

## **Hybrid composite materials on basis of reactoplastic matrix reinforced with textile fibres from process of tyres recycling**

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**Abstract.** The paper deals with a testing of composite materials reinforced with fabric, which were obtained after a recycling process of used tyres and a matrix is on a base of reactoplastics. The aim of the research was to set a possible utilization of unsorted textile waste from the process of the tyres recycling in the area of the polymeric composite systems. The subject of performed experiments was the hybrid polymeric composite, whose continuous phase was in a form of a two-component epoxy adhesive and a discontinuous phase (reinforcing particles) in a form of Polyamide PA (fibres) and rubber particles (granules of different sizes). An influence of a tensile stress, an elongation and an impact strength on the newly suggested hybrid composite materials were experimentally tested.

**Key words:** hybrid polymeric composite, mechanical properties, morphology of fibres, tyre recycling.

### **INTRODUCTION**

Used tyres, or tyres, whose parameters do not meet the requirements specified by the relevant rules of the road safety are recovered or disposed in accordance with the law. A principle of the ecological liquidation of tyres is a separation of various types of materials from which the tyres consist of. The output products of the recycling are the recycled rubber and other parts of the tyres that mean textile fibres and steel wires. Nowadays the number of worn tyres reaches 10 millions each year in all the world (Fang et al., 2000; Valášek et al., 2013). Technologies dealing with the recycling of products from the ecological liquidation of tyres have been constantly developing.

We can obtain a valuable raw material - crushed rubber from used tyres. It is widely used as an ingredient in asphalt, concrete filler, layer base of roads, rail crossings, coatings, paints, running tracks, playgrounds etc. Separate steel parts are also used in engineering and metallurgical industries. The last component of used tyres is the fabric. Fibres from the process of the tyres recycling are of polyamide how it is visible from the results of the thermal analysis (Parres et al., 2009).

A shattering of tyres for a purpose of gaining a granulate is one of effective possibilities of their recycling. This material is possible to use as a filler for a production of other rubber products and composite materials. An utilization of the textile waste is problematic because of a contamination with the rubber granulate. Various exploiters of this waste reach considerable differences in a composition of the textile

waste / rubber granulate (Parres et al., 2009; Knapčíková et al., 2014). It depends on a production technology and on a degree of purification in cyclone. A reason for that is a fact that this waste does not belong among primarily gained secondary raw-material.

The epoxy resins are typical reactoplastics. They dispose of three-dimensional structure. They show higher rigidity, strength and heat stability. The epoxy resins are brittle and prone to the initiation of cracks (Valášek, 2014; Valášek et al., 2014). The epoxy resin properties can be changed by adding appropriate types of fillers. Shi Ai Xu et al. (Shi Ai Xu et al., 2013) present an example of the modification of the epoxy resins by means of a liquid rubber (CTBN liquid rubber). They reached an improvement of failure properties of the epoxy resins.

Reactoplastics are used for the material recycling of various types of the waste (Valášek & Müller, 2014).

The aim of performed experiment was to describe the change of mechanical qualities of the epoxy resin filled with the recycled rubber and textile fibres from process of tyres recyclation with changeable amount of the filler.

A basic assumption for an optimum choice of materials is the knowledge of the applied material behaviour. The aim of the research was to set a possible utilization of unsorted textile waste from the process of the tyres recycling in the area of the polymeric composite systems. The research was realized according to the requirements of two significant firms dealing with the tyres recyclation in the Czech Republic.

## MATERIALS AND METHODS

The subject of performed experiments was the hybrid polymeric composite, whose continuous phase was in a form of a two-component epoxy adhesive (RAPID F) and a discontinuous phase (reinforcing particles) in a form of Polyamide PA (fibres) and rubber particles (granules of different sizes).

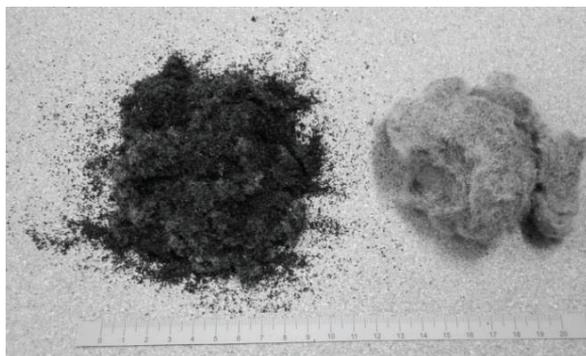
Epoxy resins are suitable for filling with organic as well as inorganic particles (Valášek & Müller, 2014). The adhesive RAPID F is a low-molecular epoxy resin prepared from bisphenol A and epichlorhydrin. It is distinguished for high liquidity and increased speed of a hardening. The processing time is till 10 minutes at the temperature  $23 \pm 5$  °C. The hardening is reached at least in 24 hours. The resin secondarily hardens during several days. The hardening of the resin is an exothermic reaction. The hardening process was watched by means of a thermocamera. The resin temperature before mixing was ca 22 °C. The temperature increased to 28 till 30 °C after mixing of the resin and the hardener. After adding the filler into the matrix the temperature ranged in the interval 29 to 34 °C. The matrix was chosen according to the requirement of the firm using this type of the resin in the production. The fibres were not treated before mixing with the resin. This requirement came out from an expected practical application. It was required only the storing temperature ranging in the interval 15 to 27 °C.

An influence of a tensile stress, an elongation and an impact strength on the newly suggested hybrid composite materials were experimentally tested.

The concentration of the components was expressed in volume percentage.

Two products of significant firms dealing with the tyres recyclation process were used for the research (Fig. 1). The fillers in the Fig. 1 were prepared by two different producers and each of the fillers contains different ratio of the rubber and the textile fibres. A different representation of the rubber fraction portion followed from the

analysis of these two products. The evaluation was performed by means of a sieve analysis.



**Figure 1.** Filler (left: filler for composites A, B, C; right: filler for composites D, E, F).

Following composite systems were tested in the research (Table 1)

**Table 1.** Tested variants of composite systems

Composite type	Mass of filler (a rubber, a fibre) (g)	Characteristic – volume ratio (fibre/rubber/matrix) (%)
Composite A	2	0.18/1.85/97.98
Composite B	5	0.44/4.64/94.95
Composite C	10	0.88/9.22/89.90
Composite D	2	1.61/0.72/97.67
Composite E	5	4.03/1.79/94.18
Composite F	10	8.06/3.58/88.36

A mean of recycled fibre and rubber particles was evaluated on a basis of a picture analysis. The evaluation was performed in a microscope Jenavert PA HD with a camera ARTCAM 300 MI. A width of the fibre was  $16.32 \pm 4.21 \mu\text{m}$ . The length of the fibre was  $2667.47 \pm 2079.47 \mu\text{m}$ . Also Parres et al. (2009) state a huge variability of the length of recycled fibres. The average size of rubber particles was  $410.38 \pm 146.30 \mu\text{m}$ .

The hybrid composite systems were tested from two points of view. The first one was the material testing. The reason is an application in a form of a production of self-contained products. The second point of view – in the interaction with the adherent that means the adhesive bond. The reason is a hypothetical utilization as the filled adhesive.

By mixing of the specified matrix – filler phases ratio the composite was made, which was used for the preparation of test specimens according to the specified standards. The composite mixture was left for the total hardening for 330 hours. The secondary hardening of the composite mixture was ensured.

Fifteen test specimens were always prepared for testing. The test specimen was not tested in the case that a defect was ascertained by a visual check. Own testing was performed at 10 pieces of test specimens. The surface roughness was measured on the profilograph SurfTest 301 (a value of cut off was 0.8 mm). The surface roughness of the

matrix was Ra  $0.29 \pm 0.05 \mu\text{m}$ , Rz  $1.80 \pm 0.42 \mu\text{m}$  in the bottom part of the specimen and Ra  $0.48 \pm 0.14 \mu\text{m}$ , Rz  $2.85 \pm 0.66 \mu\text{m}$  in the upper part of the specimen. The surface roughness of the composites A to F was  $0.37 \pm 0.10 \mu\text{m}$ , Rz  $2.17 \pm 0.37 \mu\text{m}$  in the bottom part of the specimen and Ra  $1.20 \pm 0.41 \mu\text{m}$ , Rz  $5.90 \pm 2.09 \mu\text{m}$  in the upper part of the specimen.

The theoretical density of the composite systems was calculated on the basis of the physical relationships, the real density was stated on the basis of the ratio of weight and volume of the composites (Berthelot, 1998). An important first-class quality of the composite system – porosity (P) was calculated according to the equation (1):

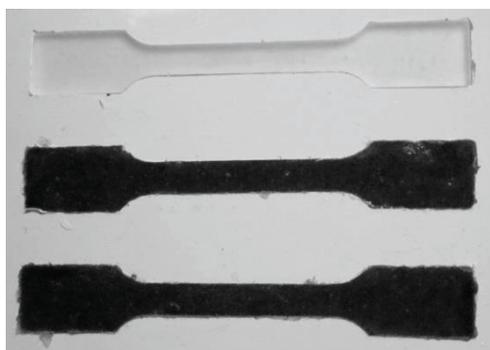
$$P = \frac{\rho_{The} - \rho_{Rea}}{\rho_{The}} \quad (1)$$

where: P – porosity (%);  $\rho_{The}$  – theoretical composite density ( $\text{g cm}^{-3}$ );  $\rho_{Rea}$  – real composite density ( $\text{g cm}^{-3}$ ).

The porosity of the composites A to F was 5.6 to 8.1%. The porosity was caused by a presence of air bubbles in the matrix and by the distribution of the filler in the matrix.

**Tensile test:** The test specimens for the tensile properties determination according to the standard CSN EN ISO 527-1 (Plastics – Determination of tensile properties – Part 1: General principles) were prepared according to the standard CSN EN ISO 3167 (Plastics – Multipurpose test specimens). By the destructive testing the tensile strength and the elongation were determined.

The moulds for casting of the test specimens were made from the material Lukapren N using prepared models. The shape and sizes of moulds meet the corresponding standards (Fig. 2).



**Figure 2.** Mould and casts of test specimens for tensile test.

**Adhesive bonds:** The basis of adhesive bonds laboratory testing was the determination of the tensile lap-shear strength of rigid-to-rigid bonded assemblies according to the standard CSN EN 1465 (Equivalent is BS 1465).

Specimens of all the tested materials were obtained identically – cutting from the semi-products in the hydraulic guillotine sheet metal machine. Laboratory tests of the adhesive bonds were performed using the standard test specimens made according to the standard CSN EN 1465 (dimensions  $100 \pm 0.25 \times 25 \pm 0.25 \times 1.6 \pm 0.1 \text{ mm}$  and lapped length of  $12.5 \pm 0.25 \text{ mm}$ ) from the constructional plain carbon steel S235J0.

The surfaces of 1.5 mm thick steel sheets were at first blasted using the synthetic corundum of a fraction F80 under the angle of  $90^\circ$ . Using the profilograph Surftest 301 the following values were determined: Ra  $1.28 \pm 0.12 \mu\text{m}$ , Rz  $6.2 \pm 0.86 \mu\text{m}$ .

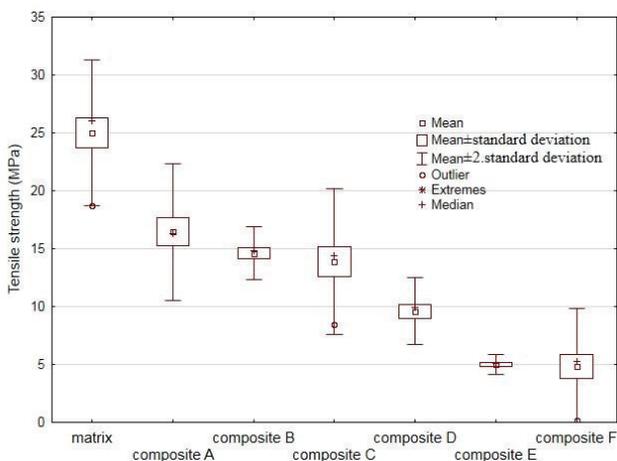
Then the surface was cleaned and degreased using acetone and prepared to the application. The surface preparation is important and should guarantee good strength on the boundary adherent/adhesive/adherent (Novák, 2011; Hricová, 2014; Legutko et al., 2014). An even thickness of the adhesive layer was reached by a constant pressure 0.5 MPa. The lapping was according to the standard  $12.5 \pm 0.25$  mm.

The tensile strength and the elongation test (the adhesive bond, the cast of the test specimens) were performed using the universal tensile strength testing machine LABTest 5.50ST (a sensing unit AST type KAF 50 kN, an evaluating software Test&Motion). A speed of the deformation corresponded to  $6 \text{ mm min}^{-1}$ .

**Impact strength:** The impact strength was set in an apparatus Dynstat determined for the testing of plastics. The test specimen preparation and impact tests were performed according to the standard CSN 64 0611 (Determination of the impact resistance of rigid plastics by means of Dynstat apparatus). By the destructive testing the impact strength was determined.

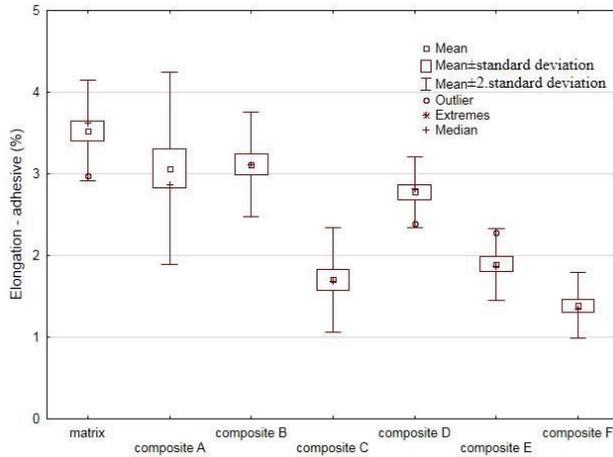
## RESULTS AND DISCUSSION

Tensile strength of composites showed lower values compared with the matrix (the epoxy adhesive) – see Fig. 3. The composite systems A, B and C showed higher tensile strength than the composite system D, E and F because composites A, B, C consist of smaller percentage of the fibres. It is obvious from the results that the textile fibre acts the tensile strength in a negative way.



**Figure 3.** Tensile test – CSN EN ISO 527-1 – tensile strength of matrix and composite systems.

Fig. 4 shows the results of the tests focused on the evaluation of the elongation of the matrix and composite systems. It is obvious from the results that higher ratio of the filler decreases the elongation values.



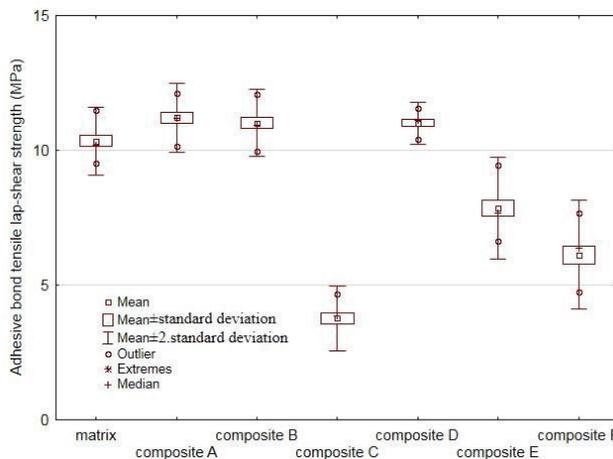
**Figure 4.** Tensile test - CSN EN ISO 527-1 – elongation of matrix and composite systems.

The tested sets were mutually compared using F-test from the point of view of the influence of various filler concentrations of cast test specimens on the tensile strength and the elongation.

The zero hypothesis  $H_0$  presents the state when there is no statistically significant difference ( $p > 0.05$ ) among tested sets of data from their mean values point of view.

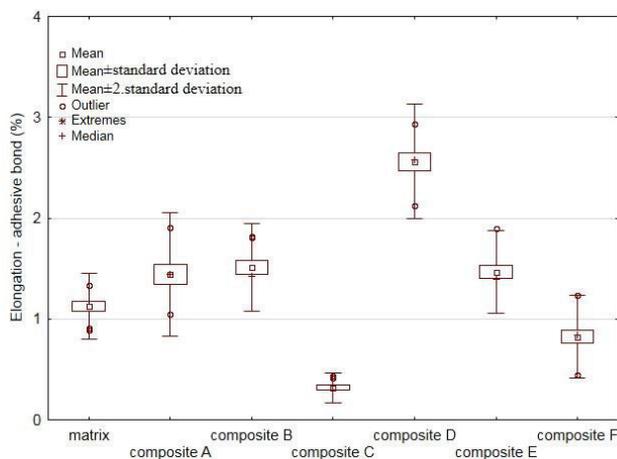
The tensile strength ( $p = 0.0000$ ) and the elongation ( $p = 0.0000$ ) did not certify the hypothesis  $H_0$ , so there is the difference among particular tested filler concentrations in relation to the adhesive bond strength and to the elongation in the reliability level 0.05.

It is obvious from the strength results of the adhesive bonds that the composite systems A, B and D reach higher values of the adhesive bond tensile lap-shear strength (Fig. 5) than the matrix. The strength results of the interaction with the adhesive bonded material (the adherent) show different behaviour compared with the results of the tensile test.



**Figure 5.** Adhesive bond tensile lap-shear strength – CSN EN 1465.

The composite systems A, B, D and E proved higher values of the adhesive bond elongation (Fig. 6). Higher concentration of the filler in the area of the adhesive bonds acts the resultant tested mechanical properties of the adhesive bond in the negative way (Fig. 6).



**Figure 6.** Elongation of adhesive bond – CSN EN 1465.

The tested sets were mutually compared using F-test from the point of view of the influence of various filler concentrations on the tensile lap-shear strength and the elongation of the adhesive bonds.

The zero hypothesis  $H_0$  presents the state when there is no statistically significant difference ( $p > 0.05$ ) among tested sets of data from their mean values point of view. The adhesive bond tensile lap-shear strength ( $p = 0.0000$ ) and the elongation of the adhesive bond ( $p = 0.0000$ ) did not certify the hypothesis  $H_0$ , so there is the difference among particular tested filler concentrations in relation to the adhesive bond tensile lap-shear strength and the elongation in the reliability level 0.05.

Fig. 7 shows the results of the impact strength. The positive influence of the filler on the impact strength of the composite system is visible from the results. Higher values of the impact strength are reached at the composites D, E and namely F. These composites contain higher volume ratio of the textile fibres.

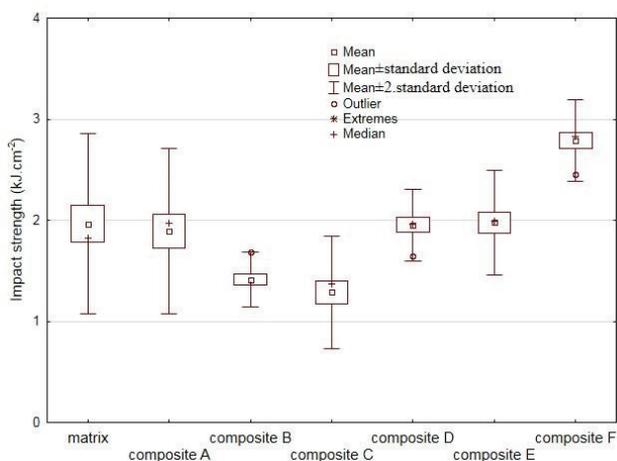
The tested sets were mutually compared using F-test from the influence of various filler concentrations on the impact strength point of view.

The zero hypothesis  $H_0$  presents the state when there is no statistically significant difference ( $p > 0.05$ ) among tested sets of data from their mean values point of view. The impact strength ( $p = 0.0000$ ) did not certify the hypothesis  $H_0$ , so there is the difference among particular tested filler concentrations in relation to the impact strength in the reliability level 0.05.

Results of Dadfar and Ghadami showed that it came to improving the fracture toughness owing to increasing content of the rubber modifier (Dadfar & Ghadami, 2013). It is possible to agree with the results. The filler in the form of the rubber granules and the textile waste has a similar function.

On the basis of laboratory results it is possible to agree to the statement of Jiao Weizhou et al. that epoxy adhesives are of low impact strength and that they are brittle (Jiao Weizhou et al., 2009).

The presumption was certified that the presence of rubber particles lower the tensile strength. The mechanical properties showed throughout negative trend (Ferreira et al., 2013; Kejval & Müller, 2013; Valášek, 2014; Valášek & Müller, 2014). These conclusions were confirmed by the results of the research. Adhesive bonds with maximum 15 vol.% of the filler do not show the statistically significant fall of the tensile lap-shear strength. The experiment results proved the increase of the adhesive bond strength.



**Figure 7.** Impact strength – CSN 64 0611.

## CONCLUSIONS

The knowledge of applied materials behaviour is the basic presumption of the optimum material choice.

The research results proved the possibility to use the waste textile fibres and the rubber granules in the composite systems on the basis of the reactoplastics.

Following statements can be said in the end:

- The tensile strength was decreasing. The fall of the tensile strength was 80.1% at the application of the filler. The composite systems A, B and C (they contain higher percentage of the rubber) showed the strength fall of 34 till 44%. The composite systems D, E and F showed the fall of 61 till 80%.
- The elongation of the composite systems was of no explicit trend.
- The tensile lap-shear strength showed the increase (the composites A, B and D).
- The impact strength showed increased values at the composite systems which contained higher ratio of the textile fibres.

The composite material can be used at a renovation of machine parts, for a production of new products e.g. a roofing material.

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