

## ADHESION OF EPOXY COATINGS WITH MICROMETER SIZE FILLERS TO STEEL SUBSTRATE<sup>1</sup>

PAULINA MAYER, ANNA DMITRUK

*Wroclaw University of Science and Technology, Faculty of Mechanical Engineering, Wroclaw, Poland*  
*e-mail: paulina.mayer@pwr.edu.pl; anna.dmitruk@pwr.edu.pl*

MAREK LUBECKI, MICHAŁ STOSIAK

*Wroclaw University of Science and Technology, Faculty of Mechanical Engineering, Department of Maintenance and Operation of Logistics, Transportation and Hydraulic Systems, Wroclaw, Poland*  
*e-mail: marek.lubecki@pwr.edu.pl; michal.stosiak@pwr.edu.pl*

The paper concerns pull-off strength results of epoxy coatings without and with five different fillers. Polymer coatings were applied to a steel substrate that was degreased and/or pretreated by the means of abrasive blasting using electrocorundum and cast steel shot with different grain sizes. The roughness profile and the basic roughness parameters were determined. The results showed a decisive effect of substrate preparation on the coating adhesion.

*Keywords:* epoxy coatings, pull-off test, coatings with powder fillers, adhesion of coatings

### 1. Introduction

Polymeric coatings, due to their high durability, good adhesion to most substrates, ease of application and renovation as well as low manufacturing costs are commonly used to protect various surfaces against negative effects of ambient factors (Kotnarowska, 2012). They are applied to many types of substrates including metals, concrete, wood, plastics and ceramics. Technical coatings are used to ensure the material to possess the desired mechanical, electrical and thermal properties, i.e. coatings with improved hardness, abrasion resistance or resistance to high temperatures as was presented by Hallman *et al.* (2012) and Zhang *et al.* (2011). A properly prepared surface of the metal (steel, aluminum) substrate is a factor affecting the adhesion of the coating (Dmitruk *et al.*, 2018). The preparation of the steel substrate prior to the application of the coating primarily consists of removing all kinds of impurities and improving surface roughness. The most common contaminants are dust, dirt, oils, lubricants, water and moisture, and metal oxides that prevent good adhesion of the coating to the substrate. The necessity of obtaining the appropriate surface roughness of the substrate results from the very nature of the coating application process, because its adhesion is caused primarily by mechanical anchoring of the first layer of the coating in the unevenness of the surface of the substrate. An improperly prepared surface of the steel substrate may cause many negative effects, e.g. reduction of adhesion of the coating to the substrate, development of subcoating corrosion, formation of bubbles, cracks and peels on the coating, reduction of smoothness and gloss of the coating (Vaca-Cortes *et al.*, 1998).

For a coating to fulfill its function, good adhesion between it and the substrate is one of the most crucial factors. The methods of quality evaluation of such connections include: knife, peel,

---

<sup>1</sup>The work is related to a paper at PCM-CMM-2019.

hot water immersion, cathodic disbondment, salt spray, pull-off and bending tests which were distinguished in works of Vaca-Cortes *et al.* (1998) and Dmitruk *et al.* (2019).

The use of fillers strongly influences the change in the properties of thermoplastics and chemo-hardening polymer materials. It was verified experimentally by several research groups all over the world that nanoparticles of metallic or inorganic type prove the ability to reinforce effectively thermoplastic and also thermosetting polymer matrices (Singh *et al.*, 2002). This effect is at the same time accompanied by improvements in fracture toughness and impact energy which, however, strongly depends on the filler volume content (Wetzel *et al.*, 2001). The unique nanocomposite effects can only be effective if the nanoparticles are well dispersed in the surrounding polymer matrix. It has been shown that a considerable improvement of mechanical and tribological properties can already be achieved at a very low filler volume content, somewhere in the range of 1-5 vol.% (Zhai *et al.*, 2006; Shi *et al.*, 2009; Bauer *et al.*, 2006; Zhou *et al.*, 2009).

## 2. Materials

Epidian 652 with IDA hardness (Ciech-Sarzyna) was used to obtain coatings. Five different fillers in 20% wg. were added to the epoxy resin. The properties of the fillers (Hoffmann Mineral company) are shown in Table 1. Sillitin Z 89, Sillitin N 85 and Sillikolloid P 87 are a natural combination of corpuscular silica and lamellar kaolinite. These two elements together form a loose structure which offers unique advantages in terms of application possibilities when used as a functional filler. Aktisil EM and Aktisil AM are the activated varieties of Sillitin Z 86, produced by modification of the surface with epoxy or amino silane, respectively.

**Table 1**

Name	Sillitin Z 89	Sillitin N 85	Sillikolloid P87	Aktisil EM	Aktisil AM
Brightness Y (DIN 53 163:82)	86	82	82	82	82
Brightness Z (DIN 53 163:77)	86	75	76	77	77
Density [g/cm <sup>3</sup> ]	2.6	2.6	2.6	2.6	2.6
Particle size distribution					
D50 [μm]	1.9	3.0	1.5	2.2	2.2
D97 [μm]	9	16	6	10	10

The steel substrate was made of DC04 steel with thickness 2.0 mm, and dimensions of the produced sheets were 30 × 250 mm. The steel substrate was treated by abrading blasting using two different abrasives and after that it was cleaned by acetone. The coatings without and with fillers was mixed and then carefully applied by hand on the steel surface without and with mechanical treatment. The finished coatings were allowed to dry for 24 hours.

In the produced coatings, coagulation of fillers was observed as shown in Fig. 1a. High polymeric dispersing agent Tegomer® DA 626 was used to eliminate aggregated powders from agglomerates. It was added in an amount of 5% by weight in relation to the filler. No coagulation phenomenon was observed in the obtained coatings with the dispersing agent (Fig. 1b).

The roughness was measured with a profilometer (Marh Surf PS 10) by the contact method determining the arithmetic mean deviation of the roughness profile  $Ra$  and the roughness height by 10 points  $Rz$  (Fig. 2).

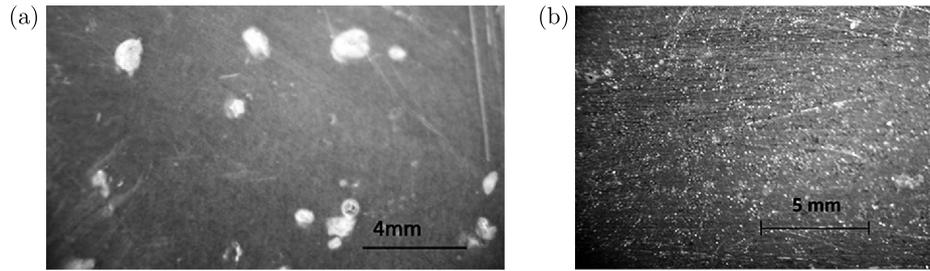


Fig. 1. Coating without (a) and with (b) dispersing agent

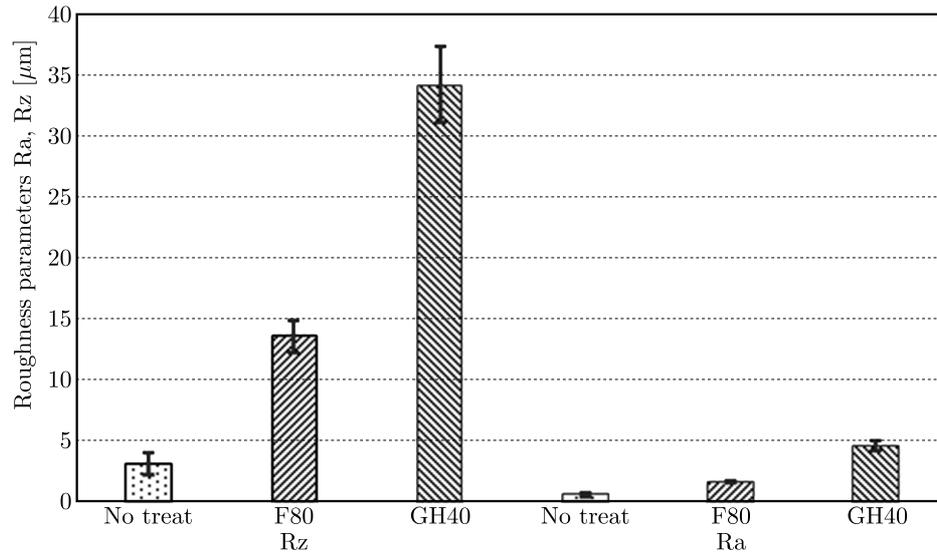


Fig. 2.  $R_a$  and  $R_z$  roughness parameters of the steel surface without and after abrasive blasting

Figures 3, 4 and 5 present graphs of the roughness profiles on steel sheets before and after abrasive blasting using F80 grain size corundum and GH40 cast steel shot.

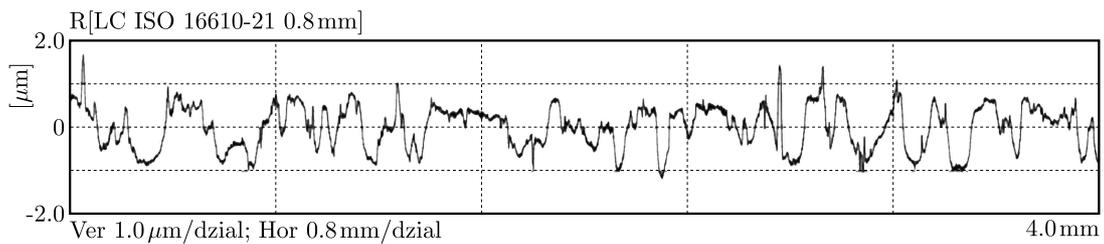


Fig. 3. Steel sheet roughness profile without abrasive blasting

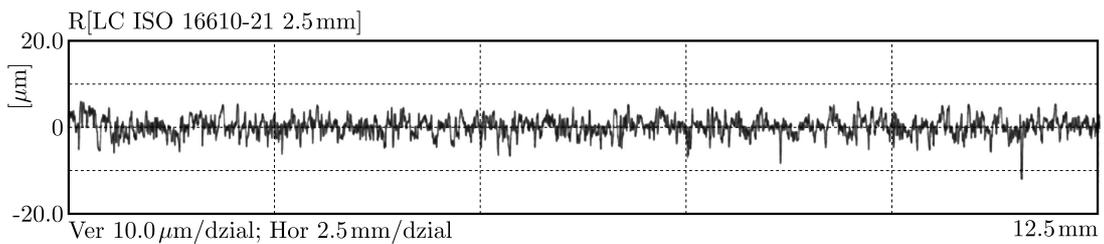


Fig. 4. Steel sheet roughness profile after abrasive blasting using F80 grain size corundum

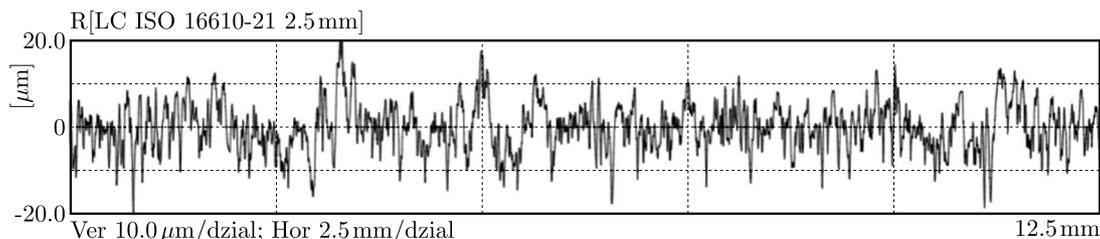


Fig. 5. Steel sheet roughness profile after abrasive blasting using GH40 cast steel shot

For a substrate without abrasive blasting  $Ra = 0.57 \pm 0.14 \mu\text{m}$ ,  $Rz = 3.12 \pm 0.86 \mu\text{m}$ , while for a substrate after pretreatment with aluminum oxide F80 and cast steel shot G40 it was respectively  $Ra = 1.65 \pm 0.04 \mu\text{m}$ ,  $Rz = 13.52 \pm 1.28 \mu\text{m}$  and  $Ra = 4.53 \pm 0.41 \mu\text{m}$ ,  $Rz = 34.15 \pm 3.0 \mu\text{m}$ . The use of abrasive blasting resulted in a significant increase in the roughness of the steel substrate.

### 3. Methods

The adhesion of the obtained coatings was determined in accordance with PN-EN ISO 4624-2004 standard (PKN, 2004). This method consists in measuring the pull-off strength of the coatings on the unit interface. The measure of adhesion of the coating is the smallest stress required to pull-off the weakest boundary layer (adhesive detachment mechanism) or the weakest point of the tested coating system (cohesive detachment mechanism). In order to investigate the adhesion of the obtained coatings, a 20 mm diameter measuring punch was glued to the surface of the coating using methyl methacrylate (MMA) adhesive and pulled off after 24 hours using DeFelsko PosiTest AT-A (Pull-off adhesion tester). The adhesion and impact tests were carried out at room temperature at 51% humidity for the epoxy coatings.

The tests were carried out on epoxy coatings applied to a steel substrate after degreasing with acetone and after abrasive blasting using F80 aluminum oxide and GH40 cast steel shot as an abrasive. Adhesion of the coatings without fillers and with five different fillers was tested. In addition, the effect of the applied dispersing agent on the adhesion between the polymer and the steel substrate was investigated.

#### 3.1. Pull-off strength of epoxy coatings

Adhesive detachment occurred in all tested samples. In Fig. 6, there is shown an example of a sample after the pull-off test, and in Fig. 7 the steel surface after detachment is presented. On the steel surface, no fragments of coatings were observed. Epoxy coatings without fillers possess the highest pull-off strength. The coatings applied to the substrate after abrasive blasting with GH40 steel shot have higher pull-off strength values in comparison to those without pretreatment.

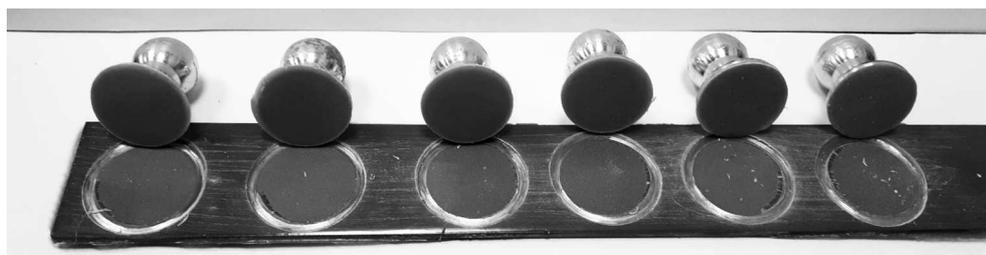


Fig. 6. An example of a sample after the pull-off test

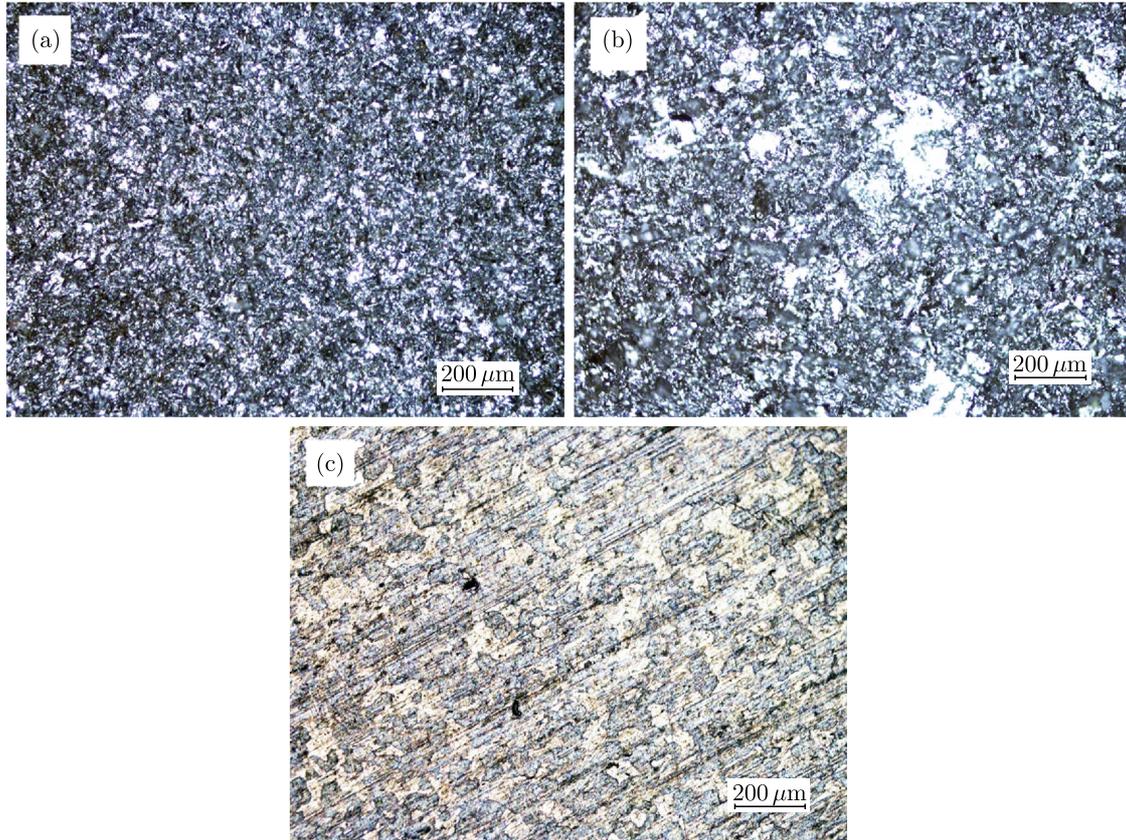


Fig. 7. Surface of the steel substrate after pull-off tests: (a) abrasive blasting F80, (b) abrasive blasting GH40, (c) degreasing

Comparing the obtained results of the pull-off strength of the coatings applied to the substrate with and without mechanical treatment, it can be clearly stated that the effect of preparation of the substrate has a huge impact. An increased roughness after machining resulted in a more than five times increase in adhesion for most of the samples. Adhesion of the coatings applied to the surface after mechanical treatment with GH40 cast steel shot is about 10% higher than after abrasive blasting using F80 grain size corundum. The increase of the pull-off strength is not proportional to the increase in the roughness parameters of the substrate. The over-expansion of the specific surface of the substrate may lead to deterioration of the coating adhesion as the formed irregularities can prevent the polymer from complete wetting of the metal sheet. Mutual interlocking in this case might be insufficient, reducing the mechanical adhesion.

Among all of the used fillers, the highest pull-off strength values were obtained for coatings with the addition of Actisil AM. In contrast, Sillitin Z 89 filler reduced the adhesion, in relation to pure epoxy coatings by about 40%, for samples after abrasive blasting.

Analyzing the obtained results of the epoxy coatings with fillers without the dispersing agent (Fig. 8) and with it (Fig. 9), significant differences in the pull-off resistance are noticed. Addition of the dispersing agent caused a decrease in the pull-off strength, but only for the coatings applied to the substrates after abrasive blasting. This phenomenon can be explained by a decrease in wetting of the unevenness (cavities) of the steel substrate by the coating. However, for the coatings with the dispersing agent on a substrate only degreased, more than a double increase in strength was observed. Addition of the dispersing agent has minimized the effect of the type of fillers on the pull-off strength.

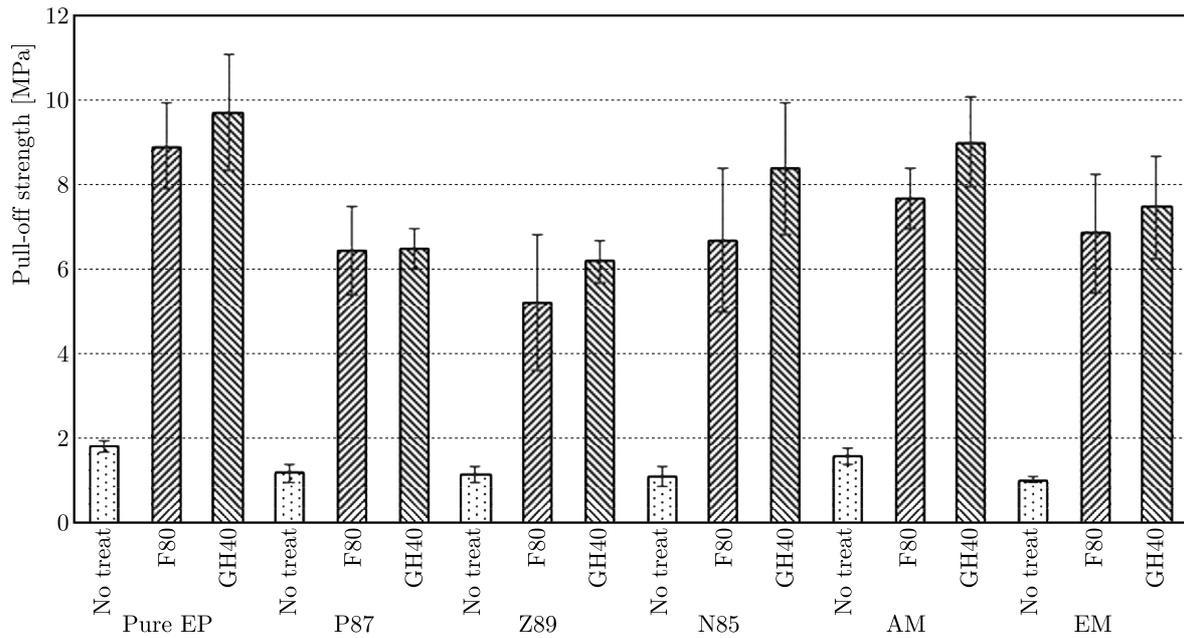


Fig. 8. Pull-off strength results for epoxy coatings without and with fillers

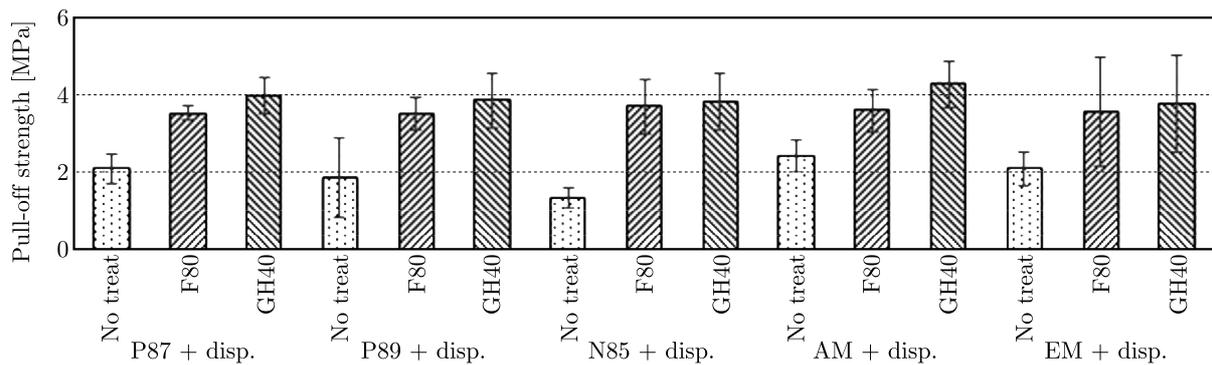


Fig. 9. Pull-off strength results for epoxy coatings without and with fillers and the dispersing agent

#### 4. Results

- The use of mechanical treatment resulted in the development of a specific surface area and change of its geometry which increased the contact of the substrate with the coating. It was confirmed by the obtained results of the pull-off strength, which were higher for the coatings on substrates with greater roughness.
- The use of mechanical treatment resulted in an almost five-fold increase in the pull-off strength for most samples.
- The addition of fillers to the epoxy coating reduced the adhesion to the steel substrate. Nevertheless, for the reinforced coatings, the best pull-off strength results were obtained for Aktisil EM filler.
- An adhesive detachment mechanism occurred in all of the tested coatings, what indicates that the manufactured coatings are uniform and homogenous as their cohesion forces are not deteriorated.
- Addition of a dispersing agent significantly affects the pull-off strength results. The adhesion of coatings that were applied to the substrate after mechanical pretreatment decreased almost twice, whereas in those only degreased, it increased.

## References

1. BAUER F., DECKER U., ERNST H., FINDEISEN M., LANGGUTH H., MEHNERT R., SAUERLAND V., HINTERWALDNER R., 2006, Functionalized inorganic/organic nanocomposites as new basic raw materials for adhesives and sealants, Part 2, *International Journal of Adhesion and Adhesives*, **26**, 567-570
2. DMITRUK A., MAYER P., PACH J., 2018, Pull-off strength of thermoplastic fiber-reinforced composite coatings, *Journal of Adhesion Science and Technology*, **32**, 9, 997-1006
3. DMITRUK A., MAYER P., PACH J., 2019, Pull-off strength and abrasion resistance of anti-corrosive polymer and composite coatings, *International Journal of Surface Science and Engineering*, **13**, 1, 50-59
4. HALLMANN L., ULMER P., REUSSER E., HÄMMERLE C., 2012, Effect of blasting pressure, abrasive particle size and grade on phase transformation and morphological change of dental zirconia surface, *Surface and Coatings Technology*, **206**, 4293-4302
5. KOTNAROWSKA D., 2012, The influence of operational factors on the surface condition of epoxy and polyurethane coatings (in Polish), *Przetwórstwo Tworzyw*, **3**, 230-234
6. PN-EN ISO 4624:2016-05. Paints and varnishes - pull-off test for adhesion
7. SHI X., NGUYEN T.A., SUO Z., LIU Y., AVCI R., 2009, Effect of nanoparticles on the anticorrosion and mechanical properties of epoxy coating, *Surface and Coatings Technology*, **204**, 237-245
8. SINGH R.P., ZHANG M., CHAN D., 2002, Toughening of a brittle thermosetting polymer: effects of reinforcement particle size and volume fraction, *Journal of Materials Science*, **37**, 781-788
9. VACA-CORTES E., LORENZO M.A., IRSA J.O., WHEAT H.G., CARRASQUILLO R.L., 1998, Adhesion testing of epoxy coating, Research Report No. 1256-6, Texas Department of Transportation
10. WETZEL B., HAUPERT F., FRIEDRICH K., ZHANG M.Q., RONG M.Z., 2001, Mechanical and tribological properties of particulate and nanoparticulate reinforced polymer composites, *Proceedings of the 13th ECCM Brugge*, Belgium
11. ZHAI L., LING G., LI J., WANG Y., 2006, The effect of nanoparticles on the adhesion of epoxy adhesive, *Materials Letters*, **60**, 3031-3033
12. ZHANG T., BAO Y., GAWNE D.T., MASON P., 2011, Effect of a moving flame on the temperature of polymer coatings and substrates, *Progress in Organic Coatings*, **70**, 45-51
13. ZHOU H., LIU H.Y., ZHOU H., ZHANG Y., GAO X., MAI Y.W., 2016, On adhesive properties of nano-silica/epoxy bonded single-lap joints, *Materials and Design*, **95**, 212-218

*Manuscript received October 31, 2019; accepted for print February 12, 2020*