COMPARATIVE ANALYSIS OF WETTING ABILITY OF ALUMINUM SHEETS WITH DIFFERENT SURFACE ROUGHNESS PARAMETERS BY EPOXY ADHESIVE

Analiza porównawcza zdolności zwilżania przez klej epoksydowy blach aluminiowych o różnych parametrach chropowatości powierzchni

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A b s t r a c t: The article presents the analysis of the wetting ability by Epidian 5 epoxy resin with hardener Z1 of EN AW-2017A aluminum alloy sheets. The material sheets were subjected to the selected methods of processing in order to obtain different parameters of the geometric development of the surface. The energy state of the surface layer was examined on the prepared surfaces, taking the polar and non-polar free surface energy components into account. On the basis of the obtained results, wetting envelopes were determined, which represent the limit value of the surface energy components of the wetting liquid, ensuring good wetting. As part of the research, an analysis of the possibilities of achieving maximum adhesion work between a solid and a liquid in the event of changes in the contact angle was also conducted. This analysis allows one to determine how the parameters of the test adhesive deviate from the ideal, i.e. those for which the surface tension at the interface reaches the minimum value. Based on the results of the analysis, a summary was prepared, showing the ability of the adhesive to wet surfaces with different roughness parameters.
K e y w o r d s: free surface energy, contact angle, aluminum alloy, epoxy adhesives

S t r e s z c z e n i e: W artykule przedstawiono analizę zdolności zwilżania przez klej Epidian 5 z utwardzaczem Z1 powierzchni blach ze stopu aluminium EN AW-2017A. Blachy poddano wybranym sposobom obróbki w celu uzyskania odmiennych parametrów rozwinięcia geometrycznego powierzchni. Na tak przygotowanych powierzchniach przeprowadzono badania stanu energetycznego warstwy wierzchniej z uwzględnieniem składowej polarnej i niepolarnej swobodnej energii powierzchniowej. Na podstawie otrzymanych wyników wyznaczono krzywe zwilżania, które przedstawiają graniczną wartość składowych swobodnej energii powierzchniowej cieczy zwilżającej, zapewniające uzyskanie dobrego zwilżania. W ramach badań przeprowadzono również analizę możliwości osiągnięcia maksymalnej pracy adhezji między ciałem stałym, a cieczą w przypadku zmian kąta zwilżania. Analiza ta pozwoliła na określenie na ile parametry badanego kleju odbiegają od idealnych, czyli takich, dla których napięcie powierzchniowe na granicy faz osiąga wartość minimalną. W oparciu o uzyskane wyniki analiz dokonano zestawienia obrazującego zdolność kleju do zwilżania powierzchni o różnych parametrach chropowatości.
S ł o w a k l u c z o w e: swobodna energia powierzchniowa, kąt zwilżania, stop aluminium, kleje epoksydowe

Introduction

Adhesive technology, due to a number of its advantages, is of a great importance in industrial production. In particular, it has found applications in the aerospace, automotive or maritime industries to connect aluminum structures. Bonding offers the possibility of joining elements with large surfaces made of various materials that may vary in thickness. The structures produced using the mentioned technology are characterized by a lower weight and lack of thermal deformation or residual stresses that arise due to heating [8, 14, 15].

As research shows, one of the important factors affecting the creation of adhesive joints is to achieve a strong adhesive bond [3, 4, 10, 12]. This is possible through the selection of a suitable adhesive, which would be characterized by a lower value of surface free energy than the material that we want to connect. It is also important to maintain a balance between the polar and dispersion components of the surface free energy of adhesive and the joined elements. Knowledge of the energy properties of the surface of the material and the applied adhesive composition makes it possible to conduct a wettability analysis that helps to assess the accuracy of the selection of the adhesive or the application of a specific method for preparing the surface of the material. Analysis using wetting envelopes can be an effective, simple, safe and economical tool for assessing the strength of an adhesive joint in the aspect of optimizing adhesive properties [5, 6, 11].

Aluminum alloy surface is normally covered with a naturally formed oxide layer on the surface and adsorbed impurities which must be removed to provide a strong adhesive bond. For this purpose, various mechanical, chemical or electrochemical methods of surface treatment are used. These methods not only ensure sufficient surfacing in the geometrical sense by shaping favorable horizontal parameters from the point of view of gluing technology but also change the energy properties of the surface layer of the material. Chemical methods involving digestion in acidic or alkaline baths or the application of adhesive agents in the form of e.g. silicone compounds are not environmentally friendly. Therefore, they require the use of complicated process installations and appropriate procedures for dealing with consumed chemical compounds. All this means that simple, safe and effective methods of improving the quality of adhesive joints are sought after [1, 2, 7, 9, 10, 13, 16].

The aim of the study is to determine whether knowledge of wetting envelopes, representing the limit values of the components of surface free energy of the liquid ensures a good wetting, can be helpful in the design of adhesive joints.

Methodology

The wetting ability analysis of adhesive Epidian 5 with Z1 hardener was performed for EN AW 2017A aluminum alloy sheets subjected to selected machining methods to obtain different surface geometrical parameters.

Samples with dimensions of 25 x 100 mm and a thickness of 2 mm were prepared according to the variants presented in Table 1.

Tab. 1. Variants for the preparation of EN AW-2017A aluminum alloy samples

Variant	Method of treatment
A	Untreated
B	Treatment with non-woven fabric with a grain size of 80
C	Treatment with non-woven fabric with a grain size of 180
D	Treatment with non-woven fabric with a grain size of 320

For this purpose, disks of abrasive material of different grain size were used (Table 1). Each sample was processed according to the scheme presented in Fig. 1. Three alternating tool transitions to the workpiece were carried out.

After the treatment, the samples were cleaned in two stages with a Loctite 7061 degreasing agent. The first stage consisted of washing the samples twice with a degreasing agent and wiping them with a paper towel. In the second stage, after cleaning the samples, the degreasing agent was allowed to evaporate.

The HOMMEL TESTER T1000 contact profilometer was used to measure 2D surface roughness parameters.

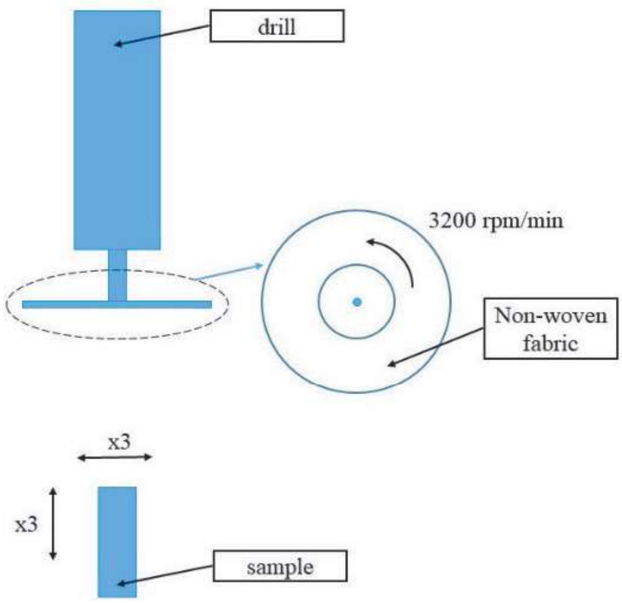

Fig 1. Diagram of processing of EN AW-2017A aluminum alloy samples

The length of the elementary section was selected in accordance with the recommendations of PN-EN ISO 4288: 2011 and equaled 0.8 mm. Measurement of surface roughness parameters was repeated five times for all variants of the treatment.

Surface-free energy and its components, for the treated samples of EN AW-2017A aluminum alloy, were determined using the Owens-Wendt method [17]. On the surface of EN AW-2017A aluminum alloy samples treated with non-woven fabric with different grain size (Table 1), droplets of measuring liquids: distilled water and diodomethane, with a volume of 4 µl, were dosed. The tests were repeated twenty times for each treatment variant. Distilled water is a strongly polar liquid and its value of polar component SEP γ_w^P is 51 mJ/m^2 with the total value of SEP γ_w equal to 72,8 mJ/m^2. Diodomethane, on the other hand, has a polar component of 2.3 mJ/m^2 and a dispersion component of 48.5 mJ/m^2.

On the basis of the surface tension measurements, the components of surface free energy of Epidian 5 resin with hardener Z1 were determined by the hanging drop method. Using a syringe, 20 µl droplets of epoxy adhesive were dispensed and 20 surface tension measurements were carried out. Then, with an NICHIRYO Le-20 automatic pipette, 5 ± 0.02 µl epoxy adhesive drops were placed on the surface of the DC0 steel (the total surface free energy of the value 30,3 mJ/m^2, non-polar component – 28,3 mJ/m^2) and 20 wetting angle measurements were taken. The measurements were carried out at 21°C ± 1.The components of the surface-free energy of the adhesive were determined on the basis of the intersection of the wetting envelope determined on the basis of the contact angle measurements with DC01 steel with a straight line representing the value of the surface tension of the adhesive [6].

Surface tension and contact angles measurements that were used for the calculations were performed on the KRÜSS DSA30 goniometer using an automatic module for obtaining and analyzing the results.

Experimental results

Table 2 presents the average values of measurements of surface roughness parameters of the EN AW-2017A aluminum alloy subjected to the selected methods of surface preparation. The following parameters were registered: Ra parameter - arithmetic mean deviation of the roughness profile and Rz parameter- the maximum height of the profile

Based on the obtained results, it was found that subjecting the samples of the EN AW-2017A aluminum alloy with non-woven fabric with different grain size contributes to the increase of recorded roughness parameters in comparison to the untreated samples. The greatest increase in surface roughness was observed for samples treated with non-woven fabric with a grain size of 80.

The average values of contact angle measurements and components of surface-free energy together with the total value calculated using the Owens-Wendt method for EN AW-2017A aluminum alloy subjected to the selected surface preparation methods are presented in Table 3 and Figure 2.

Tab. 2. The selected 2D surface roughness parameters of EN AW-2017A aluminum alloy samples subjected to the selected surface preparation methods

Variant [1)]		Ra [µm]	Rz [µm]
A	Average value	0,11	0,52
B	Average value	0,59	5,78
C	Average value	0,40	2,79
D	Average value	0,41	3,02

[1)] variant of surface preparation method according to Table 1

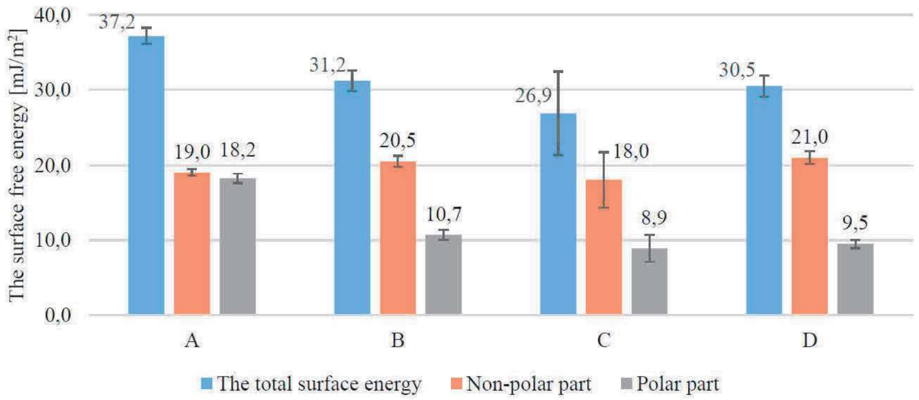

Fig. 2. The results of calculation of surface free energy of EN AW-2017A aluminum alloy (variant of surface preparation method according to Table 1)

Tab. 3. The average value of contact angle and the surface free energy determined by the method of Owens-Wendt together with the components for the EN AW-2017A aluminum alloy samples subjected to the selected surface preparation methods

Variant [1)]	The average value of the contact angle [°]		Owens-Wendt method		
	Distilled water	Diodomethane	The surface free energy [mJ/m²]	Non-polar component [mJ/m²]	Polar component [mJ/m²]
A	66,7	66,5	37,2	19,0	18,2
B	77,2	66,4	31,2	20,5	10,7
C	82,6	72,0	26,9	18,0	8,9
D	78,9	66,0	30,5	21,0	9,5

[1)] variant of surface preparation method according to Table 1

The highest value of total surface-free energy was found for the samples not subjected to mechanical treatment with abrasive nonwoven. It amounts to 37,2 mJ/m^2 with a non-polar component equal to 19 mJ/m^2, whose share in total surface energy is 51.1%. The use of treatment using the non-woven a fiber reduces the total value of the surface free energy. At the same time it can be seen that the change in the share of component values in the total surface free energy compared to the untreated samples. The treated surfaces are characterized by lower values of the polar component of the surface free energy and a smaller share of this component in the total SEP value.

The measurements of surface tension and contact angle during the application of drops of Epidian 5 resin with Z1 hardener on DC01 steel samples were used to determine the components of the surface free energy of the tested adhesive. This adhesive is characterized by a total surface free energy equal to 36,6 mJ/m^2 of a polar component 27,4 mJ/m^2 [6].

Based on the determined work of adhesion curves the values of the maximum work of adhesion between the solid and the liquid that forms on the surface the contact angle of a certain value were also determined. Table 4 summarizes the results of calculations.

Wetting envelopes were determined for the analyzed variants of treatment of EN AW-2017A aluminum alloy, which represent the representation of boundary components of surface free energy of the liquid ensuring good wetting (Fig. 3-Fig. 6) [6]. In the graphs, there was marked the point corresponding to the values of components of the surface free energy of the Epidian 5 resin with Z1 hardener.

The diagrams presented in the drawings show that the best wetting conditions for adhesive Epidian 5 with Z1 hardener were obtained for the surface of the EN AW 2017A aluminum alloy that had not been treated (contact angle θ=19,48°). In the case of mechanical treatment with non-woven fabric with different grain size, the value of the surface free energy and the polar component of the mentioned adhesive designate the points on the

Tab. 4. Summary of the values of the work of adhesion between the surface layer of the EN AW-2017A aluminum alloy and the Epidian 5 with the Z1 hardener adhesive while maintaining a specific contact angle, and the maximum value of the work of adhesion possible to be achieved under certain wetting conditions

Variant [1)]	Contact angle [°]	Ra [µm]	Max. work of adhesion [mJ/m^2]	Adhesion work Epidian 5/Z1/100:10 [mJ/m^2]
A	19,48	0,11	76,59	71,10
B	46,68	0,59	74,02	61,71
C	56,18	0,40	69,13	56,97
D	50,12	0,41	74,33	60,70

[1)] variant of surface preparation method according to Table 1

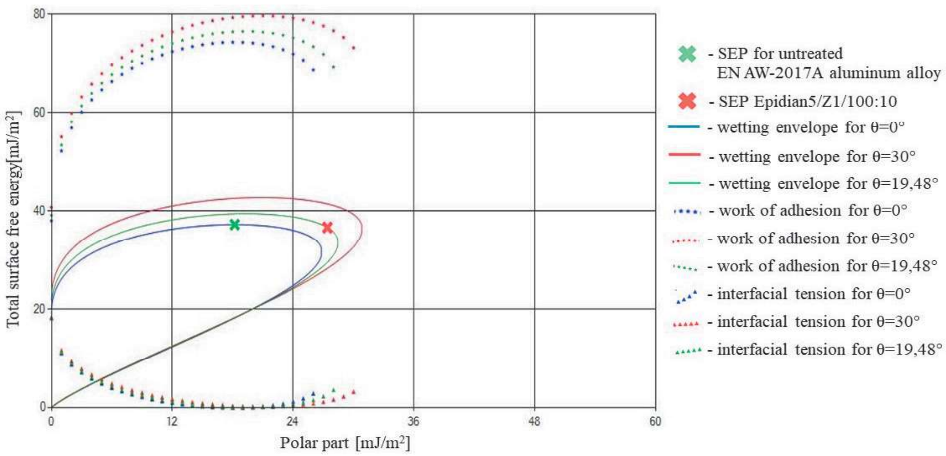

Fig 3. The wetting envelopes, adhesion and interfacial tension curves determined for untreated EN AW-2017A aluminum alloy samples with the marked value of surface free energy of the adhesive Epidian 5 with Z1 hardener

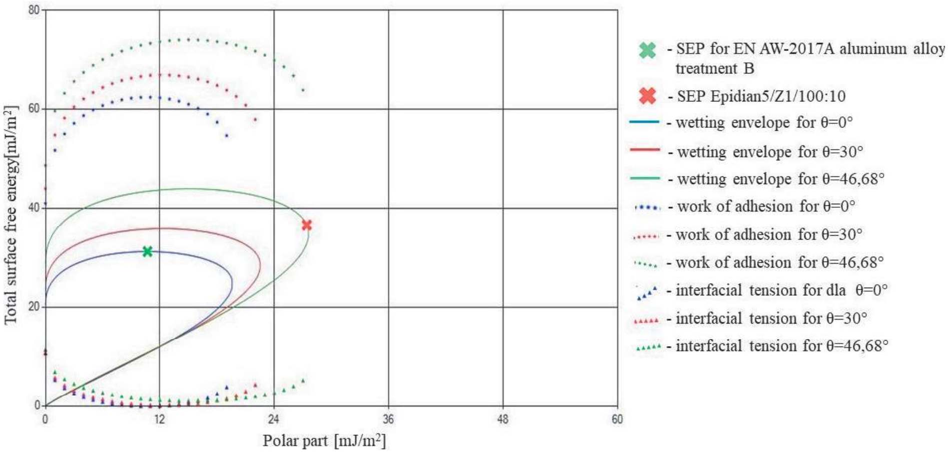

Fig 4. The wetting envelopes, adhesion and interfacial tension curves determined for treated with non-woven fabric with a granulation of 80 EN AW-2017A aluminum alloy samples with the marked value of surface free energy of the adhesive Epidian 5 with Z1 hardener

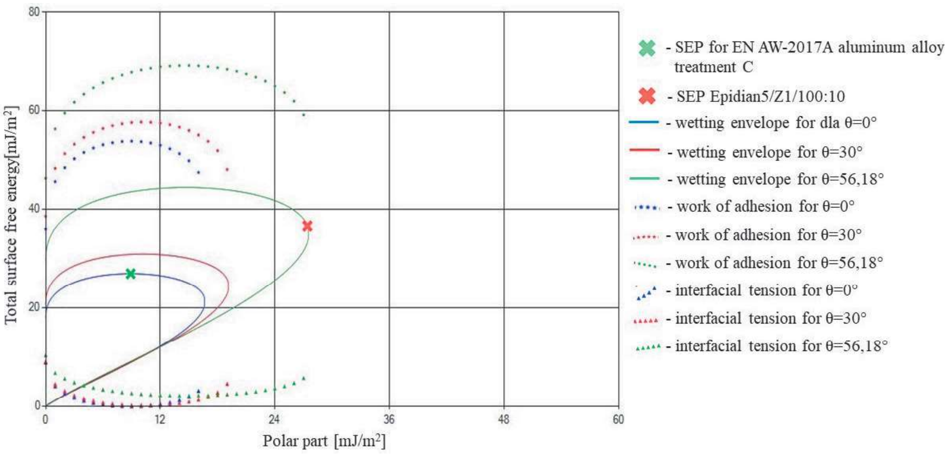

Fig 5. The wetting envelopes, adhesion and interfacial tension curves determined for treated with non-woven fabric with a granulation of 180 EN AW-2017A aluminum alloy samples with marked value of surface free energy of the adhesive Epidian 5 with Z1 hardener

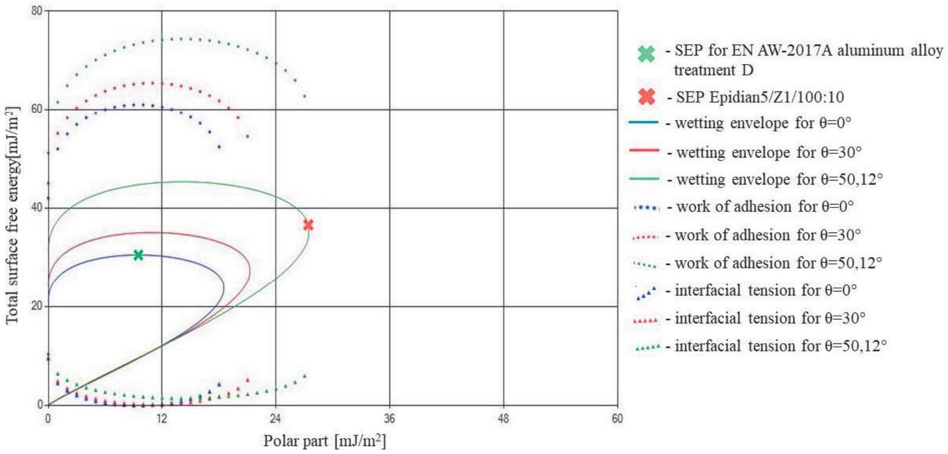

Fig 6. The wetting envelopes, adhesion and interfacial tension curves determined for treated with non-woven fabric with a granulation of 320 EN AW-2017A aluminum alloy samples with marked value of surface free energy of the adhesive Epidian 5 with Z1 hardener

charts that are further away from the wetting envelopes illustrating very good and ideal wetting. Treatment with non-woven fabric adversely affects the surface wettability of adhesive Epidian 5 with Z1 hardener. This is caused by a decrease of the polar component of surface free energy of the treated aluminum, which in turn reduces Van der Waals forces, and thus reduces the value of the work of adhesion due to the increase of the interfacial tension. The values of surface free energy for the aluminum treated non-woven fabric are similar. The change in the strength of connections is therefore here the influence of mechanical and chemical adhesion.

The results of research and analyses lead to the conclusion that the mechanical removal of the oxide layer from the surface of the EN AW-2017 aluminum reduces the surface energy. When comparing variants B, C and D, it can be stated that for the accepted form of machining geometrically developing the surface, there was no effect on the energy properties of the surface layer and the work of adhesion.

Presented at work charts can be helpful for analyzing the possibility of modifying the adhesive in order to achieve, at a given contact angle, the maximum value of the work of adhesion. In the case of the considered methods of preparing the surface of the aluminum alloy EN AW-2017A, the values of the work of adhesion determined for the theoretical contact angles formed between the material and Epidian 5 with the Z1 hardener adhesive do not reach the values corresponding to the maximum. this gives the opportunity to modify the adhesive composition to achieve optimal adhesive properties. At the maximum value of the adhesion work, the minimum value of the interfacial tension is simultaneously achieved. In the case of Epidian 5 with hardener Z1 adhesive, to achieve this state, it was necessary to reduce the polar component of the surface free energy.

Conclusions

The analysis of the wetting ability of the Epidian 5 with hardener Z1 adhesive of the surface of the EN AW-2017A aluminum alloy subjected to the selected methods of treatment in order to obtain different parameters of the geometric development of the surface allowed for the determination of theoretical contact angles between the analyzed adhesive and the material. On the basis of studies it was found that despite the increase in the surface roughness parameters favorable from the point of view of adhesion, treatment with non-woven fabric adversely affects the values of the surface free energy of the EN AW-2017A aluminum alloy as a result of removing the high-energy oxide layer.

The best „matching" of the adhesive and the material was obtained in the case of untreated samples. The points characterizing Epidian 5 with hardener Z1 adhesive are closest to the wetting envelopes corresponding to the ideal wetting angle. In order to ensure the perfect wettability of the EN AW-2017A aluminum alloy surface treated with non-woven fabric, one should strive to reduce the total surface free energy of the adhesive compositions to values close to the value of the surface free energy of the material, with particular regard to its polar component.

The analysis of the surface free energy of epoxy adhesives for a construction material subjected to the selected methods of surface preparation allowed for a quick analysis of the adhesive's ability to wet the surface of materials. Knowing the components of the free-surface energy of the adhesive, it can be determined without the need for destructive testing, whether it will be able to wet the surface of the material well and what value of work of adhesion will be achieved. The presented analysis allows one to determine the adhesive-surface configuration allowing to achieve optimal adhesive properties.

References

[1] Boutar Y., Naïmi S., Mezlini S., Ali M.B.S. 2016. "Effect of surface treatment on the shear strength of aluminum adhesive single-lap joints for automotive applications". International Journal of Adhesion & Adhesives (67): 38 43. http://dx.doi.org/10.1016/j.ijadhadh.2015.12.023.

[2] Budhe S., Ghumatkar A., Birajdar N., Banea M.D. 2015. "Effect of surface roughness using different adherend materials on the adhesive bond strength". Applied Adhesion Science 3(20). https://doi.org/10.1186/s40563-015-0050-4.

[3] Domińczuk J. 2011. "Właściwości adhezyjne warstwy wierzchniej materiałów konstrukcyjnych". Postęp Nauki i Techniki (9): 28 37.

[4] Domińczuk J. 2012. „Wpływ stanu energetycznego warstwy wierzchniej na wytrzymałość połączenia adhezyjnego". Postępy Nauki i Techniki (13): 30 36.

[5] Domińczuk J., Krawczuk A. 2016. „Analiza zdolności kleju do zwilżania powierzchni o określonych właściwościach energetycznych". Technologia i Automatyzacja Montażu (2): 60 64.

[6] Domińczuk J., Krawczuk A., Kuczmaszewski J. 2016. "Energia powierzchniowa wybranych klejów epoksydowych". Technologia i Automatyzacja Montażu (2): 47 52.

[7] Ghumatkar A., Sekhar R., Sandip Budhe S. 2017. "Experimental study on different adherend surface roughness on the adhesive bond strength". Materials Today: Proceedings 4(8): 7801 7809. https://doi.org/10.1016/j.matpr.2017.07.115.

[8] Hu Y., Yuan B., Cheng F., Hu X. 2019. "NaOH etching and resin pre-coating treatments for stronger adhesive bonding between CFRP and aluminium alloy". Composites Part B: Engineering (178): 107478. https://doi.org/10.1016/j.compositesb.2019.107478.

[9] Kłonica M., Kuczmaszewski J. 2011. „Badania porównawcze wytrzymałości na ścinanie zakładkowych połączeń klejowych po oczyszczaniu mechanicznym i ozonowaniu". Technologia i Automatyzacja Montażu (4): 45 48.

[10] Kłonica M., Kuczmaszewski J. 2015. „Badania porównawcze stanu energetycznego warstwy wierzchniej stopu aluminium AZ91HP po obróbce ściernej i frezowaniu”. Mechanik (8-9): 212-216. http://dx.doi.org/10.17814/mechanik.2015.8-9.372.
[11] Krawczuk A., Domińczuk J. 2015. „Analiza możliwości wykorzystania krzywych zwilżania do optymalizacji procesów adhezyjnych”. Technologia i Automatyzacja Montażu (4): 45-49.
[12] Leena K., Athira K.K., Bhuvaneswari S., Suraj S., Lakshmana Rao V. 2016. “Effect of surface pre-treatment on surface characteristics and adhesive bond strength of aluminium alloy”. International Journal of Adhesion & Adhesives (70): 265–270.
[13] Saleema N., Sarkar D.K., Paynter R.W., Gallant D., Eskandarian M. 2012. “A simple surface treatment and characterization of AA 6061 aluminum alloy surface for adhesive bonding applications”. Applied Surface Science (261): 742 748. http://dx.doi.org/10.1016/j.apsusc.2012.08.091.
[14] Wu C., Chen C., He L., Yan W. 2018. “Comparison on damage tolerance of scarf and stepped-lap bonded composite joints under quasi-static loading.” Composites Part B: Engineering (155): 19-30. https://doi.org/10.1016/j.compositesb.2018.08.031.
[15] Zain N.M., Ahmad S., Ali E.S. 2014. “Effect of surface treatments on the durability of green polyurethane adhesive bonded aluminium alloy”. International Journal of Adhesion & Adhesives (55): 43-55. http://dx.doi.org/10.1016/j.ijadhadh.2014.07.007.
[16] Zimniak Z., Wróblewski R. 2018. „Wpływ aktywacji powierzchni aluminium 7075 na wytrzymałość połączenia klejowego”. Przegląd Spawalnictwa 90(2). http://dx doi org/10 26628/ps v90i2 858.
[17] Żenkiewicz M. 2007. „Analiza głównych metod badania swobodnej energii powierzchniowej materiałów polimerowych”. Polimery 52(10): 760-767.

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