Significance of Adhesion Theories in Area of Flexible Bonded Structural Joints in Construction Sector

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Abstract. The issue of bonding has been a part of humanity throughout its history. The greatest development in this area occurred in the second half of the 20th century and the issue of bonding has gained a lot of interest from the scientific community. Over the past 80 years, there has been a significant increase in scientific research and grants aimed at defining adhesion. We now know that examining the adhesion of materials, determining the adhesive properties or simply evaluating the test results and assessing the failure of the test specimens is not possible without understanding the basic principles and theories of bonding, i.e. adhesive joining. The presented paper is focused on the description of fundamental adhesion theories and their usage in the field of structural bonding in construction industry. The importance of understanding to adhesive properties of used products is demonstrated on an example of four different surfaces in combination with representatives of high strength flexible adhesive systems intended for façade applications. Representatives with high surface polarity, medium-high polarity and low polarity were deliberately selected. The one-way ANOVA was performed to analyse the impact of surface adhesive properties on adhesion of bonded joints. It was confirmed that the riskiest is bonding of polymer-based materials. For all selected materials it was concluded that the hypotheses of the adsorption theory, which, to some extent, also includes the assumptions of other adhesive theories, seem to be the most fundamental for the presented researched area.

1. Introduction

The issue of bonding has been a part of humanity throughout its history. While by the beginning of the 20th century, all adhesives used until then were entirely natural-based, only with the end of the second wave of the industrial revolution came the first adhesive based on synthetic polymers. As reported by Petrie [1], nowadays, it is almost impossible to find a product commonly used in households, industry, transportation, or anywhere else that does not contain adhesive in any form. We now also know that examining the adhesion of materials, determining the adhesive properties, or simply evaluating the test results and assessing the failure of the test samples is not possible without understanding the basic principles and theories of bonding because adhesion of materials is not only related to adhesive properties but also to the resistance of the bonded surface to mechanical stresses [1-3]. The durability/strength and lifespan of a bonded joint is most often affected by four basic parameters [2-4]:

- adhesion of the adhesive to the adherend;
- adhesive properties of the adherends;
- cohesion of the adhesive; and
- durability/strength of the adherends, as the bonded joint is as strong as the adherends are resistant.

The main prerequisite for a perfect joint of two adherends is their ideal adhesive properties [1-5]. As mentioned above, scientists have been studying the phenomenon of adhesion in modern history since the 1920s [3,6], and more attention has been paid to this issue since the 1940s [1]. That is primarily why we can use several theoretical models to define the bond of the adhesive and the adherends. The so-called traditional ones include mechanical, electrostatic, diffusion and adsorption theories, see Table 1 [3,6]. It is obvious that the number of theories has been gradually expanded, or more precisely that some methods has been elaborated in more detail with the development of technologies and new materials. However, the approach to the assessment of adherends has remained almost unchanged. The assessment of the adherend based on the use of only one theory is insufficient, as the definition of adhesion is based on the search for interdisciplinary connections.

Table 1. Overview of basic adhesion theories [3,6].

Traditional Theories	Current Theories	Scale of Assessment
Mechanical	Mechanical	microscopic
Electrostatic	Electrostatic	macroscopic
Diffusion	Diffusion	molecular
Adsorption	Wettability	molecular
	Chemical	atomic
	Weak – Boundary – Layer	molecular
	Acid – Base	molecular

1.1. Mechanical theory
This is one of the basic theories of adhesion which was introduced by McBain and Hopkins around 1925 [7]. This theory is applicable mainly to porous materials, as it is based on the assumption that the surface of each material is wrinkled in some way which allows the adhesive to penetrate better into all gaps and irregularities. Thus, the theory works better for rough or additionally roughened surfaces while it is not applicable to smooth surfaces on its own. As mentioned above, the properties of the joint are not only affected by the adhesive properties of the adherend but also by the properties of the adhesive itself. It is therefore necessary to use a high viscosity adhesive to fulfil the mechanical theory hypothesis. Part of the presented paper is devoted to the verification of the hypothesis of this theory, when materials with different degrees of porosity and with different surface polarity were selected for this purpose.

1.2. Electrostatic theory
The use of this theory is not so common [1,6,8-10], as it assumes that electrostatic discharges occur between the adherend and the adhesive. These provide a stable joint, which can only be unintentionally broken in the event of depolarization or forcible tearing off of the surfaces. As stated by Derjaguin et al. [10], the potential of the theory is used mainly in the bonding of polymers and sheets, and, contrarily, in the case of other materials, the results of the theory are negligible, because there is not a sufficiently strong electrostatic charge between the surfaces.

1.3. Diffusion theory
This theory is especially applicable to polymer-based materials, as it is based on the hypothesis that the joint strength can be increased by mutual diffusion (interdiffusion) of the adherend and the adhesive. Thus, the more similar the chemical composition of the adherends, the stronger the joint should be [1-2,6,9]. The mutual diffusion of materials usually does not occur spontaneously, but it is necessary to use aids. This, for example, is a usage of a primer before applying the adhesive.

1.4. Adsorption theory

Currently, more adsorption theories are used (see Table 1), but all presume that the basis of adhesion is the interaction of the molecules of the adherend and the adhesive [4].

1.4.1. Surface wettability theory. The main prerequisite for perfect adhesion is the creation of a bond forces, i.e. interaction of the adherend and the adhesive through mutual contact [6,8-9]. This process is called surface wettability because the adhesive must be able to wet the entire adherend. The hypothesis of this theory is similar to the assumption of the mechanical theory, where the adhesive penetrates into all the gaps of the surface and, after curing, forms a non-detachable joint.

1.4.2. Chemical theory. It is necessary for the adhesive and the adherend to interact with each other to form a firm joint. This interaction occurs through the formation of primary chemical bonds. While according to Ebnesajjad [6], these are covalent, hydrogen, Lifshitz – van der Waals forces and the interaction of acid bases, according to Mittal and Pizzi [9], it can only be the action of covalent bonds, the strength of which is more than tenfold. However, the chemical bond between the adhesive and the adherend does not occur without the existence of interacting chemical groups. For some materials, the interaction occurs naturally, sometimes it is necessary to modify the adherend, for example by applying a primer or deep impregnation of the surface, or by choosing another, more effective, adhesive. It is obvious that the main hypothesis of this theory is similar to the assumptions of the mechanical theory and the surface wettability theory.

1.4.3. Theory of Weak – Boundary – Layer. The idea of this theory was first described by Bikerman [2-3,6-7,9] and accepts that the failure of the bonded joint occurs either due to poor cohesion or due to poor adhesive properties of the top layer of the substrate. This theory is directly related to the surface wettability and can be divided into three stages: The first stage occurs before the implementation of the bonded joint. Suitable examples are metals [6,9], in particular, for example, aluminium or copper, which oxidize on contact with air and form a thin layer of aluminium oxide or copper carbonate on their surface. Although this layer protects the surface from continued oxidation, it completely degrades its adhesive properties. For this reason, it is necessary to remove this layer before bonding. Another material that requires surface treatment are polymers (plastics), the surface energy of which is usually very low, but can be increased by a suitable treatment method; The second stage is related to the drying and curing of the adhesive. During this phase, the adhesive is exposed to the surrounding environment, which, by its aggressiveness, can significantly affect the properties of the marginal layers of the joint; The last stage is the life of the bonded joint. During its life, the joint is exposed to the environment as well as to the effects of, for example, paints, cleaners, etc., which often reduce the strength and adhesion of the marginal layers [9], and, at the same time, also the effect of weather conditions is involved.

2. Materials and methods
2.1. Selection of adherends

As already mentioned in the previous chapter, the strength and lifespan of the bonded joint is mainly influenced by the choice of a suitable adherend material and the adhesive system. Bonded substrates are exposed to various stresses during their life, or during the life of the joint, which they must safely withstand. Each material has specific properties such as strength, elasticity, toughness, etc. However, for many materials, these properties are affected by changes in the temperature or humidity of the environment, which brings a secondary stress into the joint [11]. Even though, the bonded joint is able to significantly eliminate these stresses frequent changes can cause fatigue and damage [2]. Three representatives of porous materials and one representative of non-porous materials were selected. These namely include Cetris Basic (CB) cement-bonded particleboard, solid wood of Larix sibirica (SW), solid three-layer board panels (TLB), and wood-plastic composite (WPC). At the same time, these are materials with high as well as low polarity of the surface that significantly affects the

adhesive properties, which is suitable for the study of the area. The basic material characteristics of the selected adherends relevant to the researched area are listed in Table 2.

Table 2. Overview of relevant material properties - adherends.

Material characteristic/ Adherend	CB	SW	TLB	WPC
Density [kg/m3]	1,275.0	640.0	850.0	1,250.0
Modulus of elasticity [MPa]	6,800.0	7,400.0	13,800.0	5,500.0
Tensile strength perpendicular to the board's plain [MPa]	0.63	7.8	1.5	N/A

2.2. Selection of adhesives
When choosing an adhesive or an adhesive system, it is necessary to consider the suitability for a specific application. The choice may also be influenced by the viscosity, homogeneity and thickness of the applied adhesive layer. The selection of a suitable adhesive is one of the basic preconditions for a strong and durable joint [2].

The presented paper is focused on facade applications therefore the use of structural adhesive is necessary. There are a number of adhesives produced by various manufacturers that can potentially be used for structural façade joints. Perhaps the best known are silicone sealants, which are most often used when it is necessary to create a flexible structural joint between glass and aluminium or steel, or between glass and glass [12-15]. Structural silicone joints are relatively thick and flexible, which allows them to adapt to different thermal stresses between glass and metal. However, due to their low resistance, structural silicones are not suitable for transferring the higher shearing stress required for some types of materials, such as ceramic or composite cladding. Therefore, polymer-based structure adhesives are currently used in this field. In particular, these are polyurethanes and modified polymers, which have similar tensile properties as silicones, but at significantly higher stresses. For this purpose, two representatives of the polyurethane group from two different manufacturers and two representatives of the silyl-modified polymers group from the same manufacturer were selected. These are products suitable for the implementation of structural joints with high flexibility, which are part of adhesive systems. The use of a system cleaner was necessary for all selected representatives, as the cleanliness and subsequent wettability of the contact surfaces are one of the basic parameters that has a significant effect on the strength and life of the bonded joint. It was also necessary to use a primer. Selected material characteristics of the used systems are listed in Table 3.

Table 3. Overview of relevant material properties - adhesives.

Adhesive/ Material characteristic	PU/1	PU/2	MS/1	MS/2
Density [kg/m3]	1,180	1,200	1,450	1,500
Tensile strength [MPa]	2.50	9.00	2.30	1.80
Shear strength [MPa]	2.00	5.50	2.00	2.25
Elongation at Break [%]	c. 500	c. 600	c. 400	c. 330

2.3. Methods
As already mentioned in the Adsorption theory section, the critical surface energy or surface tension affects the bonding potential of the selected adherend. This property is called the surface polarity of substances. The higher the surface tension value, the more polar the surface is. Therefore, it is suitable to use an adhesive with a lower polarity than the adherend has to make the bonded joint. In this case, the contact surface of the adherend will be wetted. In some cases, especially regarding plastics, it is necessary to increase their polarity by modifying the adherend [4]. As some selected manufacturers recommend a modification of the adherend and others do not, an assessment of the wettability of the adherend was performed prior to the production of test samples for adhesion assessment purposes. The output of the test method is to determine the size of the contact angle α. If the surface energy of the

solid is greater than the sum of the surface energy of the liquid and the solid-liquid interfacial energy, the liquid spreads over the surface of the solid in a continuous layer. The smaller the contact angle observed, the better the wettability of the surface [3,13,16-17]. If spreading does not occur and a drop of liquid assumes an equilibrium shape on the surface of the solid, it is possible to measure the contact angle and define the wettability of the surface. An assessment of both unmodified and modified surfaces was performed, when a mechanical treatment by sandpaper with grit size P80, which is recommended by the manufacturer of the PU/1 system, was performed.

Another parameter that significantly affects the strength of the joint is the selected geometry. A suitable example to demonstrate this statement is a single lap joint, as its shear strength is directly proportional to the width of the joint. The wider the joint, the greater the forces it can withstand [8]. On the other hand, if the length of the overlap were to increase, the increase in strength would not be entirely guaranteed, as it is no longer a directly proportional relationship [2]. Therefore, it is not appropriate to estimate the failure of another joint design based on the results of one type of geometry. Each joint should be designed and assessed for the specific method of adhesive application. For this purpose, a test method defined as the so-called pull-off technique was chosen, which is frequently used for the evaluation of adhesion strength. The adhesive was applied between the examined adherend and a square disc with (50 ± 0.1) mm side, therefore, the size of the studied area was approx. 2,500 mm^2. The adhesive in the thickness of 3 mm, as recommended by manufacturers, was applied to surface already treated with a primer. The test samples were kept in a clean environment with air temperature (23 ± 2) °C; humidity (50 ± 5) % and air circulation ≤ 0.2 m/s for 14 days. Subsequently, testing was performed on the mechanical press Heckert FP 10/1 in similar boundary conditions. The pull-out speed was set to (250 ± 50) N/s and at least 6 samples represent one set. All samples with values outside the range $\pm 20\%$ of the average were removed. The analysis of variance (one-way ANOVA) was performed to evaluate the recorded data and to determine the significance of surface modification on adhesion. The value of maximum load was used as a response and a significance level of $p = 0.05$ was considered.

3. Results and discussion

3.1. Contact angle measurement and surface wettability analysis

Many technological processes depend on how well the liquid wets the surface of the solid. This ability is quantified by assessing the wettability of selected adherends, or more precisely by measuring the contact angle. Measurements are made to better understand the interactions between the solid and the liquid. For the presented purposes, the method of static measurement of the contact angle of the drop defined in ČSN EN 828 [18] was chosen. We usually encounter several degrees of wettability; if the angle $\alpha \geq 90°$, the wettability of the observed surface is insufficient, otherwise the wettability is sufficient and no additional surface treatment is necessary. Measurements were performed on 3 test samples of each adherend. Distilled water was used as the measuring liquid. Drops of measuring liquids were applied with a micropipette in the form of drops with a volume of 3–6 μl. In general, the wetting liquid must not react with the measured surface, otherwise the results would be significantly skewed and therefore not usable for further evaluation. The arithmetic mean, including the standard deviation, was determined from the measured values, see Table 4. Graphical outputs of the surface wettability assessment by the sitting drop technique are visible in Figure 1. The measured values of the contact angle of wood cladding, i. e. three-layer glued board and solid wood, show that the wettability of these surfaces is sufficient even without surface treatment, so it can be assumed that adhesion of these surfaces will not be the main reason for possible premature joint failure. On the contrary, the surfaces of the cement-bonded particleboard and the wood-plastic composite are poorly wettable and their treatment is necessary to improve the adhesive properties.

Table 4. Assessment of wettability of the surface of façade cladding without surface treatment.

Adherend	Contact angle α [°]	Wettability assessment [-]
CB	107.98 ± 2.38	poorly wettable surface
SW	69.21 ± 8.76	sufficiently wettable surface
TLB	83.29 ± 6.95	sufficiently wettable surface
WPC	118.77 ± 1.27	poorly wettable surface
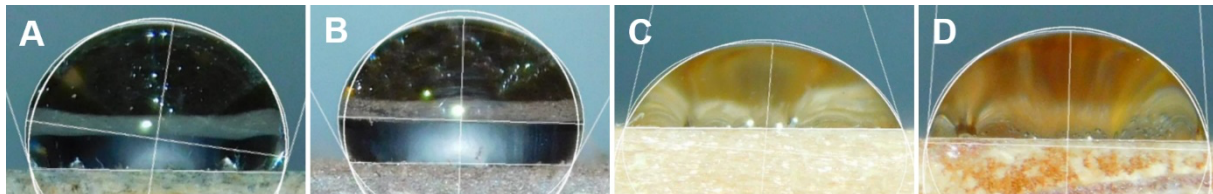
Figure 1. Determination of wettability of the adherend without surface treatment: a) Cetris Basic; b) WPC; c) three-layer glued board; d) solid wood.

Subsequently, all observed surfaces were mechanically treated with P80 sandpaper. As can be seen from the outputs in Table 5 and from the graphical assessment in Figure 2, a 100% improvement in wettability was not observed only in case of the surface of the wood-plastic cladding. Moreover, in combination with MS polymers, the surface of the cladding was first only cleaned with the recommended cleaning agent, as the manufacturer of the surface roughening system does not directly recommend it. This step was sufficient only in the case of wooden adherends and the Cetris Basic cladding. As in the case of the PU systems, the wood-plastic surface had to be mechanically treated to improve the wettability of the surface; the mere treatment with a primer was insufficient.

Table 5. Assessment of wettability of the surface of façade cladding with surface treatment.

Adherend	Contact angle α [°]	Wettability assessment [-]
CB	33.53 ± 5.76	ideally wettable surface
SW	0	ideally wettable surface
TLB	0	ideally wettable surface
WPC	98.89 ± 1.28	insufficiently wettable surface
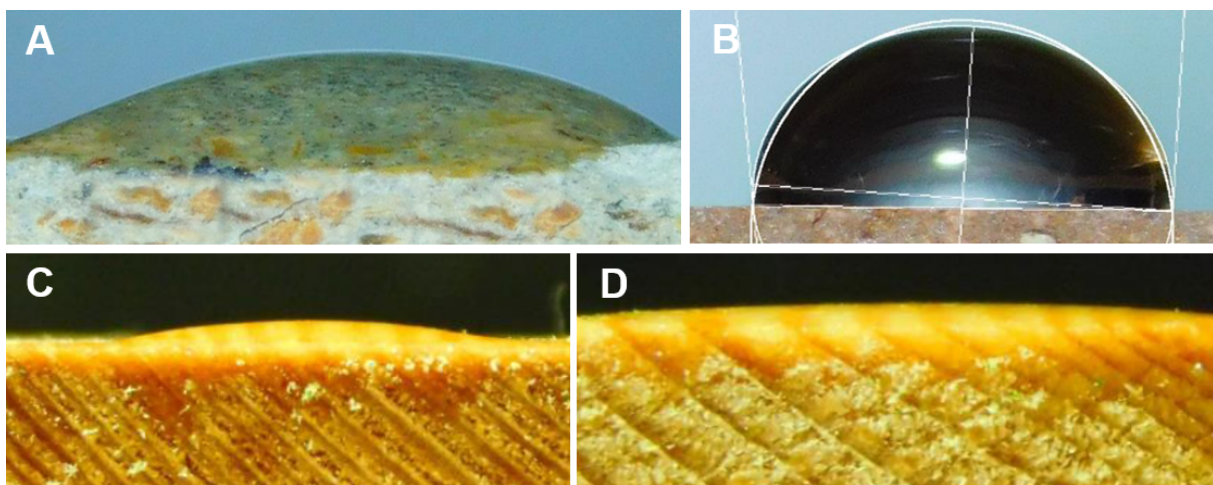
Figure 2. Determination of wettability of the adherend after surface treatment: a) Cetris Basic; b) WPC; c) three-layer glued board; d) solid wood.

3.2. Analysis of the effect of wettability on surface adhesive properties

As can be seen from the outputs of the previous subchapter. Three surfaces with good wettability and one surface with insufficient wettability were tested. Although the surface of the wood-plastic adherend was additionally roughened and creased, the hypothesis of mechanical theory was not fully confirmed. On the other hand, it is a non-porous material, therefore there was insufficient penetration of the selected liquid into all pores of the adherend material.

In order to assess the adhesive properties of the selected adherends, the adhesion was measured by means of a tensile test. As can be seen from the graphical comparison in Figure 3, the adhesive properties of the selected substrates were influenced not only by the morphology of the surface, i.e. the degree of its creasing, but also by the compatibility with the selected adhesive system. The measured results thus significantly support the hypothesis of the electrostatic theory.
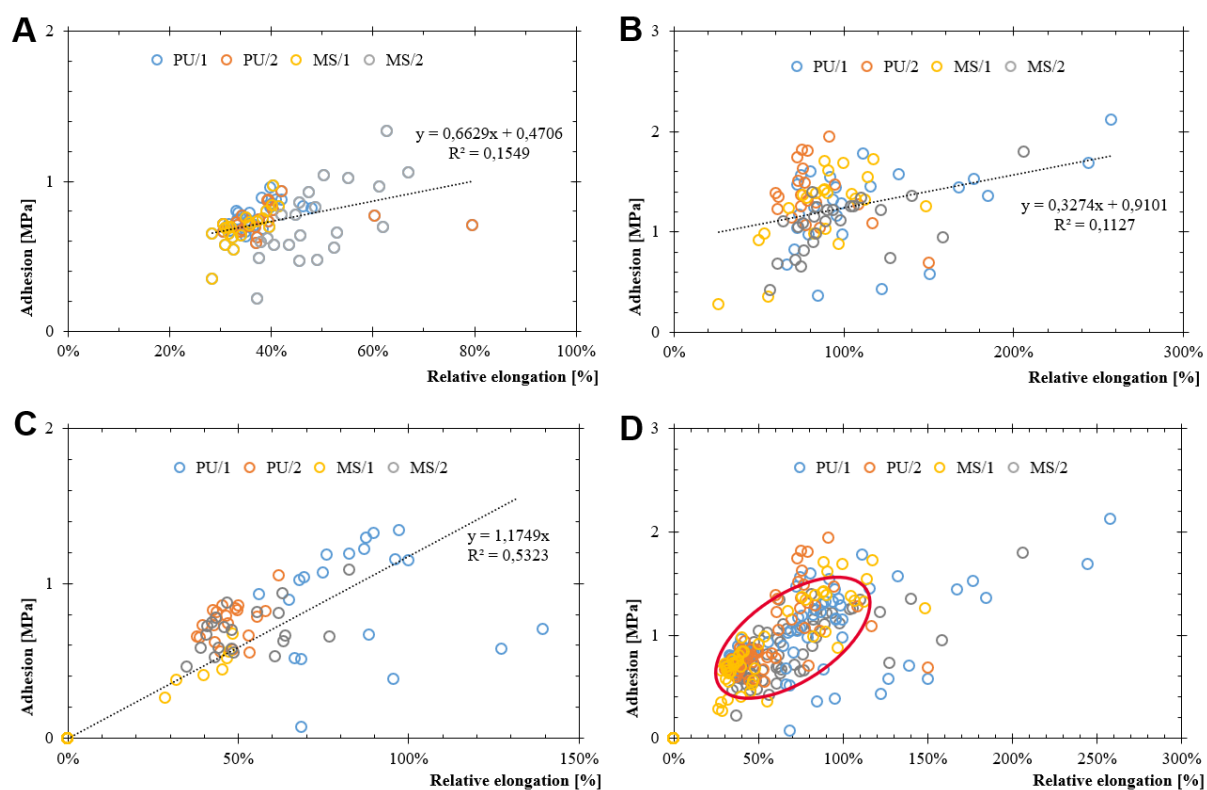

Figure 3. Comparison of adhesion to selected adherends and their compatibility with the selected mounting systems: a) Cetris Basic; b) solid wood; c) WPC; d) comparison of the deformation of the bonded joint with respect to the mounting system.

Although the wettability of the WPC surface was insufficient even after roughening, the adhesion measured in combination with this adherend is according to the trend line the most consistent. The adhesive properties of the adherends affect not only the amount of stress that the bonded joint can withstand, but also the relative elongation of the joint, as well as the type of failure. The data shown in Figure 3 a) demonstrate that the failure of the bonded joint most often occurred when a stress of about 0.8 MPa and a relative elongation of 30–40 % was reached. Relatively identical results were obtained in combination with the PU/1, PU/2 and MS/2 systems. These systems are directly designed for bonding of façade cladding and are more flexible when compared to the MS/1 system. This is a feature that makes it easier for the bonded joint to withstand changes in the joint stress caused, for example, by volume changes in the façade cladding and the supporting substructure. The compatibility of the selected adherend was also confirmed by the predominant type of joint failure method; the substrate

failure in 88 % of the measurements was so-called fiber-tear failure. The tensile strength of the Cetris board perpendicular to the board's plain is c. 0.63 MPa, therefore, the value stated by manufacturer has been exceeded by more than 20 %.

Furthermore, two types of façade cladding made of Siberian larch, i.e. solid wood planks and a massive large-format three-layer glued board, were tested. It was considered that the adhesive properties of both surfaces are very similar, and this assumption was confirmed by the results of the wettability test. Therefore, only one variant of the wood cladding was tested in the adhesion test – solid wood. The adhesive properties of this substrate are very good, the most frequent was the cohesive failure (observed in 52 % of cases), or less frequent was the so-called thin-layer cohesive failure. As can be seen from the comparison in Figure 3 b), an average stress of 1.0–1.5 MPa was achieved at an elongation of 75–110%. Although the maximum stress or elongation declared by the manufacturer of the tested adhesive systems was not reached, it is a very satisfactory assessment of the adhesive properties of the selected adherend. The last of the tested adherends was the wood-plastic. Unfortunately, the bondability of WPC is highly affected by its thermoplastic matrix [19]. The thermoplastic component of the matrix has usually a very low surface energy and bad wettability properties [19-20]. As mentioned above, it was necessary to modify the adherend in combination with MS systems, although their manufacturer does not directly recommend it. The mechanical surface treatment led to an 100% improvement of adhesion and the values of average stress at the board's plain of MS/2 were comparable to those observed in combination with PU/2. On the contrary, no improvement was observed in combination with MS/1. Some samples broke even before the tensile test was started. Moreover, the adhesive failure was observed almost in 95 % of cases.
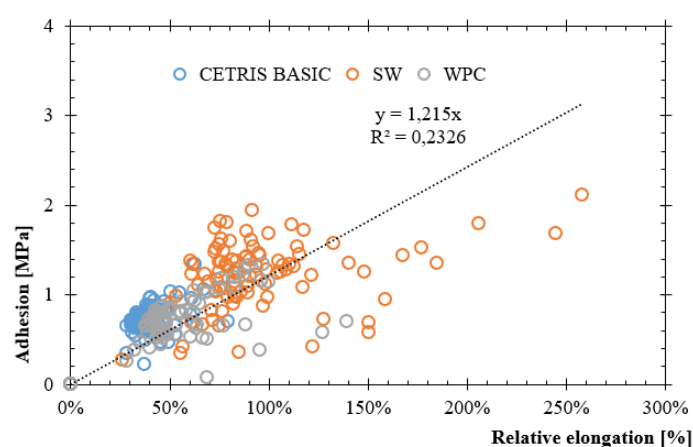
Figure 4. Comparison of the deformation properties of the bonded joint in relation to the used substrate.

The comparison presented in Figure 3 d) and Figure 4, demonstrates the effect of substrate selection on adhesive strength of the joint and the importance of material compatibility. The better the adhesive properties of the adherend, the larger elongation, and joint strength can be observed. The data recorded in combination with Cetris Basic proved that the material has good adhesive properties since it was compatible with all selected adhesive systems, moreover, the limit tensile strength of the material was reached. The data observed with the WPC substrate were consistent, see Figure 4, however, the results were highly affected by poor adhesive properties of the surface. This was confirmed by the combination with wooden substrate. As can be seen in Figure 3 d), the systems PU/2 and MS/1 reached high strength and smaller elongation before the joint failure, on the contrary, the systems PU/1 and MS/2 were extended to more than 100 %. Finally, the one-way analysis of variance (ANOVA) was performed to determine the significance of adhesion theories in area of flexible bonded structural joints. The results of the analysis only confirmed the results presented in Figure 3 and Figure

4. The adhesion to the cement-bonded particleboard was not significantly affected by poor adhesive properties which was also confirmed by minor differences in the recorded tensile strength. On the contrary, the variance of values measured with the wooden substrate was larger and less accurate. In some combinations more than 10 samples were tested since the data were more inconsistent. The ANOVA did not verify significant difference only in combination with PU/2. Similar results were obtained also in combination with WPC substrate.

4. Conclusions

The presented paper deals with the study of the surface of selected substrates in order to predict the adhesive properties. The influence of surface adhesive properties on the adhesion of a bonded joint was examined in four types of adherends, where two representatives have almost identical surface properties. Representatives with high surface polarity (SW), medium-high polarity (Cetris Basic) and low polarity (WPC) were deliberately selected. The polarity of the surface of substances is the cause of the formation of surface energy which is expressed by surface tension. The higher the surface tension value, the more polar the solid surface or the liquid is. The polarity of the surface was verified by determining the so-called wetting contact angle. The wetting contact angle is one of the few directly measurable properties of the solid/liquid/gas phase interface. This is the angle of the tangent to the surface of the drop at the point of contact of the drop with the interface. The disadvantage of the measurement is the fact that it can be easily affected by the inhomogeneity of the surface, therefore homogeneous and cohesive materials were chosen. At the same time, 4 assembly systems were tested, where the selected representatives of polyurethanes have a higher polarity than the representatives of MS polymers.

Based on the measurement of the contact angle and the determination of the wettability of the surface of selected adherends, their mechanical treatment was performed, which led to the improvement of adhesive properties of all substrates. While all assembly systems in combination with Cetris Basic and wood cladding had a very small incidence of adhesive failure, which is a sign of insufficient adhesive properties, adhesive failure occurred in 100 % of the performed tests in combination with WPC. It is therefore clear that the hypotheses of the mechanical theory are relevant, but do not lead to a significant improvement in the adhesion of the surface of plastics. On the contrary, in the case of the diffusion theory, it was proven that the use of a primer creates a layer improving the adhesive properties of the surface and, at the same time, modifying the surface tension, which facilitates the formation of a firm joint. This theory was tested mainly in combination with the Cetris Basic material. Its adhesive properties were greatly improved by mechanical surface treatment, but a very strong joint was formed using a primer. The problem was again noted in combination with WPC, where the adhesive failure of the joint occurred precisely in the microlayer between the primer and the surface of the substrate. This means that in the case of this material, the diffusion theory hypothesis was completely disproved. The hypotheses of the adsorption theory, which, to some extent, also includes the assumptions of the theories mentioned above, seem to be the most fundamental for the researched area.

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