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Mechanical properties of polymer matrix composites filled with Jatropha Curcas L.

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Abstract. Polymers and their composites are widely used for their specific properties. This paper deals with composite materials based on Jatropha oil cake. Seeds of Jatropha Curcas L. plant are pressed for gaining oil. A cake is a by-product.
A polymer composite was a subject of performed experiments. A continuous phase was in a form of a two-component epoxy adhesive and Jatropha oil cakes were a discontinuous phase (reinforcing particles).
Using Jatropha oil cake as the by-product after the mechanical extraction of the oil decreases a price of the composite system. Jatropha oil cake is the waste which is not further utilized.
The research was focused on various weight concentrations from 5 to 30%. The volume energy, the strength characteristics and the impact resistance were tested at these composite materials. Results were evaluated by means of the statistical program ANOVA. The filler moisture was 4.59 ± 0.22% WB. Mechanical properties were not changed using of the filler.
The negative influence of the filler was ascertained at the tensile strength. The positive influence was ascertained at the impact strength until 20 wt% of the filler concentration.
Owing to the high complexity of the failing process of the composite material the grid electron microscopy in the area of the fracture surface was used.

Key words: Adhesive, seeds, Jatropha oil cake, statistica.

INTRODUCTION

With regard to the environment and a high level of exhaustion of oil resources, biopolymers have a great scientific attention in recent years. Polymer composites reinforced with natural fibres have been developed over the last decade as sustainable alternatives of engineering applications. According to Kumar & Sharma (2008) the growing interest in the development of composites has become more effective thanks to the growing demand for low-cost renewable materials which could replace the traditional ones. In addition, the high price of synthetic fibres, the public awareness and the sensitivity to the preservation of the natural environment contribute to the development of new research activities in this area (Satheesh et al., 2009; Müller et al., 2015).

Naturally renewable energy has many advantages, such as the availability of feedstock, the environmental friendliness and the low-cost (Ahmed et al., 2001; Aigbodion & Pillai, 2001). Firstly, these products must comply with the technical and

industrial standards of a durability, an exposure stability, a chemical resistance, etc. They must also comply with all environmentally appropriate standards (Galusek et al., 2007). A number of vegetable oils were used for the synthesis of various polymer resins, such as polyester, epoxy, polyurethane, polyester amide, etc. (Herák et al., 2013; Ružbarský et al., 2014).

The reason to incorporate the filler into the polymer is dual: firstly, to improve the tribological, mechanical and thermal properties, secondly, to reduce the cost of the final composite.

Jatropha curcas L. is a versatile plant with diverse use, such as biodiesel, medicines, cosmetics, etc. Jatropha is a drought resistant bush or tree widely distributed in the wild and cultivated form in some areas of Central and South America, Africa and South-East Asia (Cano-Asseleih, 1986; Ružbarský et al., 2014). The first commercial applications of Jatropha curcas L. were in Lisbon where the oil was imported from Cape Verde. It was used for the manufacture of soaps and for lamps (Galusek et al., 2007).

Jatropha oil cake yield is approximately about 500–600 g kg^{-1} of Jatropha seeds (Syed et al., 2009). Oil cakes contain a large amount of minerals (Satheesh et al., 2009).

Shivamurthy & Murthy (2014) describe in the experiment the strength and the rigidity of the epoxy matrix, which is increased by an addition of microparticles prepared from the residues of Jatropha curcas L. seeds after pressing. It is necessary to describe the properties of composites with particles from renewable sources (biomaterials) and to determine the area of the application of these materials (Ružbarský et al., 2014).

Nanocomposites reinforced with 1 wt.% of expanded graphite showed an improvement by 15% in the module of elasticity compared to a pure epoxide (Mishra et al., 2000).

Rosso et al. (2006) evaluated the mechanical strength of epoxy resins with the addition of 5 vol.% of silicon dioxide nanoparticles. The silicon dioxide addition was able to improve the rigidity and the toughness of the polymer. As a consequence the module of elasticity was increased by 20% and the fracture toughness by 70%.

Isik et al. (2003) and Yasmin et al. (2003) studied the effects of a nanoclay added to the epoxy resin from 0 to 10 wt.%. Isik et al. found that the tensile strength was increased with the addition of 1 wt.% of the nanoclay. However, the module of elasticity was increased gradually with the addition of clay nanoparticles. Yasmin, et al. (2003) found an increase by 80% in the module of elasticity by addition of 10 wt.% of clay nanoparticles.

Subramanian & Sun (2006) examined the effect of the nanoclay with the addition of 0, 3, 5 and 8 wt.% to the epoxy resin. The compressive strength of composites was increased by 22% and 36% with the addition of 3 wt.% and 5 wt.% of the nanoclay.

This paper deals with the mechanical properties of Jatropha oil cakes, such as the tensile strength, the impact strength and the volume energy. The matrix is a two-component adhesive and a filler (discontinuous phase) is Jatropha oil cakes. The research objective was the utilization of the material waste from the Jatropha urcas L. pressing process. The secondary objective was to find an application which improves the mechanical properties of the composite.

MATERIALS AND METHODS

The object of the experiments was a particle polymer composite. A continuous phase was in the form of a two-component epoxy adhesive ChS Epoxy 324 Epoxy 1200 (hardener P11 – Diethylenediamine) and the discontinuous phase (reinforcing particles) in the form of Jatropha oil cakes (Fig. 1). The filler was obtained as a waste from the seeds of Jatropha curcas L. during the presswork process. This waste of the process (cakes) was dried to a moisture content 4.59 ± 0.22% WB and subsequently the size of the particles was adjusted by crushing. The filler thus obtained has not been further modified, e.g. fractionated.

The size of the particles was measured by means of an optical analysis at the stereoscopic microscope. A great variability of the filler in the form of the microparticles is apparent from performed 100 measurements. The dimension was 435 ± 256 μm.

The concentration of subcomponents was determined and expressed in weight percentages. Test specimens were made in the concentrations of 5, 10, 20, 30, 40 and 50 wt.% of Jatropha oil cake microparticles.

Figure 1. Jatropha oil cakes.

By mixing of the specified matrix – filler phases ratio the composite was made. It was used for the preparation of test specimens according to the specified standards.
The moulds for casting were made from the material Lukapren N using models. The form and size of moulds meet the corresponding standards.

The composite mixture was let to be fully cured under time 340 h. The secondary curing of the composite mixture was provided. The influence of the impact resistance, the tensile strength and the volume energy was experimentally investigated.
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, Czech Standard Institution). By the destructive testing the tensile strength σ was ascertained.

To determine the relationship between a tension force and a deformation, a device (Labortech, MPTest 5.050, Czech Republic) was used to record the course of a deformation function. The tensile test was performed according to CSN EN ISO 527-2 (2012). A deformation speed at the tensile test was 6 $mm.min^{-1}$. Ascertained values of

tensile forces were transformed by means of an equation 1 to the tensile stress and deformations were transformed by means of an equation 2 to the relative deformation.

$$\sigma = \frac{F}{S} \tag{1}$$

where: σ – tensile stress in sample (MPa); F – tensile force (N); S – appropriate cross section area of sample (mm^2).

The volume deformation energy was set as an area below a curve 'stress – strain' from zero to a maximum value of the deformation according to an equation 2.

$$\lambda = \sum_{n=0}^{n=i-1}\left[\left(\frac{\sigma_{n+1}+\sigma_n}{2}\right)\cdot\left(\varepsilon_{n+1}-\varepsilon_n\right)\right] \tag{2}$$

where: λ – volume energy ($J\ m^{-3}$); i – indicates the additional amount of strain in which the stress was determined (step of measurement – 0.001 mm), -; σ_n – tension stress at appropriate strain (MPa); σ_{n+1} – tension stress at the sequential strain (MPa); ε_n – strain (-); ε_{n+1} – sequential strain (-).

The impact strength was ascertained 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 a_n was determined. The calculation was performed according to the equation (3)

$$a_n = \frac{A_n}{b.h} \tag{3}$$

where: a_n – impact strength ($kJ\ m^{-2}$); A_n – energy consumed to breaking up test specimen (kJ); b – width of test specimen (m); h – thickness of test specimen (m).

The destructive testing conducted at the laboratory temperature 22 ± 2 °C.

Statistical hypotheses were also tested at measured sets of data by means of the program STATISTICA. A validity of the zero hypothesis (H_0) shows that there is no statistically significant difference ($p > 0.05$) among tested sets of data. On the contrary, the hypothesis H_1 denies the zero hypothesis and it says that there is a statistically significant difference among tested sets of data or a dependence among variables ($p < 0.05$).

RESULTS AND DISCUSSION

Fig. 2 shows the results of the tensile strength. A negative influence of the filler based on Jatropha oil cake microparticles is visible from the results of the experiment. The fall of the tensile strength caused by the addition of Jatropha oil cake microparticles ranged in the interval 65 to 80%. The tensile strength fall was more significant with increasing concentration of the filler.

Fig. 3 shows the results of the volume energy. A negative effect of the filler based on Jatropha oil cake microparticles is obvious from the experimental results. The fall of the module of elasticity caused by adding of Jatropha Oil Cake microparticles ranged in

the interval 75 to 92%. The fall of the volume energy was more significant with increasing concentration of the filler.

Fig. 4 shows the results of the impact strength. A positive effect of the filler based on Jatropha Oil Cake microparticles till 20 wt.% is obvious from the experimental results. The increase of the impact strength was 14% at the concentration 20 wt.%. At the concentration 30 wt.% the fall of the impact strength of 21 to 37% occurred.

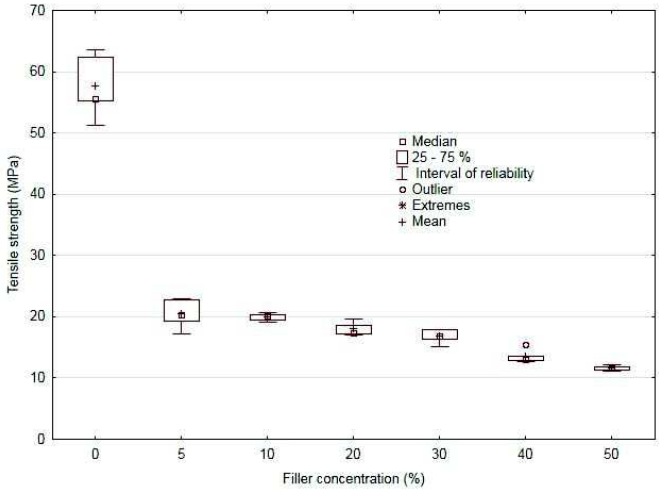

Figure 2. Effect of filler based on Jatropha oil cake microparticles on tensile strength.

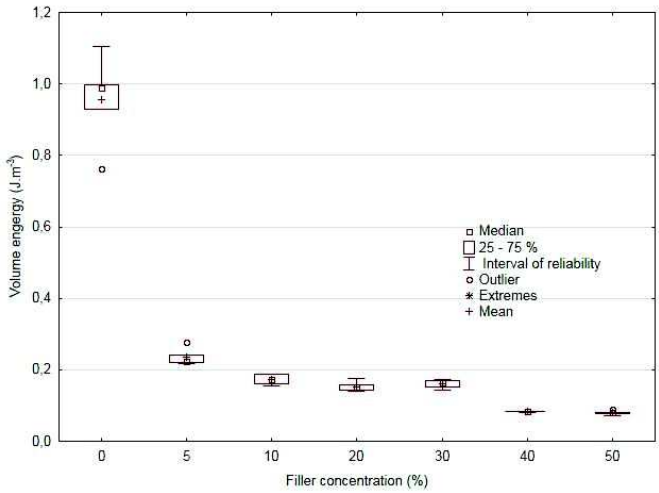

Figure 3. Effect of filler based on Jatropha oil cake microparticles on volume energy.

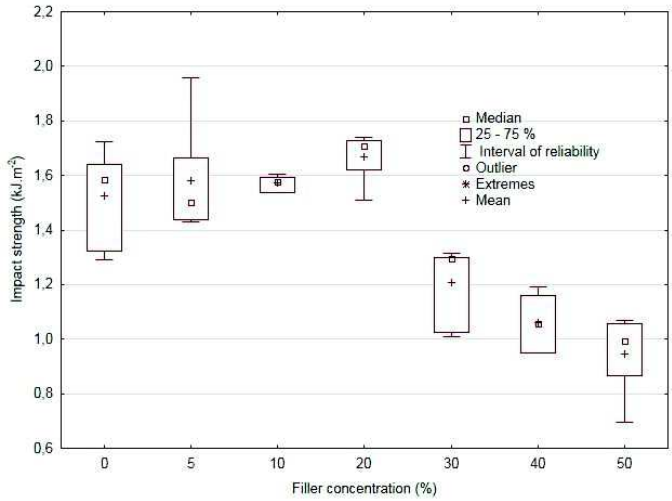

Figure 4. Effect of filler based on Jatropha oil cake microparticles on impact strength.

In terms of the filler of Jatropha oil cake influence on mechanical properties of the polymer composite material the results of ANOVA F-test are following:

- Tensile strength: at comparing all variants of the experiment the hypothesis H_0 was not certified ($p = 0.0000$), i.e. there is a difference among single tested concentrations of the filler microparticles 5, 10, 20, 30, 40 and 50 wt.% of Jatropha oil cakes and the matrix in the significance level 0.05. So, the influence of the concentration of the filler based on Jatropha oil cake microparticles on the tensile strength was statistically proved in the significance level 0.05.
- Volume energy: at comparing all variants of the experiment the hypothesis H_0 was not certified ($p = 0.0000$), i.e. there is a difference among single tested concentrations of filler microparticles 5, 10, 20, 30, 40 and 50 wt.% of Jatropha oil cakes and the matrix in the significance level 0.05. So, the influence of the concentration of the filler based on Jatropha oil cake microparticles on the volume energy was statistically proved in the significance level 0.05.
- Impact strength: at comparing all variants of the experiment the hypothesis H_0 was not certified ($p = 0.0000$), i.e. there is a difference among single tested concentrations of filler microparticles 5, 10, 20, 30, 40 and 50 wt.% of Jatropha oil cakes and the matrix in the significance level 0.05. So, the influence of the concentration of the filler based on Jatropha oil cake microparticles on the impact strength was statistically proved in the significance level 0.05.

During the experiments the assumption about the negative effect of the filler on the tensile strength was confirmed. Cho et al. (2006) indicate that the strength of the composite is reduced with increasing volume of the filler particles.

According to the research by various authors the addition of fillers to the epoxy adhesive does not produce a definite improvement or deterioration in impact strength. E.g. Dadfar & Ghadami (2013) indicate an improvement in the toughness due to the increased content of the rubber modifier. Further, it was proved that the aluminium microparticles also increased the impact strength.

The investigation showed that the epoxy adhesive retained the adhesion of the adhesive and the filler up to 50 wt.%.

Jiang-Jhy et al. (2001) proved in their experiments that the irregular shape of the particles ensured good interaction between the matrix and the filler. Farraf et al. (2008) ascertained that the irregular shape of the particles showed reaching smaller mechanical properties probably owing to the separation between the matrix and the filler. Good interaction between the filler and the matrix is visible from Figs 5 and 6. It is obvious from the result that it did not depend on the shape of the filler.

Jatropha curcas L. seeds contain a high percentage of oil which is widely used, e.g. it is used at a production of biodiesel (Samsuri & Zoveidavianpoor, 2014). The filler based on Jatropha oil cake microparticles contains residual concentration of oil. This fact made perfect wetting of the filler surface with the matrix impossible. So, it is obvious from the results that the shape and the type of the filler are essential.

A wettability of the filler with the matrix is important at filling structural two-component epoxies with the filler. The research results confirmed also ca. 2–3% porosity of created material.

Figs 5, 6 and 7 show a fracture surface of the polymer composite based on the filler in the form of Jatropha oil cake microparticles with using the electron microscope Tescan Mira 3. Bad interaction of the filler with the matrix in the form of the epoxy was proved within the experimental research by use of the electron microscopy (Figs 5, 6). Fig. 5 shows ordering of the filler microparticles.

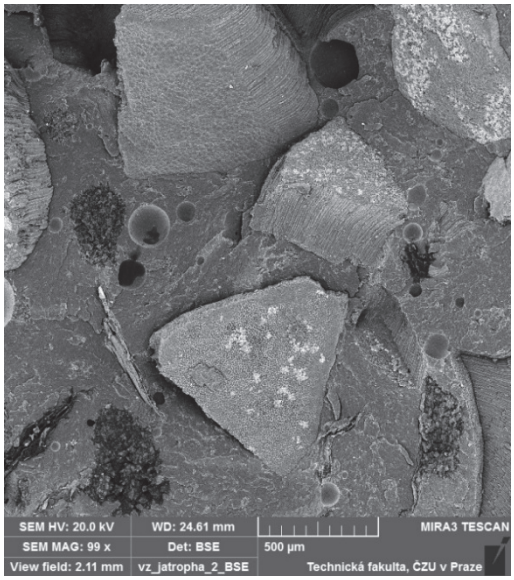

Figure 5. SEM images – fracture surface of composite based on Jatropha oil cake microparticles (composite 40 wt.% of filler).

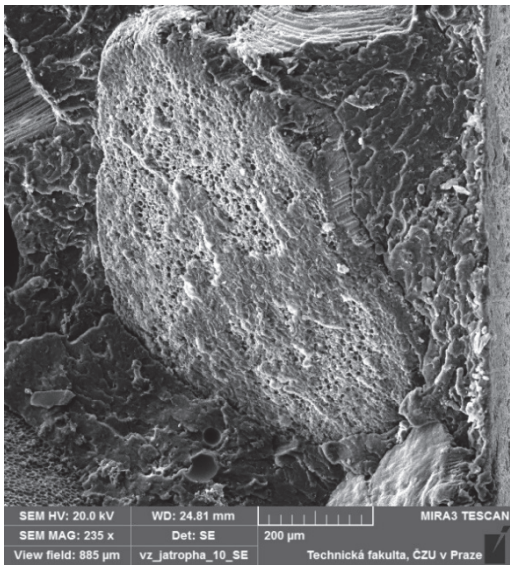

Figure 6. SEM images – fracture surface of composite based on Jatropha oil cake microparticles (filler particles in form of kernel).

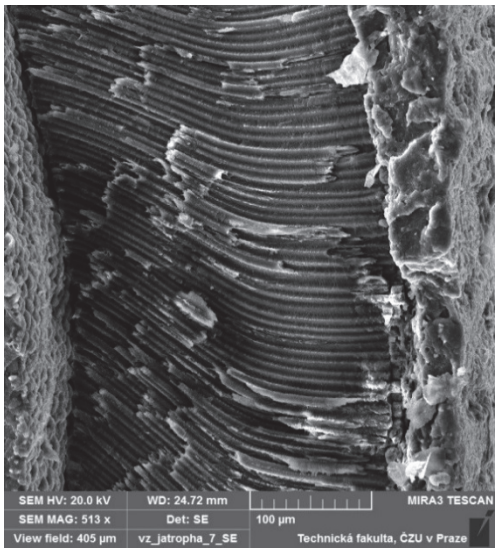

Figure 7. SEM images – fracture surface of composite based on Jatropha oil cake microparticles (filler particles in form of peel).

CONCLUSIONS

Based on the experiment it is possible to establish these conclusions:

- The results of the experiments show an obvious negative impact of the filler based on Jatropha oil cake microparticles on the tensile strength and the volume energy.

A significant decrease was of approximately 90% with increasing concentration of the filler based on Jatropha oil cake microparticles.

- The results of the experiments show an obvious positive impact of the filler based on Jatropha oil cake microparticles on the impact strength. This positive trend can be monitored up to 20 wt.%. This positive effect was due to a modification of the matrix, i.e. a structural epoxy with the filler which contained residual concentration of oil.

Increased dynamic strain resistance of the polymer particle composite system is positive. Polymer materials are characterized by reduced dynamic stress resistance. This causes problems in practice.

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