Evaluation of factors influencing adhesive bond strength

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Abstract: In the last ten years periods the bonding technology noted a great boom not only in manufacturing industry but in repairing industry, too. The expansion of chemical industry is the cause of this boom. In this way the use of bonding technology in industrial applications brings considerable cost savings. For the successful use of adhesives the knowledge of used adhesives and of further affecting factors is important. Respecting of this know-how is the presumption of the bonded joint successful design. The breaking of the technological procedure and the incorrect design are very often reasons of wrong joints. The paper contains theoretical in formation about the bonded joints creation and some results of laboratory tests inquiring into the reasons which affect the bonded joint strength. For tests the two-component epoxy adhesives were used.

Keywords: bonding; bonding technology; surface preparation; testing of adhesive bonds

Bonding technology – theory of the bonded joint creation

The bonded joint strength depends above all on the adhesion and cohesion. The resultant bonded joint strength is very important. The notion "bonded joint strength" means the complex property which depends not only on the basic forces of adhesion and cohesion, but on a row of further factors, too, which affect the resultant bonded joint strength. Therefore it is necessary to find out these factors and to respect them at the bonding technology application. The substantial factor which affects the resultant bonded joint strength is above all the finish of the bonded surface (LOCTITE 1998).

The adhesion is possible to define as the adherence force (the intermolecular attractive chemical and physical forces) acting on the contact surfaces. The major forces which affect the adhesion, but the cohesion, too, are the forces caused by the energy of the molecular system. The relation between energy U, force F and intermolecular distance r is possible to figure by the Lennardo-Jones potential (Figure 1) (Kovačič 1984; Pizzi & Mittal 2003).

The Lennardo-Jones curve is expressed by the equation (1).

$$U = -\frac{A}{2}\left(\frac{2}{r^6} - \frac{r_0^6}{r^{12}}\right) \tag{1}$$

where: U – energy (J),
A – constant,
r – real intermolecular distance (m),
r_0 – equilibrious intermolecular distance (m).

The quotient $2/r^6$ expresses the attraction of two molecules, the quotient r_0^6 expresses their repulsion. Relation for the potential intermolecular bonding energy of two particles of their intermolecular distance $r = r_0$ is:

$$U_0 = \frac{A}{2r_0^6} \tag{2}$$

where: U_0 – intermolecular potential energy (J),
A – constant,
r_0 – equilibrious intermolecular distance (m).

From this relation it is possible to express the value of the constant A. The acting force gives the equation:

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$$F = -\frac{dU}{dr} = 6A\left(\frac{1}{r^7} - \frac{r_0^6}{r^{13}}\right) \quad (3)$$

where: F – acting force (N),
U – energy (J),
A – constant,
r – real intermolecular distance (m),
r_0 – equilibrious intermolecular distance (m).

The maximum force F_{max} of the force F is in the inflexion point of the curve $U = f(r)$, then in the distance $r_{F\max}$ the equation (4) is valid

$$\frac{d^2U}{dr^2} = 0 \quad (4)$$

where: U – energy (J),
r – real intermolecular distance (m).

According to the equation (3) the equality exists (5)

$$6A\left[-\frac{7}{r_{F\max}^8} + \frac{13r_0^6}{r_{F\max}^{14}}\right] = 0 \quad (5)$$

where: A – constant,
$r_{F\max}$ – intermolecular distance of the maximum force (m),
r_0 – equilibrious intermolecular distance (m).

The maximum force F_{max} is given by the second-order derivative of the relation (3) for the conditions of the inflexion point of the curve. Then we calculate the equation:

$$r_{F\max} = r_0\left(\frac{13}{7}\right)^{1/6} = 1.1087r_0 \quad (6)$$

where: $r_{F\max}$ – intermolecular distance at the maximum force (m),
r_0 – equilibrious intermolecular distance (m).

By the substitution in the equation (3) we get:

$$F_{\max} = 6A\left[\frac{1}{(1.1087r_0)^7} - \frac{r_0^6}{(1.1087r_0)^{13}}\right] = 1.345$$

$$\left(\frac{A}{r_0^7}\right) = 2.690\left(\frac{U_0}{r_0}\right) \quad (7)$$

where: F_{max} – maximum force (N),
A – constant,
r_0 – equilibrious intermolecular distance (m),
U_0 – intermolecular potential energy (J).

From the equation (7) it follows that the maximum attractive force F_{max} between two molecules of the same molecular diameter depends on the intermolecular potential energy U_0, which is also called the intermolecular bond energy. From these relations it is possible to determine the intensity of the theoretical value of materials strength caused by the molecular energy system (Peterka 1980; Ptáček 2001).

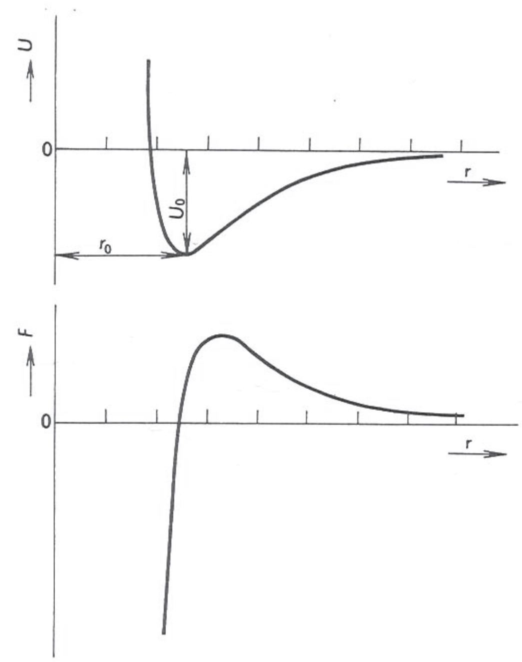

Figure 1. Lennardo-Jones curve

Except these forces named primary forces the next secondary adsorbing forces occur on the contact surfaces, which are called Van der Waals forces. These forces acting between the molecules are very low and weaken with their distance. The reach of these molecular forces is substantially lower than the surface roughness depth of machined surfaces. Therefore it is necessary that the adhesive penetrates into these inequalities and wets perfectly both surfaces. Good adhesion depends on the good wet ability of the bonded surfaces by the liquid adhesive. Therefore the surface energy of the adhesive must be lower (or maximum the same) than the critical surface energy of the bonded material. This stress is the effect of attractive forces which act between the molecules of the material surface (LOCTITE 1998; Peterka 1980). Metals are of relatively high surface energy. Therefore adhesives of relatively low surface energy ensure the basic condition for the strong bonded joint creation. Bonding plastics the relations of surface energies are critical. Using the suitable treatment of plastics surfaces e.g. by the use of primers and activators the surface energy can be positively influenced so that the bonding is possible.

Then the measure of the bonded material surface coating by the adhesive depends on the adhesive consistency, surface cleanness and roughness and on the surface inequalities form. The form and size of surface inequalities depend on the material structure, on the manufacturing process and on the surface finish, too (Peterka 1980).

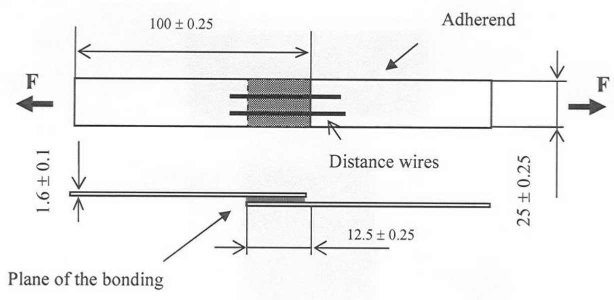

Figure 2. Form and sizes of the tested body according to ČSN EN 1465 (1997)

Cohesive forces are in the adhesive film. Cohesion is the summary of forces which bind the material particles together by interaction of valence and molecular attractive forces. It is also called internal adhesion. The intensity of cohesion is given by the so called cohesion energy which can be expressed as the energy needed for the separation of one adhesive particle from the others (the strength of the adhesive alone) (Cagle 1973; Habenicht 2002).

Valence and intermolecular forces result from the intermolecular attractive forces named Van der Waals forces and from the interconnection of polymer molecules.

Again the Lennardo-Jones potential is here of use. By means of it the relation between the cohesion strength σ_c and the elasticity of elongation E can be derived (8) (Kovačič 1984).

$$\sigma_c = 0.064 \times E \quad (8)$$

where: σ_c – ideal cohesion strength (MPa),
E – coefficient of elasticity (MPa).

The cohesion strength depends on the material and temperature parameters of a given material. Metals are of higher cohesion strength than plastics. The temperature increase causes the cohesion strength decrease. It is caused above all by the increasing mobility of the material molecules. The increase of the adhesive cohesion strength is caused by the transition from the liquid phase in the cured state.

MATERIAL AND METHODS

Labor tests with a view to the bonded surface finish determination were carried out according to the standard ČSN EN 1465 (1997) using the standardized specimens made from duralumin and steel bonded by means of two-component epoxy adhesives. This test determines the tensile lap-shear strength of rigid-to-rigid bonded assemblies. The lap joint is strained by the tensile force acting parallel with the bonded surface and with the principal axis of the specimen till to the failure. The tested body (Figure 2) is made by bonding of two tested specimens of size 100 × 25 × 1.5 mm. The specimens were bonded so that the lap length was 12.5 mm (ČSN EN 1465 1997).

Table 1 presents the chemical composition of bonded specimens. First the optimal adhesive layer thickness was determined, which was used for further tests. Following adhesive layer thicknesses were tested: 0.06 mm, 0.11 mm, 0.16 mm, 0.22 mm, 0.29 mm and 0.39 mm. Each adhesive layer thickness was se-

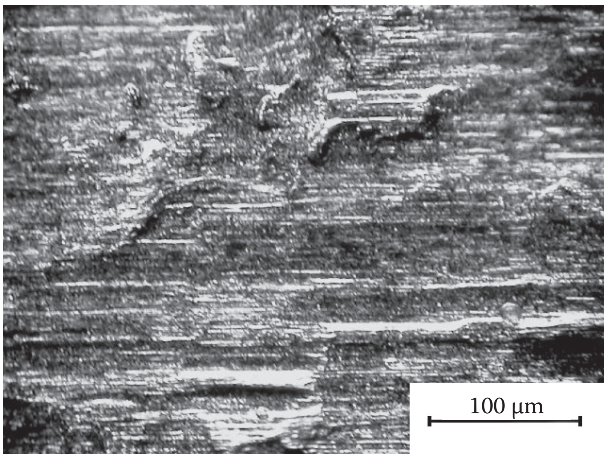

Figure 3. Local view of the steel semi product surface

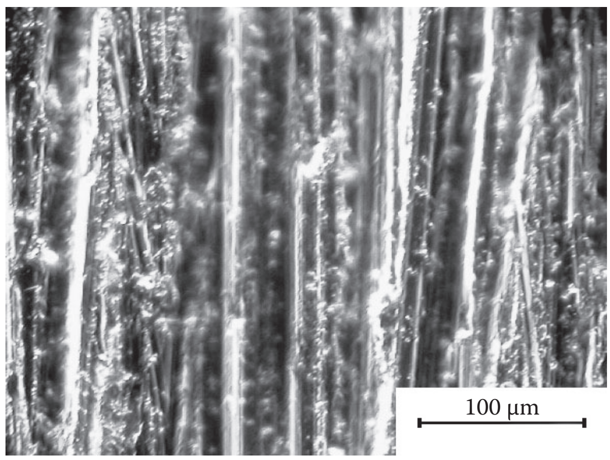

Figure 4. Local view of the surface after grinding using the abrasive cloth

Table 1. Chemical composition of bonded materials (%)

Element	C	Mn	Cr	Ni	Al	Cu	Nb	Ti	Fe	Si	Mg	Zn
Steel	0.047	0.24	0.076	0.017	0.065	0.039	0.007	0.016	99.5	–	–	–
Duralumin	–	0.51	0.003	0.003	93.197	5.012	–	0.013	0.304	0.35	0.571	0.014

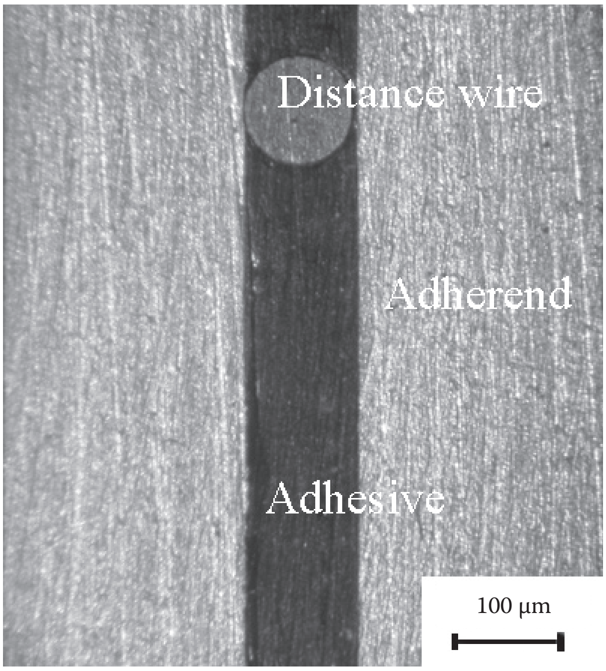

Figure 5. Bonded joint section

cured using two distance wires of requisite diameter, which were put in the bonded joint.

The determined optimal adhesive layer thickness was used at tests for the suitable surface mechanical finish determination. The mechanical finish was carried out using the abrasive cloth of different grain size 40, 100, 150, 240, 320, 400 and 500 (Figures 3 and 4). Only the surface finish using the abrasive cloth was chosen because it is easy accessible above all at repairs in not so good implemented workshops, which have not available e.g. a sand blaster.

On likewise finished specimens the surface roughness was measured using the Mitutoyo profilograph SURFTEST – 301 according to the Standard ISO 4287 (1999). The specimens were ground normal to the applied force in the test of the tensile lap-shear strength (ČSN EN 1465 1997). Above all the R_a value (the arithmetic mean of the departures of the profile from the mean line) (ISO 4287 1999).

After grinding the specimens were rinsed in perchlorethylene.

After the roughness evaluation the specimens were rinsed once more, the pertinent adhesive layer was put on, which was secured by means of two distance wires (Figure 5). The bonded parts were locked. The bonded joint was left in the laboratory for the time presented in the instructions user to reach the perfect curing at the laboratory temperature.

The shear load test was carried out using the universal tensile-strength testing machine ZDM 5. After the failure of the specimen the highest force was read on the scale, the bonded surface was measured, the failure type was determined according to ISO 10365 and the strength of the bonded joint was calculated (9).

$$\tau = \frac{F}{S} \qquad (9)$$

where: τ – shear strength (MPa),
F – highest force (N),
S – bonded surface (mm^2).

Characteristic of used two-component epoxy adhesives

Following two-component epoxy adhesives were evaluated, which components ratio was 1:1 and which cure at laboratory temperature (FIRM LITERATURE). Table 2 contains commercial names of evaluated adhesives and their specifications. In following graphs the adhesives are mentioned according to Table 2 (adhesives 1 to 5).

RESULTS

Carried out laboratory tests intended to the suitable bonded surface finish determination are elaborated in following graphs, tables and commentaries. The graphically presented relation course is expressed by the nonlinear regressional quadratic function. The function type is derived from the correlation field form, which is created by the cross points of the dependent and independent variables.

On the basis of test results the optimal thickness of the adhesive layer was determined. The determined

Table 2. Characteristics of used adhesives (FIRM LITERATURE)

Commercial name	Name used in test	Application possibility	Usable life (min)	Curing time	Thermal fastness
Bison metal	adhesive 1	metals, aluminium alloys, ceramics, wood and plastics	60	12 h	– 60 till + 100°C
Bison universal	adhesive 2		120	24 h	– 50 till + 80°C
Lepox	adhesive 3		30	48 h	100°C
Uhu 2 min	adhesive 4		2	5 min	– 60 till + 80°C
Uhu 5 min	adhesive 5		3–5	30 min	– 60 till + 80°C

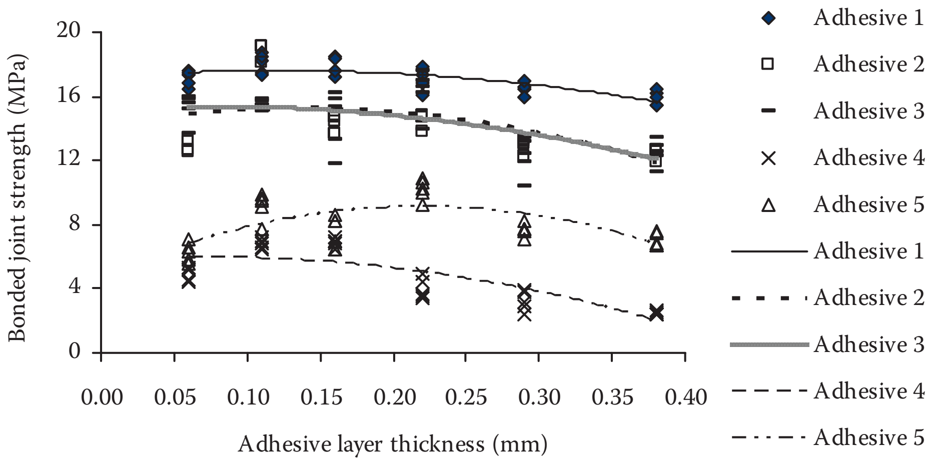

Figure 6. Bonded joint strength related to the adhesive layer thickness

optimal adhesive layer thickness was used at tests for the suitable surface finish determination. The results of the relation between adhesive layer thickness and strength are shown in Figure 6.

The coefficient of variation (10) was in this case from 1.58 to 14.1%. Coefficient of variation is calculated as the deviation between the bonded joint strength and the mean value.

$$\nu = \frac{S_0}{\bar{x}} \times 100 \qquad (10)$$

where: ν – coefficient of variation (%),
S_0 – standard deviation (MPa),
$\bar{x}$ – arithmetic mean (MPa). 2

For the correct evaluation the determination of the given relation is important, too. It is the task of the correlation analysis. The closeness of this relation is judged by means of the determination index. It ranges from 0 to 1. When the values approach to 1, the relation is more intense. The determination index $I_{\tau x}$ indicates how the dependent variable is influenced by the independent variable. The calculated values are presented in Table 3. From the determination indexes the significant till high closeness is evident.

After the optimal adhesive layer thickness determination the surface of next test specimens was finished using the abrasive cloth of various grain size. The surface roughness R_a was measured, which was different according to expectation not only using different grain size but using different specimen material, too. The mean values are presented in Table 4.

Table 3. Equations of regressional functions and their determination index

Name used in test	Functional equation	Determination index $I^2_{\tau x}$
Adhesive 1	$\tau = -28.701x^2 + 6.9249x + 17.195$	0.57
Adhesive 2	$\tau = -58.243x^2 + 16.28x + 14.149$	0.29
Adhesive 3	$\tau = -35.152x^2 + 5.216x + 15.183$	0.40
Adhesive 4	$\tau = -42.98x^2 + 6.5967x + 5.6713$	0.65
Adhesive 5	$\tau = -90.704x^2 + 39.841x + 4.7283$	0.42

Table 4. Mean values of R_a surface roughness

Abrasive cloth of grain size	40	100	150	240	320	400	500
Mean surface roughness R_a – duralumin (μm)	5.10	2.10	1.50	1.80	1.70	0.80	0.50
Mean surface roughness R_a – steel (μm)	2.40	1.27	0.76	0.58	0.53	0.51	0.42

Table 5. Equations of regressional functions and their determination index

Name used in test	Functional equation	Determination index $I^2_{\tau x}$
Adhesive 1	$\tau = -5E\text{-}05x^2 + 0.0352x + 12.647$	0.52
Adhesive 2	$\tau = -5E\text{-}05x^2 + 0.0352x + 12.647$	0.60
Adhesive 3	$\tau = -4E\text{-}05x^2 + 0.0249x + 7.3992$	0.43
Adhesive 4	$\tau = -1E\text{-}05x^2 + 0.0062x + 2.5912$	0.52
Adhesive 5	$\tau = -8E\text{-}06x^2 - 0.0015x + 4.5333$	0.71

Table 6. Equations of regressional functions and their determination index

Name used in test	Functional equation	Determination index $I^2_{\tau x}$
Adhesive 1	τ = 3E-05x^2– 0.0188x + 20.132	0.57
Adhesive 2	τ = 3E-05x^2– 0.0205x + 20.982	0.53
Adhesive 3	τ = 4E-06x^2– 0.0003x + 8.5832	0.10
Adhesive 4	τ = 8E-06x^2– 0.0117x + 6.7036	0.78
Adhesive 5	τ = 2E-05x^2– 0.0176x + 8.5319	0.71

Table 7. Test results for duralumin

Name used in test	Optimal adhesive layer thickness (mm)	Duralumin			
		Surface preparation	Grain size (μm)	Mean surface roughness R_a (μm)	Adhesive bond strength (MPa)
Adhesive 1	0.11	abrasive cloth 240	44.5	1.31	18.70
Adhesive 2	0.11	abrasive cloth 240	44.5	1.18	19.64
Adhesive 3	0.22	abrasive cloth 240	44.5	1.26	12.36
Adhesive 4	0.16	abrasive cloth 150	98.0	1.54	3.46
Adhesive 5	0.22	abrasive cloth 100	137.5	2.09	4.57

Table 8. Test results for steel

Name used in test	Optimal adhesive layer thickness (mm)	Steel			
		Surface preparation	Grain size (μm)	Mean surface roughness R_a (μm)	Adhesive bond strength (MPa)
Adhesive 1	0.11	abrasive cloth 100	137.5	1.29	19.33
Adhesive 2	0.11	abrasive cloth 100	137.5	1.27	20.05
Adhesive 3	0.22	abrasive cloth 100	137.5	1.27	9.68
Adhesive 4	0.16	abrasive cloth 100	137.5	1.26	6.16
Adhesive 5	0.22	abrasive cloth 40	462.5	2.40	7.62

The relation between bonded joint strength and grain size is shown in Figures 7 and 8, using duralumin specimens. The calculated determination index is presented in Table 5. The closeness was high. The coefficient of variation was from 2.61 to 10.56%. Only at the use of the adhesive 4 the coefficient of variation reached higher values from 2.39 to 16.84%.

The determination index of steel bonded joints is presented in Table 6. The closeness was high, too. The coefficient of variation was from 1.02 to 11.64.

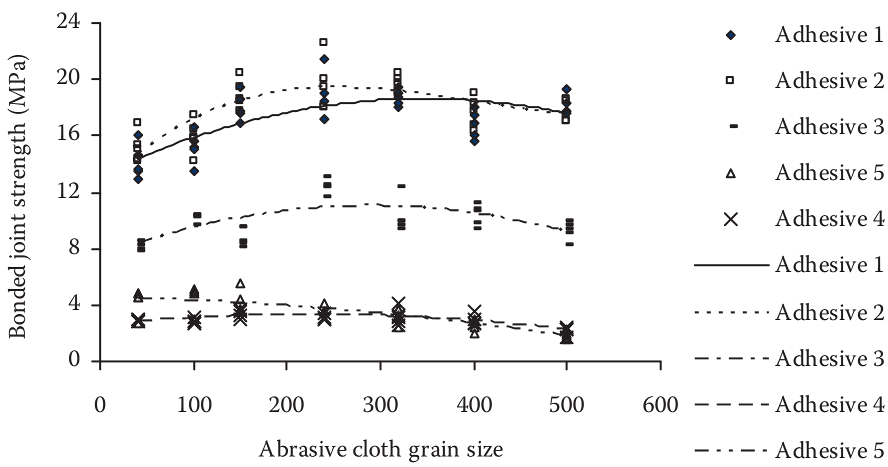

Figure 7. Relation between joint strength and abrasive cloth grain size using the duralumin specimens

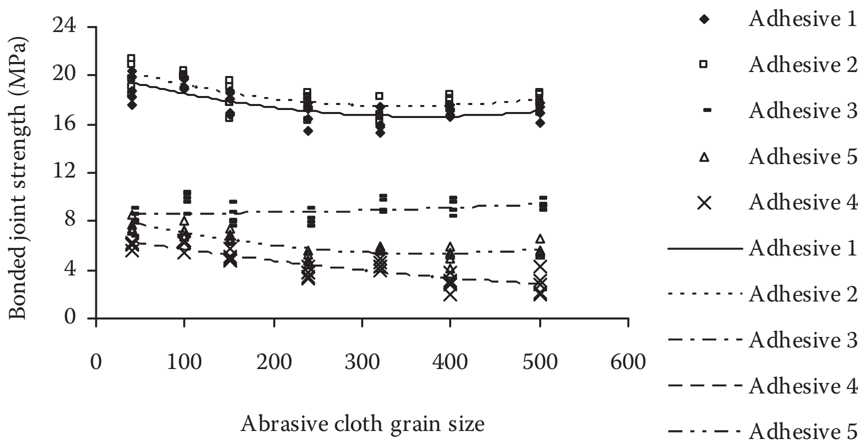

Figure 8. Relation between joint strength and abrasive cloth grain size using the steel specimens

The coefficient of variation reached higher values from 3.18 to 20% at the use of the adhesive 4 again.

The test results are arranged hand summarized in Tables 7 and 8, where the optimal adhesive layer thickness, suitable abrasive cloth, reached surface roughness R_a and bonded joint strength are presented for each of the adhesives.

CONCLUSIONS

In the paper the laboratory test results of one of basic technological properties of epoxide adhesives are presented. According to the results the adhesive layer thickness from 0.1 to 0.25 mm can be commonly determined for all tested adhesives. Concretely they were: Adhesive 1–0.11 mm, adhesive 2–0.11 mm, too, adhesive 4–0.16 mm, adhesives 3 and 5–0.22 mm. The optimal layer thickness determination is important for several reasons, which are above all higher strength of the bonded joint and low costs owing to the lower volume of consumed adhesive. At adhesive layer thickness increase over the optimal value the costs increase owing to the above mentioned reasons. Further using the equations from Table 3 the bonded joint orientation strength can be calculated by substituting the adhesive layer thickness for the variable x. E.g. using the adhesive 1 and the adhesive layer thickness 0.9 mm the bonded joint strength would be approximately to 0.1 MPa, using the adhesive 2 already at the thickness 0.65 mm. The carried out laboratory tests intended on the suitable surface finish show the importance of this factor on the resultant bonded joint strength. The adhesive 1 shows the strength difference of 15.4% at steel, of 27.7% at duralumin in dependence on the abrasive cloth choice. The adhesive 2 shows the difference of 14.3% at steel, of 22.9% at duralumin, the adhesive 3 of 17.1% at steel, of 33.4% at duralumin, the adhesive 4 of 53.89% at steel, of 37% at duralumin. The last adhesive 5 shows the strength decrease of 34.9% at steel, almost 60% at duralumin. Therefore the necessity of the correct choice of the surface mechanical preparation is evident.

For the bonded joint reliable operation not only high strength but the low strength deviation from mean values (coefficient of variation) is important, too. At the optimal surface finish the following coefficients of variation were calculated: Adhesive 1 – steel 2.8%, duralumin 8.4%, adhesive 2 – steel 1.02%, duralumin 8.3%, adhesive 3 – steel 6.37%, duralumin 3.6%, adhesive 4 – steel 6.53%, duralumin 7.66%, adhesive 5 – steel 6.73%, duralumin 16.8%. Only using the adhesive 5 relative high strength deviations from the mean values were determined (duralumin 16.8%).

At bonded joints using the adhesives named as adhesives 3, 4 and 5 the adhesive cohesive failure was determined. Using adhesives 1 and 2 the cohesive failure at steel surface and adhesive cohesive failure at duralumin surface always occurs. Using the duralumin specimens the adhesive cohesive failure is always effected by the deformation of the bonded material. The deformation effects the spalling and in this way the adhesive cohesive failure of the bonded joint occurs.

On the basis of carried out laboratory measurements it is possible to say that the same adhesive behaves different when bonding different materials. Therefore it demands different technological processes at the bonded surfaces finish. Ahead of the bonding technology application it is necessary to carry out a preliminary test verification of single adhesives or to acquire these information from the manufacturer. Bonded joint is a very exacting system and therefore it is necessary to pay attention to single factors which influence the bonded joint strength.

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Abstrakt

Müller M., Hrabě P., Chotěborský R., Herák D. (2006): **Hodnocení faktorů ovlivňujících pevnost lepeného spoje.** Res. Agr. Eng., **52**: 30–37.

V posledních desetiletích s rozmachem chemického průmyslu zaznamenala i technologie lepení prudký vývoj nejen ve výrobním, ale i v opravárenském průmyslu. Využívání technologie lepení v průmyslových aplikacích přináší značné úspory. Pro úspěšné používání lepidel v praxi je důležitá znalost technologických vlastností používaných lepidel a dalších faktorů, které lepený spoj ovlivňují. Respektování těchto znalostí je předpokladem úspěšného návrhu lepeného spoje. Velmi častým důvodem špatného lepeného spoje je nedodržování technologických postupů při návrhu a tvorbě vznikajícího spoje. Příspěvek přináší teoretické poznatky o vzniku lepeného spoje a je zaměřen na některé výsledky laboratorních experimentů, ovlivňující pevnost lepeného spoje. K experimentům byla použita konstrukční dvousložková epoxidová lepidla.

Klíčová slova: lepení; technika lepení; úprava povrchu; testování lepených spojů

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