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Original Research

ANALYSIS OF THE EFFECT OF ASSEMBLY JOINTS TECHNOLOGY ON THE STRENGTH OF ADHESIVE JOINTS OF POLY(METHYL METHACRYLATE) (PMMA) USED IN ADVERTISING ELEMENTS

ANALIZA WPŁYWU TECHNOLOGII WYKONANIA POŁĄCZEŃ MONTAŻOWYCH NA WYTRZYMAŁOŚĆ POŁĄCZEŃ ADHEZYJNYCH POLIMETAKRYLANU METYLU, STOSOWANYCH W ELEMENTACH REKLAMOWYCH

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Abstract

The aim of the article was to present issues related to the influence of the assembly joints technology (the type of adhesive material and the adhesive application) on the strength of the adhesive joints of poly(methyl methacrylate) (PMMA) used in the production of advertising elements. Angular adhesive joints, which are one of the types of joints made in the construction of advertising elements, were used in the study. The bonded material was poly(methyl methacrylate) (PMMA) of 8 mm thick. Two types of adhesives were used to make the adhesive joints: one-component solvent-based adhesives and methylene chloride as a solvent. Experimental tests determined the wettability of the surface of the adherends (with and without a protective coating) and the work of adhesion based on the measurement of the contact angle, as well as the strength of adhesive joints in accordance with the ISO 4578 standard. Based on the results of strength tests, it can be seen that the use of a solvent allows for obtaining equally strong joints as with the use of solvent-based adhesives. The purposefulness of using coatings protecting not only against dirt or mechanical damage in the form of scratches was also confirmed, but also the possibility of increasing (security) adhesive properties, defined in this case by wettability.

Key words: adhesive joint, angular joint, poly(methyl methacrylate), wettability, contact angle, strength.

Streszczenie

Celem artykułu było przedstawienie zagadnień związanych z wpływem technologii połączeń montażowych (rodzaju materiału klejącego i sposobu aplikacji kleju) na wytrzymałość połączeń klejowych z poli(metakrylanu metylu) (PMMA) stosowanych w produkcji elementów reklamowych. W badaniach wykorzystano połączenia klejowe kątowe, które są jednym z rodzajów połączeń wykonywanych w konstrukcji elementów reklamowych. Łączonym materiałem był poli(metakrylan metylu) (PMMA) o grubości 8 mm. Do wykonania połączeń klejowych zastosowano dwa rodzaje jednoskładnikowych klejów rozpuszczalnikowych oraz chlorek metylenu, jako rozpuszczalnik. W badaniach doświadczalnych określono zwilżalność powierzchni klejonych elementów (z powłoką ochronną i bez) oraz pracę adhezyjną na podstawie pomiaru kąta zwilżania, a także wytrzymałość połączeń klejowych zgodnie z normą ISO 4578. Na podstawie wyników badań wytrzymałościowych można stwierdzić, że zastosowanie rozpuszczalnika pozwala na uzyskanie połączeń równie wytrzymałych, jak przy zastosowaniu klejów rozpuszczalnikowych. Potwierdzono również celowość stosowania powłok zabezpieczających nie tylko przed zabrudzeniami czy uszkodzeniami mechanicznymi w postaci zarysowań, ale także w celu zwiększenia (zabezpieczenia) właściwości adhezyjnych, określanych w tym przypadku przez zwilżalność.

Słowa kluczowe: połączenie adhezyjne, połączenie kątowa, poli(metakrylan metylu), zwilżalność, kąt zwilżania, wytrzymałość.

1. Introduction

Poly(methyl methacrylate) (PMMA) is a polymer material that is used in many areas of industry, including in the automotive, construction, medical and advertising industries [1,2]. This material is characterized by good mechanical properties and low toxicity. In addition, it is one of the hardest thermoplastics, resistant to UV. Poly(methyl methacrylate) (PMMA) is used for various types of advertising panels, spatial letters and forms, illuminated boards or spatial forms. PMMA is also used in the construction industry, the aviation industry (aircraft windows), the food industry, the automotive industry (e.g. license plates, headlight elements) and others [3]. Among the various types of assembly joints in advertising elements, angular connections can be distinguished (Fig. 1), e.g. in various types of stands.

leaflet racks | accessories for displays and stands | holders for shelfstoppers | other POS accessories

Figure 1. Examples of angular joints in the advertising industry [4]

Poly(methyl methacrylate) (PMMA) is available in many colors and textures, which allows to choose according to aesthetic requirements. In the production process, many plates made of poly(methyl methacrylate) are coated with a scratch-resistant coating [3].

One of the types of PMMA assembly joints is bonding [5], although this material is difficult to bonding due to its chemical resistance (and for this reason it is often used in elements of medical equipment). But, among others due to the requirements for the aesthetics and transparency of the joining point, this method of the joints is often chosen. Adhesives used to join metals and other polymer materials do not show strong adhesion to PMMA.

Solvent adhesives used to join PMMA are usually chemically aggressive adhesives, as they contain dichloromethane. They have a low viscosity and are very volatile, i.e. they produce a fairly high vapor pressure, and for this reason, dosing, e.g. with a syringe, causes the adhesive to constantly drip from the dispensing tip. For this reason, they can leave residues on the surface of PMMA components, and this material is often used for aesthetic reasons. Therefore, it is important to adopt an appropriate process for applying the adhesive to the adherends. These adhesives join the elements by partial dissolution of the surface layer of the surface of the element and evaporation. Thanks to this, the assembly joints are joints created using the phenomenon of diffusion [2,6].

The aim of the conducted research was to evaluate the technology of preparing the adhesive joint, and in particular the application of the adhesive on the joined surfaces, in terms of the adhesive joints strength determined by the failure force and to determine the wettability of the surfaces of the adherend.

2. Materials and methods

The subject of the research were poly(methyl methacrylate) (PMMA) angular adhesive joints, which are one of the types of assembly joints made in the construction of advertising elements. The experimental tests carried out consisted in determining the wettability of the surfaces of the joined materials (with and without a protective coating), making angular adhesive joints and subjecting them to strength tests.

2.1. Adherend

The samples that were subjected to the bonding process were made of poly(methyl methacrylate) (PMMA) with the following dimensions 8 x 30 x 50 mm, which were then subjected to the bonding process. The thickness of the elements resulted from the thickness of the materials used in the company for advertising construction elements. PMMA samples were made of a colorless plate produced by casting in special moulds. The product obtained in this way has lower internal stresses and is of better quality than extruded boards. In addition, cast plates are more chemically resistant, have a harder, more even surface and better optical properties [3]. The plate from which the samples were made had a high transparency of about 92%.

Table 1. Selected properties of samples made of cast PMMA plates [7]

Properties	Casting board
Density (max) g/cm^3	1.19
Rockwell hardness, M scale	95
Young's modulus, min., MPa	3300
Relative elongation at break, min., %	5.5
Thermal expansion coefficient, mm/m/°C	0.065

2.2. Preparing conditions of adhesive joints

2.2.1. Surface treatment method

Bonding surfaces must be dry and clean, especially free of oil, grease or release agents [2]. Another aspect in the surface treatment of this adherend is its roughness. This material is characterized by a small surface roughness, so the share of mechanical anchors of the adhesive practically does not affect the strength of the adhesive joints. For aesthetic reasons, in many cases it is not possible to apply mechanical treatment of the surface. The influence of surface roughness on the strength of joints has been described in numerous works [8-13]. Therefore, in this case, it is necessary to focus on the use of the appropriate type of the adhesive, which allows the connection to be made using partial dissolution of the surface of the joined material and the phenomenon of diffusion.

The surface of the joined samples was covered on both sides with a special protective coating. Due to the fact that it should remain on the product as long as possible (as the surface of the joined material is susceptible to scratches and dirt), it was removed just before the bonding process. Considering the material properties, the surface of the PMMA samples was only wiped with a dry cloth. No degreasing agent, e.g. based on alcohol, was used, because the use of alcohol could damage the surface of this material.

2.2.3. Adhesive joint and adhesive

The subject of the tests was angular adhesive joints of the polymer material, which was PMMA with a thickness of 8 mm, the diagram of which is shown in Figure 2. The samples were laser cut.

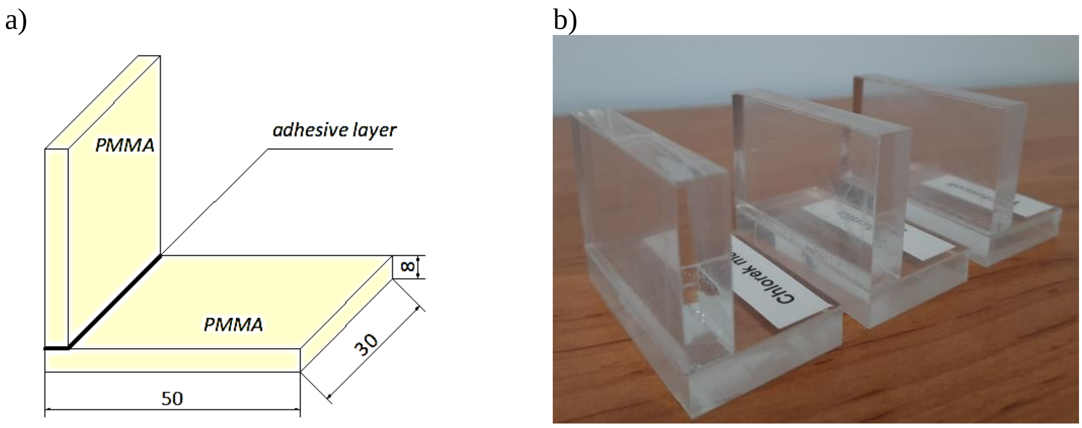

Figure 2. Angular adhesive joints: a) scheme (dimensions in mm), b) real view

Two types of adhesives were used to make three series of adhesive joints (5 joints in each series):

- one-component solvent transparent adhesive ACRIFIX 1S 0116 (Röhm GmbH Acrylic Products, Weiterstadt, Germany)
- low-viscosity solvent-based adhesive Technicoll® 108 (RUDERER KLEBETECHNIK GnbH, Zorneding, Germany),

and methylene chloride (dichloromethane, CH_2Cl_2, distributor: STANLAB, Lublin, Poland), as a solvent (Table 2).

Joints made with the use of the adhesive were an example of adhesive joints, while with the use of solvent - adhesive joints, but made taking into account the phenomenon of diffusion.

Methylene chloride (dichloromethane) is an organic chemical compound from the group of alkyl halides; chlorine derivative of methane. At room temperature and atmospheric pressure, it is a colorless, rapidly evaporating liquid. It has a characteristic, sweetish smell. Methylene chloride (dichloromethane) is the base product of many solvents supporting the removal of e.g. old paint coatings from metals, wood, stone, etc. Methylene chloride is used as a very effective solvent and diluent [14].

Table 2. Selected properties of methylene chloride [based on the safety data sheet]

Properties	**Date/ description**
Form	colorless liquid
Composition	dichloromethane
Chemical formula	CH_2Cl_2
Molar mass	84.93 g/mol
Solubility in water	16 g/dm^3 (20˚C)
Melting temperature	– 95˚C
Boiling point	39.5 – 40.5˚C
Temperature of self-ignition	605˚C
pH value	neutralne
Density	1.326 g/cm^3 (20˚C)

The listed solvent adhesives, according to the manufacturers' catalog information, are dedicated to making angle joints. Selected properties of the adhesives used are listed in Table 3.

When making adhesive joints with these adhesives, it is recommended that the pieces to be bonded fit snugly together, as small gaps will not be filled.

ACRIFIX® 1S 0116 was applied from a nozzled bottle to the edge of one of the items to be bonded, which, starting from one side, is then placed in contact with the second. After a short holding time, the bond is locked in position. Slight pressure applied to the bonded surface during drying also reduced bubble formation.

In the case of Technicoll® adhesive, the joining technology consisted in immersing the edge of one of the adherend in the Technicoll® adhesive, and after 5 to 10 seconds of surface impact (etching), the bonded elements were joined and pressed. This is one of the recommended, depending on the size of the surfaces of the elements to be joined, variants of the joining technology with the use of this group of adhesives.

Table 3. Selected properties of solvent adhesives [15]

Properties	**Solvent adhesive**	
	ACRIFIX 1S 0116	**Technicoll® 108**
Chemical base	contains ethylformate, nitroethane, butan-1-ol	dichloromethane: a solution of acrylic polymer in a mixture of solvents
Viscosity at 20°C, mPa·s	650-900	5
Density at 20°C, g/cm^3	1.0	1.3
Refractive index, n_D^{20}	~1.39	
Color	clear to yellowish; color does not affect bonding properties	colourless change of colour does not influence characteristics of adhesion
Flash point (DIN 53213)	< 4°C	
Curing	physically, through evaporation and absorption in the bonded articles	
Wet-bonding time		approx. 5-10 seconds (one-sided application)
Thickener	approx. 10%	flakes of polycarbonate (max. 15%)

The solvent will usually evaporate residue-free. Under best circumstances the resistance to temperature is similar to the one of the feedstock. Because variation may occur during the bonding process, the resistance to temperature may be lower.

The joint was made by pressing the edges of two PMMA specimens together in a previously prepared fixture, containing a wooden stop in the form of an angle bar. The upper sample was pressed against the lower with a small steel weight (>200g). Joining took place at room temperature (approx. 20°C) with air humidity (40-60%). The curing time in the methylene chloride mixture was in the range of 30-60 seconds at room temperature (approx. 20°C).

2.3. Research description

The experimental studies carried out included:

a) determining the wettability of the surface and the work of adhesion (Wa) based on the measurement of the contact angle,
b) performance of strength tests.

Determination of surface wettability and work of adhesion concerned poly(methyl methacrylate) (PMMA) samples with and without a protective coating. Strength tests included tests of angular joints made with two types of the solvent adhesives and with the use of a solvent.

Wettability of the surface of poly(methyl methacrylate) samples and the work of adhesion were determined on the basis of direct measurement of the contact angle (Θ) with distilled water. There were 3 samples per each batch: uncoated and adhesive coated.

The work of adhesion was calculated on the basis of the value of the contact angle using the formula (1) [6]:

$$W_a = \gamma_L (1 + \cos\Theta) \quad (1)$$

where the symbol γ_L denotes the surface free energy of the wetting liquid.

The contact angle Θ of the surface of the epoxy coatings was measured using a contact angle analyzer - Phoenix 150 goniometer with Surfaceware 7 software. Work of adhesion (Wa) value Wa using Surfaceware 7 calculation options.

Drops of the measuring liquid were applied to the analyzed substrate using a micropipette mounted above the tested plate. The volume of drops of the measuring liquids ranged from 5 to 7 μl. Five drops of the distilled water were applied to the surface of the tested samples. The picture of the drop was taken with a digital camera with the lens placed parallel to the tested substrate. After that, 10-13 measurements were made and the mean per each sample batch was calculated. The contact angle was measured immediately following the application of a drop of the measuring liquid (after a few seconds). The measurements of the contact angle were performed at a temperature of 21±1°C and air humidity of 28±2%. Before contact angle measurements, the adherends surfaces were wiped with a dry cloth.

Adhesive joints were subjected to strength tests, in accordance with the ISO 4578 standard, on the Zwick/Roell Z150 testing machine, using the appropriate fastening in the machine's screw-wedge holders.

3. Results

3.1. Results of contact angle measurements, determination of adhesion work and assessment of surface wettability

The results of the contact angle measurements and calculated of work of adhesion are presented in Tables 4 – 7 and Figure 3.

Table 4. Measurement results of contact angle and calculation work of adhesion for samples without protective coatings – without degreasing

Type of measuring liquid	Measurement series number	Contact angle [°] (average of 10-13 measurements)	Work of adhesion [mN/m]
Distilled water	1	52.25	117.34
	2	55.89	113.56
	3	51.04	117.79
Mean		53.06	116.23
Standard deviation		2.52	2.32

Table 5. Measurement results of contact angle and calculation work of adhesion for samples without protective coatings – with degreasing

Type of measuring liquid	Measurement series number	Contact angle [°] (average of 10-13 measurements)	Work of adhesion [mN/m]
Distilled water	1	66.17	101.47
	2	62.09	106.77
	3	63.12	106.64
Mean		63.79	104.96
Standard deviation		2.12	3.02

Table 6. Measurement results of contact angle and calculation work of adhesion for samples with protective coatings – without degreasing

Type of measuring liquid	Measurement series number	Contact angle [°] (average of 10-13 measurements)	Work of adhesion [mN/m]
Distilled water	1	66.53	102.36
	2	63.04	105.59
	3	68.00	99.96
Mean		65.86	102.64
Standard deviation		2.55	2.83

Table 7. Measurement results of contact angle and calculation work of adhesion for samples with protective coatings – with degreasing

Type of measuring liquid	Measurement series number	Contact angle [°] (average of 10-13 measurements)	Work of adhesion [mN/m]
Distilled water	1	59.28	110.01
	2	63.61	105.10
	3	57.10	111.80
Mean		60.00	108.94
Standard deviation		3.31	3.52

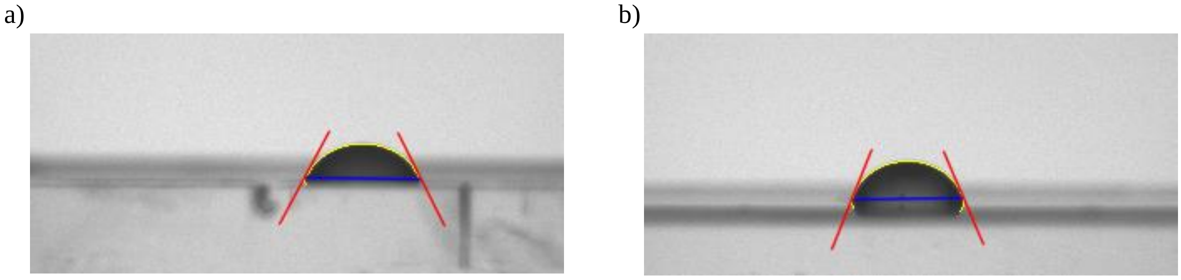

Figure 3. Contact angle of the surface of the samples (without degreasing): a) without protective coatings, b) with protective coatings

When analysing the results presented in tables 4-7, the following dependencies were observed:

1) uncoated samples are characterized by the lowest value of the contact angle (average value), amounting to 53.06°, hence they are characterized by the highest wettability and the highest work of adhesion (Wa is 116.23 mN/m),

2) the use of degreasing contributes to an increase in the value of the contact angle by about 20%, which means a deterioration of the surface wettability and a decrease in the value of the work of adhesion by about 10%,

3) samples with a protective coating obtained a greater value of the contact angle than without the coating by nearly 25%, which means that they have lower surface wettability.

Due to the fact that the protective coating was removed just before the contact angle measurements, there was no danger of soiling or reducing the energy activity of the surface. It was noticed that the protective coating itself is covered with impurities, because degreasing even this coating contributes to the increase of its wettability.

3.2. Strength test results

A comparison of the failure force of the analyzed variants of PMMA adhesive joints is shown in Figure 4 and Figure 5.

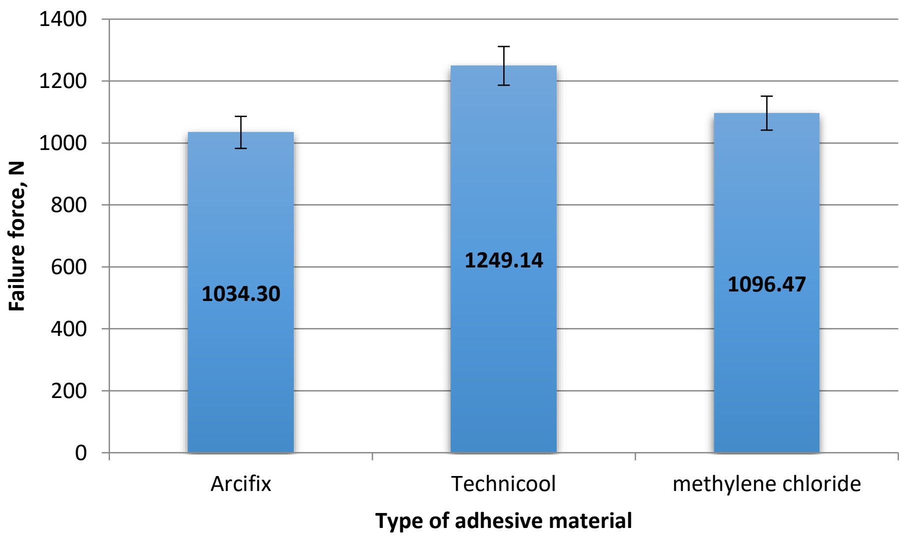

Figure 4. The value of the failure force of PMMA angular assembly joints

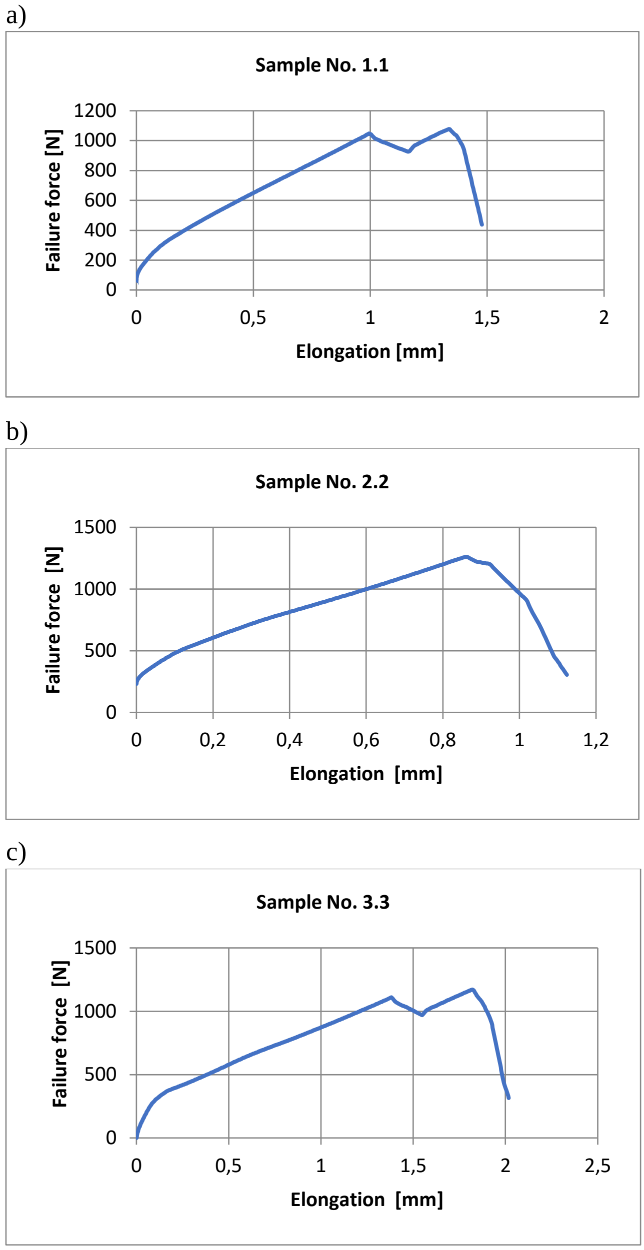

Figure 5. Examples of the elongation-failure force relationship for assembly joints made with: a) Acrifix adhesive, b) Technicool adhesive, c) methylene chloride solvent

On the basis of the obtained results (Figure 4) it was noticed that the highest value of the failure force was characteristic for the adhesive joints made with the use of one-component solvent adhesive Technicool (1249.14 N). On the other hand, joints made with the second Arcifix adhesive (also one-component solvent) had about 17% lower strength. Both adhesives used are one-component solvent adhesives, dedicated to joining polymeric materials and to making angular and T-shaped joints. It can be assumed that both the chemical composition and the assembly technology recommended for a given type of adhesive resulted in differences in the value of the failure force.

The joints made with methylene chloride solvent had a lower strength (1096.47 N) by about 12% than the highest value obtained in the case of the adhesive joints made with the one-component solvent adhesive Technicool (1249.14 N). Both types of assembly joints required the use of adhesive materials, which allows to obtain a transparent joint, which is important when making advertising construction elements.

Relationships between elongation-failure force (Figure 5) for individual types of assembly joints are similar.

In addition to the type of adhesive material in the work [5] it was noted that the decisive factor, among others, the value of the failure force of samples of PMMA and PVC adhesive joints are the properties of the bonded material.

3.3. Basic descriptive statistics

A basic descriptive statistics was presented in Table 8 and it was performed on the results from the strength test.

Basic statistics is the basis for further analyzes and inferences. Based on the data presented in Table 8, one can notice the repeatable accuracy of making adhesive joints, regardless of the adhesive material used.

Table 8. Parameters of descriptive statistics of the joints failure force

Type of adhesive material used to make PMMA joints	**Basic descriptive statistics**									
	No. of tests	Mean	Median	Mode	Mode size	Range	Variance	Standard deviation	Skewness	Kurtosis
Acrifix adhesive	15	1034.30	1035.16	multiple	1	25.63	69.0465	8.3094	-0.0369	-1.2816
Technicool adhesive	14	1249.14	1249.64	multiple	1	25.39	71.2871	8.4432	-0.1391	-1.2910
methylene chloride solvent	17	1096.47	1096.80	multiple	1	25.26	70.2013	8.3786	-0.0136	-1.2748

4. Conclusions

Based on the presented results, the following observations can be made regarding individual groups of studies:

1) based on the results of the measurement of the contact angle and the assessment of wettability, it was noted that:

- the surface of the element made of poly(methyl methacrylate) (PMMA) without the protective coating is characterized by greater wettability (smaller value of the contact angle) than the surface of the samples with the protective coating,
- due to the fact that the protective coating was removed just before the contact angle measurements, there was no risk of soiling or reducing the energy activity of the surface,
- it was noticed that the protective coating itself is covered with impurities, because degreasing even this coating contributes to the increase in its wettability,
- it can be confirmed that the use of coatings protecting not only against dirt or dust, or protection against mechanical damage in the form of scratches, but also the ability to protect and increase adhesive properties, defined in this case by wettability, can be confirmed.

2) the obtained results of the failure force of the angular joints allow to assume that:

- both the adhesive application technology and the type of adhesive will affect the strength of the joints,
- the use of a solvent allows for an equally strong joints as with the use of solvent adhesives (with a failure forces comparable to adhesive joints made by some solvent adhesives).

The use of solvent adhesives and solvents is one of the methods of making assembly joints of poly(methyl methacrylate). The surface of elements made of poly(methyl methacrylate) (PMMA) is characterized by very low roughness, therefore the use of the assumptions of the theory of diffusion adhesion in this case becomes significant. The small surface roughness does not allow for effective joints of such elements, because the phenomenon of mechanical adhesion will not occur in the general component of the adhesion, due to the lack of possibility of mechanical anchoring of the adhesive. Therefore, the use of solvent adhesives or solvents allows for effective joining of such elements, although, as it results from the research, it is necessary to analyze, among others, adhesive application technology, resulting from the properties of the adhesive, including its viscosity.

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