strength to the EPO, due to its good wettability. This effect is enhanced in PE joints that also have poorer wettability.

Also the adhesive type is important in determining the effect of Mg surface concentration on bond properties [24].

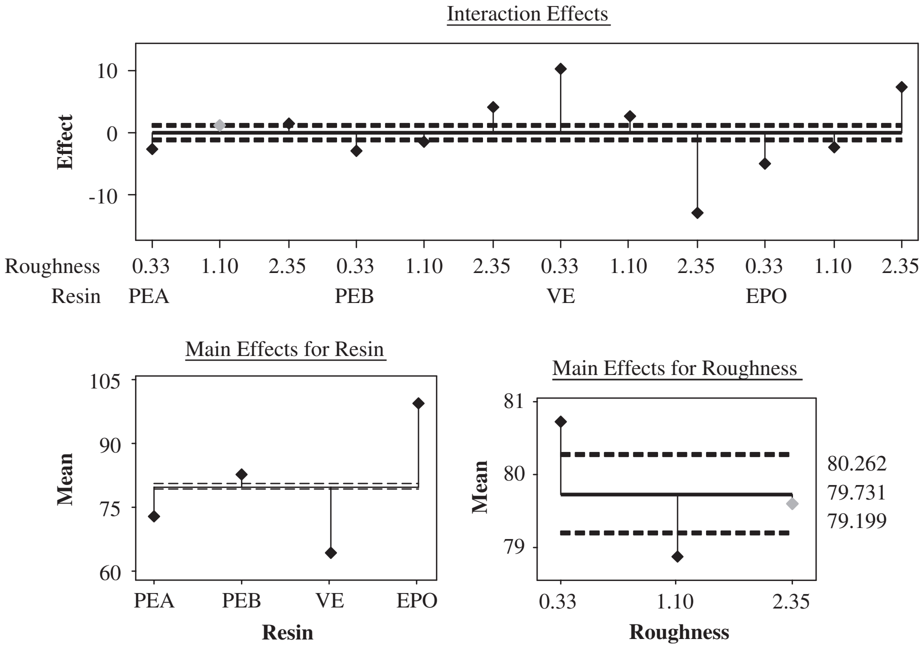

Fig. 9. Analysis of means (α = 0.05).

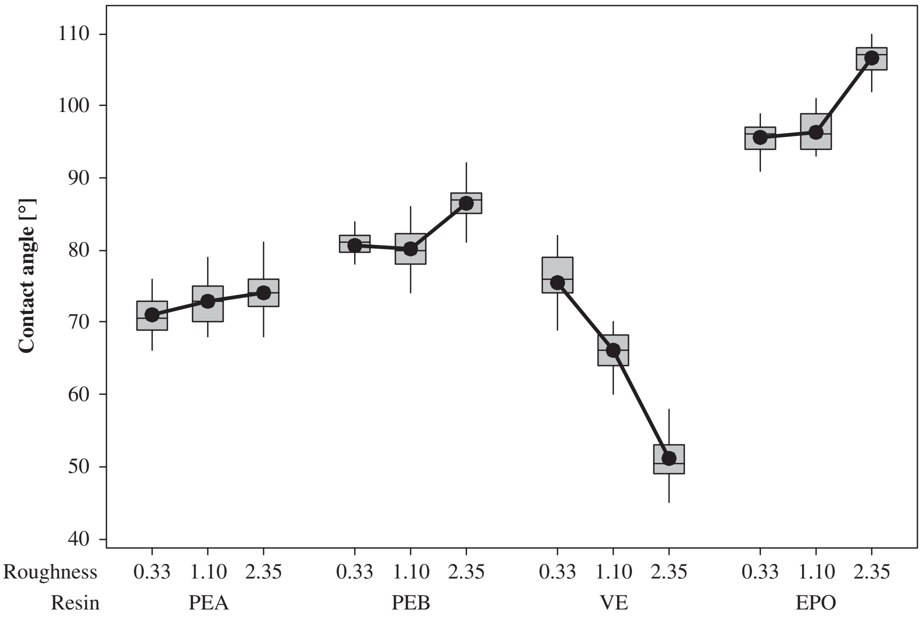

Fig. 10. Boxplot for the contact angle.

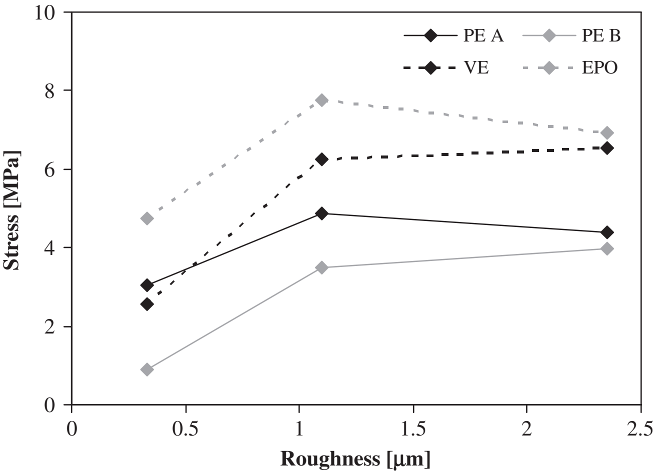

Fig. 11. Stress/roughness curves.

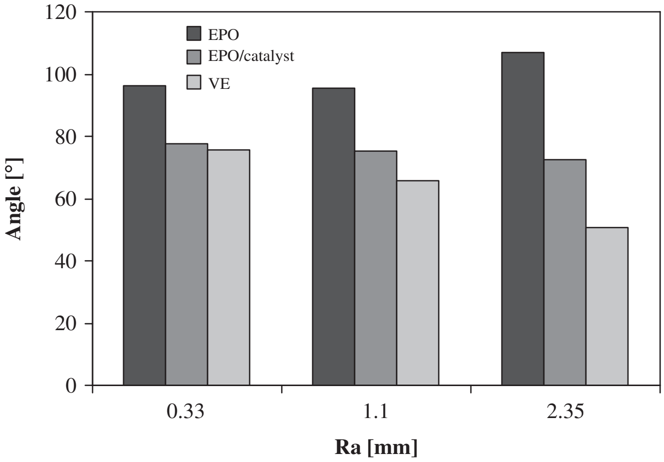

Fig. 12. Wettability for the resin/hardner mix.

The epoxy adhesive contains an alkaline curing agent, the system epoxy/hardener can react with humidity to form products that are strongly alkaline and consequently they tend to destabilise the aluminium oxide. The presence of Mg oxide in the aluminium oxide film is expected to improve the stability in alkaline environment of aluminium oxide due to the fact that Mg oxide is thermodynamically passive at high pH. Then this last phenomenon is favourable to the adhesion with EPO resin. As for the VE resin the hardener is slightly acid and this phenomenon is not present.

Moreover, the wettability of the EPO resin is improved in the mixing due to the high percentage of catalyst (see Fig. 12). In this figure the wettability of EPO with and without catalyst is compared to the one of VE (that is the best among all the tested resins).

In addition, no evident improvements are evident in the joint resistance at increasing roughness between 1.10 and 2.35 µm.

3.3.1. Analysis of variance

Fig. 11 evidences a possible interaction. The analysis of variance shows that the interaction is not significant, so it is possible to use an additive model without interaction. The result of this analysis is that the effect of the two factors is highly significant since *P*-value is lower than 0.01.

The means analysis (Fig. 13) shows no interaction between the effects (roughness and resins). This entails that the effect of increased roughness is the same on all resins.

Fig. 14 shows that both resin and roughness have an influence on the joint resistance. This analysis confirms that

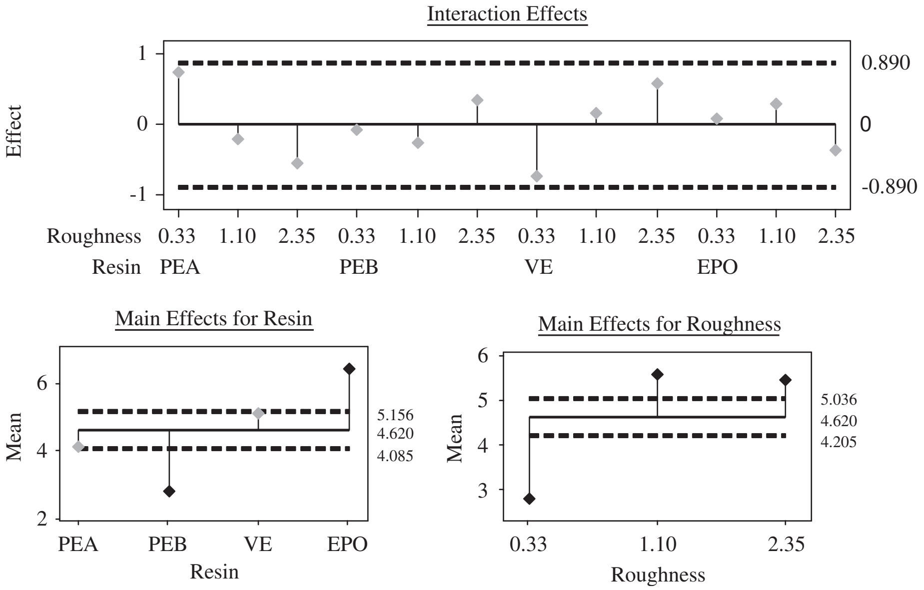

Fig. 13. Analysis of means.

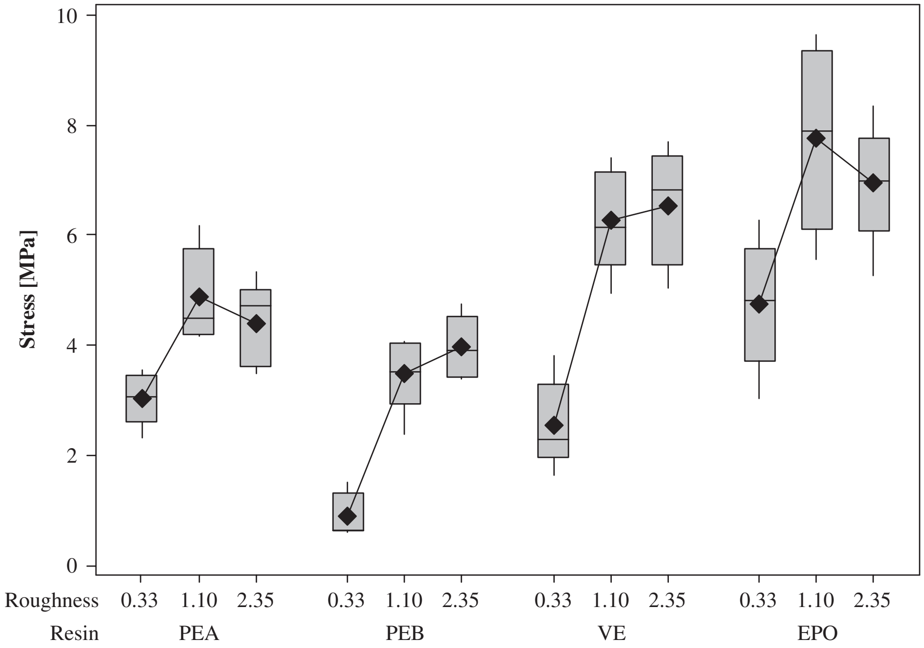

Fig. 14. Boxplot for the stress.

the mechanical abrasion is effective on the joint resistance, but there is no significant improvement at increasing roughness (at least in the analysed roughness range).

4. Conclusions

Studies on wettability:

- Of the four resins in our study, VE has shown the best wettability, EPO the worst.
- Roughness has a different effect on the wettability of each kind of resin. In particular, it has a more significant effect on VE and EPO resin. In the first case, wettability improves, in the other it worsens when roughness increases. In the other two cases (PEA and PEB resin) the roughness effect is less consistent.

Studies on joint resistance:

- *Resin has an influence on the joint resistance*: in spite of their wettability characteristics, the EPO resin joint is the most resistant, thanks to: the intrinsic strength of the adhesive, the effect of mixing with catalyst and the enhanced stability of aluminium oxide

in alkaline environment, while the polyester joints are poorer.
- *Roughness also has an influence on the joint resistance*: the mechanical abrasion is effective on the joint resistance until an ‘‘optimal’’ topography is reached, in fact no significant improvements at increasing roughness is evidenced (at least in the analysed roughness range).
- No interaction is found between resin and roughness.

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