EPDM rubber material utilization in epoxy composite systems

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Abstract. Observing of possibilities for secondary raw materials utilization should rank among the key interests of the society. Nowadays, there are a lot of modern workplaces which are devoted to the possibilities of collecting, processing and using rubber materials. EPDM waste rubber in the form of particles is one of the many products of these workplaces. One of the possibilities for recycling this waste particles material is their interaction with another polymeric material. A reactoplastic which is filled with these particles comes into consideration. This way of utilization of the material is inexpensive and simple. The paper focuses on chosen mechanical qualities of the Epoxy/EPDM waste rubber composite. The waste rubber was gained as one of the outputs of a recycling line of the firm Gumoeko, Ltd., the reactoplastic was represented by a two-component epoxy resin. Distraction of rubber particles in the epoxy matrix was achieved by mechanical mixing without using the technology of vacuum. In the paper, the porosity, tensile strength and shear strength of the composites with various concentrations of EPDM are described. The resulting composite systems may find their application in the field of agriculture - especially during joining and sealing materials of larger units where high quality connections are not required.

Key words: agriculture, mechanical properties, reactoplastic, waste.

INTRODUCTION

EPDM rubber (ethylene propylene diene monomer (M-class) rubber) is an example of a synthetic elastomer. EPDM rubber finds application in a number of industrial branches owing to its properties (high resistance to weather influences, etc.), it is used for production of washers and tubes which are applied in the area of the automotive industry. It can be also found in a number of various machines which are applied in agriculture (tractors, hardvestors, etc.). Waste EPDM rubber can be processed on modern lines whose outcomes are EPDM particles or powder. One of the possibilities for using waste inorganic particles is their inclusion into plastics – this is material utilization. Waste material recycling in the form of plastics filler is common. Materials with hard inorganic particles (SiC, Corundum) which are abrasive wear resistant can be mentioned in the agriculture (Valasek & Muller, 2012; Muller & Valasek, 2013b). Interaction of waste fillers with epoxy resins can lead to an increase in materials suitable for adhesive bonding as well as cementing (Valasek & Muller, 2013a). For example, Ku et al. (2010) used glass powder (5–12.5 wt %) with the dimension of 2–32 μm for increasing the values of a tensile strength. It is necessary to realize that adhesive bonding is a method of joining which is used in agriculture – see the cooperation between the companies Henkel and New Holland (Muller and Valasek, 2013a). Only the possibility of filling epoxy resins with waste particles and the possible application of these materials in agriculture are the topics of this paper.

Many authors have dealt with the possibilities of filling polymeric materials with waste rubber. For example, Subramaniyan et al. (2012) used waste rubber particles for improving the mechanical properties of a polyurethane foam filled with Kenaf fibres. Using waste rubber together with fibers is also described by Cerbu and Curtu (2011) who increased the impact strength of a composite on a basis of an epoxy resin, glass fibers, and waste rubber particles. Valasek et al. (2013) describe the comparable values of the shear strength of the bonds created by an unfilled epoxy resin and by a resin filled with waste rubber particles of the size of 27.6 ± 14.2 μm. The properties of epoxy resin can be changed by adding of appropriate types of fillers. Dadfar and Ghadami (2013) used carboxyl-terminated butadiene acrylonitrile for optimizing the failure properties of an epoxy resin reinforced by glass fibres. Xu et al. (2013) present an example of the modification of epoxy resins by means of a liquid rubber (CTBN liquid rubber).

The aim of the performed experiment is to describe the influence of EPDM waste rubber on a change in the mechanical properties of two-component epoxy resins which are commonly used for joining materials in agriculture and are easily fillable with inorganic fillers. The paper defines the change in the tensile strength and lap-shear tensile strength and describes the porosity and hardness of the achieved materials. The performed experiment defines potential application areas of the achieved material, not only in the area of agricultural machines – it defines the cohesive strength of the material and its adhesive strength in the boundary of a steel adherent and an adhesive.

MATERIALS AND METHODS

Preparation of test samples

Rubber EPDM granulate was gained from the company Gumoeko, Ltd. The granulate is produced by mechanical shattering of the waste from the production of washers and tubes for the automotive industry, above all. The mixture of the granulate is disposed of impurities during processing. The producer states a possibility for using the granulate for sports surfaces, isolations and paddings. This type of granulate is not dangerous waste. A particular size of particles was measured by using a stereoscopic microscope.

The rubber EPDM granulate was added to epoxy resins Eco Epoxy 1200/324 (EE) and Glue Epox rapid (GR). They are two-component resins which are suitable for filling with inorganic types of fillers. The filler concentration in the epoxy resins was expressed by volume percentages: 5–20 vol.%. Dispersion of the rubber particles was achieved by mechanical mixing. The test samples were prepared by casting (into forms made from a silicone rubber) according to relevant standards and were hardened according to the technological requirements of the resins producer.

Laboratory tests

The test samples determined for specification of the cohesive strength by means of the tensile strength were prepared according to the requirements of the standard CSN EN ISO 3167. The test samples were tested on a universal testing machine. The speed of the cross beam motion was 6 mm min^{-1}. The setting of the tensile characteristics was performed in accordance with the standard CSN EN ISO 527 (see Fig. 1).

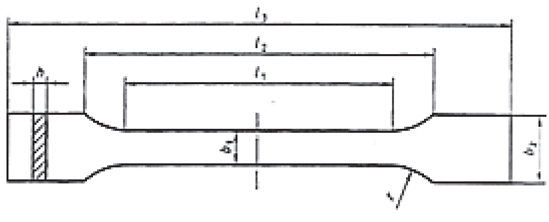

Figure 1. Multipurpose-made test sample – Tensile strength (CSN EN ISO 3167, 2004).

For the lap-shear strength description in the boundary of the adherent and the filled system, overlapped assemblies were made (CSN EN 1465, see Fig. 2). Surface preparation is important (Novak, 2011). The surface of 1.5 mm thick steel sheets (S235J0) onto which the composite system was applied was at first blasted using the synthetic corundum of the fraction F80 under the angle of 90°. In this way, the average surface roughness of Ra = 1.39 ± 0.36 µm, Rz = 9.9 ± 0.31 µm was reached. Then the surface was cleaned and degreased using perchlorethylene and prepared for the composite mixture application.

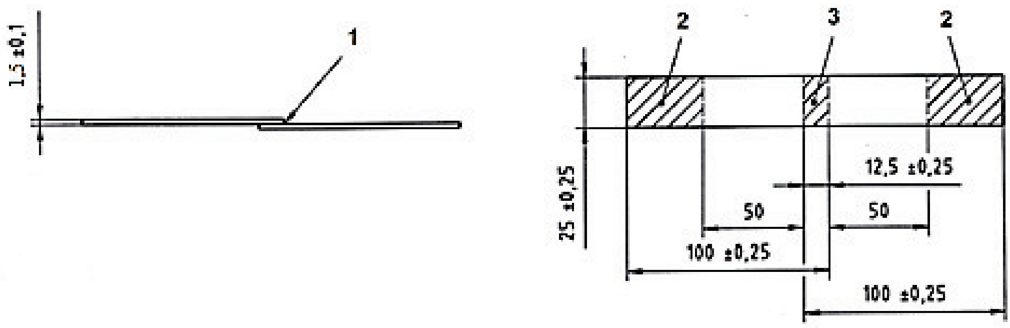

Figure 2. Test sample – Lap-shear tensile strength (CSN EN 1465, 1997).

The evaluation of the failure of the test sample surface and adhesive layers was performed using a stereoscopic microscope. A statistical evaluation of the results was performed by means of the programme Statistica – one-factor ANOVA, reliability level $\alpha = 0.05$. For the statistical comparison, the T-test was used when the zero hypothesis H_0 ($p > 0.05$) states an agreement of the statistical sets of data.

RESULTS DISCUSSION

The density of the EPDM rubber waste particles corresponded to 1.12 g cm^{-3}, the density of Eco Epoxy 1200/324 was 1.15 g cm^{-3} and the density of Glue Epox Rapid 1.15 g cm^{-3}. The mutual interaction between the resin and the particles is influenced by their morphology. Only the interphase interaction is the key in filled materials. The

EPDM waste particles were subjected on the stereoscopic microscope. The average value of particle size was 1,092 ± 403 μm. Fig. 3 presents the distribution of particle sizes and their morphology.

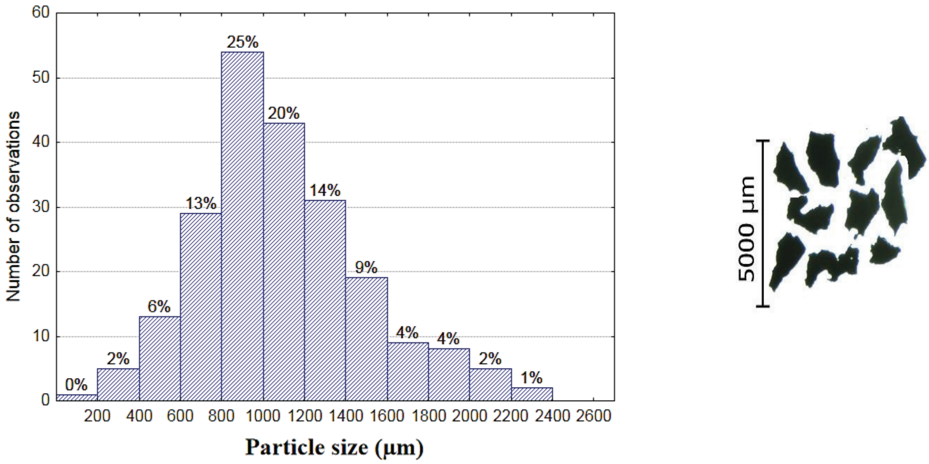

Figure 3. Histogram – size of the EPDM waste rubber particles and their morphology.

The strength of the filled epoxy resins in the boundary of the steel adherent and the adhesive was found by the methodology of testing the adhesive bonds shear-strength (CSN EN 1465). The failure always occurred in the adhesive bond, it was the adhesive failure in all cases – so the weak place of the bond was situated directly in the boundary of the steel adherent and the filled epoxy resin. All filled systems showed considerably lower shear strength compared to unfilled epoxy resin (see Fig. 4).

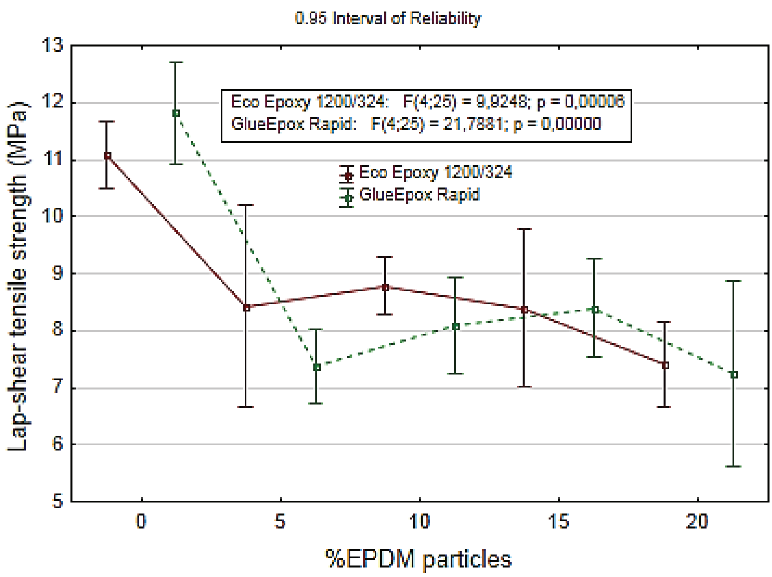

Figure 4. Lap-shear tensile strength.

The statistical tests of the data were tested by the F-test, where an agreement of dispersion variances was confirmed in all cases. Consequently, the T-test was used for comparison of the data sets for particular types of filled epoxy resins (see Table 1). The zero hypothesis H_0 describes confirmation of the hypothesis – there is no statistically significant difference among the compared sets of data.

Table 1. T-test – Lap-shear tensile strength

T-test		Eco Epoxy 1200/324				
H_0:($p > 0.05$)		0	5	10	15	20
GlueEpox	0		0.00	0.00	0.00	0.00
	5	0.00		0.62	0.97	0.20
	10	0.00	0.12		0.50	0.00
	15	0.00	0.03	0.53		0.14
	20	0.00	0.85	0.26	0.13	

The cohesive strength of filled epoxy resins was evaluated by the means of the tensile strength (CSN EN ISO 3167, see Fig. 5). These values were considerably lower compared to unfilled resin.

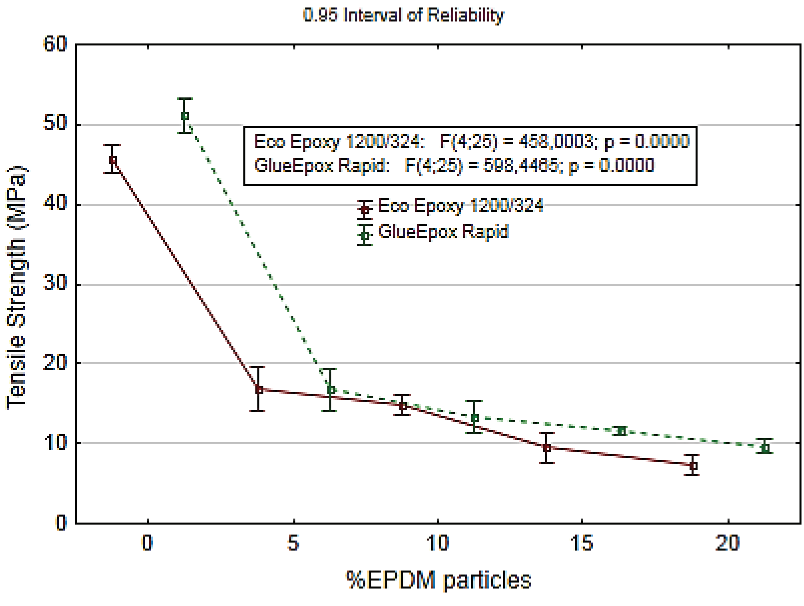

Figure 5. Tensile strength.

The statistical sets of data were compared again (see Table 2). The F-test did not confirm the agreement of dispersion variances in some cases, in this case, a two-selective T-test with dispersion variances inequality was used.

Table 2. T-test –Tensile strength

T-test	H_0:(p > 0.05)	Eco Epoxy 1200/324 0	5	10	15	20
GlueEpox	0		0.00	0.00	0.00	0.00
	5	0.00		0.12	0.00	0.00
	10	0.00	0.02		0.00	0.00
	15	0.00	0.00	0.06		0.03
	20	0.00	0.00	0.00	0.00	

Fig. 6 shows a typical failure area of filled epoxy resins. The epoxy matrix is brittle – the failure starts directly in the matrix. EPDM rubber particles are tenacious and they are capable of considerable elastic deformation (as is usual for elastomers). At the moment of maximum carrying capacity of the test sample, it comes to the failure of the matrix at first and consequently to the failure of elastic particles. However, it is obvious from the experiment that increasing the concentration of EPDM rubber particles considerably decreases the tensile strength of the material – it decreases the cohesive strength of the system.

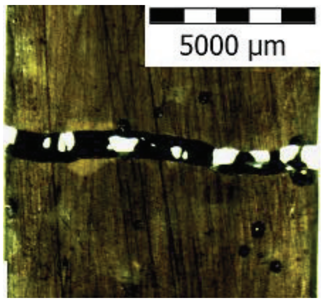

Figure 6. Failure of a brittle matrix with filler (Glue Epox with 20% of EPDM rubber particles).

The inclusion of EPDM rubber particles led to the rapid fall of the lap-shear tensile strength and the tensile strength of both reviewed epoxy resins. The lap-shear tensile strength of the Eco Epoxy resin decreased from the value 11.08 ± 0.47 MPa to the value 7.41 ± 0.64 MPa (at Glue Epox from 11.82 ± 0.72 MPa to 7.24 ± 1.41 MPa). High standard deviations are worth noticing too – the variation coefficient of 19.5%. The sheer fall of the tensile strength was proportional to the filler concentration in the matrix. The tensile strength decreased from the value 45.75 ± 1.48 MPa to the value 7.32 ± 1.16 MPa for Eco Epoxy (for Glue Epox, from 51.17 ± 1.87 MPa to 9.67 ± 0.75 MPa). The variation coefficient did not exceed 15%. The abovementioned confirms the fact that the presence of particles considerably decreases the observed characteristics. Air pores were easily identified under the stereoscopic microscope when evaluating the failure areas by a picture analysis. The presence of pores is related to the preparation technology of the test samples when no special technology (e.g. vacuum) was used and this was owing to the effort to minimize costs. The air pores can lead to a decrease of mechanical properties.

The presence of waste particles decreases the final price. So long as we consider zero price of the waste raw material, the particles used decrease the price of epoxide at the concentration 20% up to 24% (this can be up to € 3.38 at the considered retail price of the epoxy resin Glue Epox Rapid, see Fig. 7). The lines handling the waste material utilization do not offer these products at zero prices, however, the offered price per 1 kg is considerably lower than the price of the epoxide.

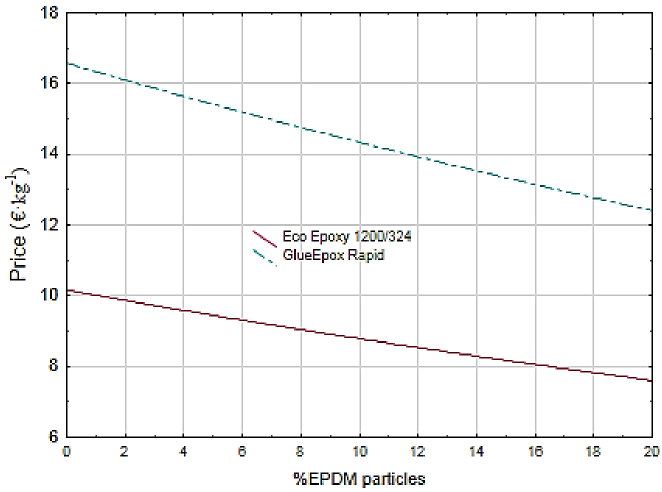

Figure 7. Description of the price of filled systems.

CONCLUSIONS

The results stated above narrow the spectrum of utilization of these materials. It is not possible to use these materials for the bonds and applications requiring high strength and reliability owing to their properties. In the area of agriculture, these materials can, above all, prospectively be used for cementing the adhesive bonding of larger units where high strength of the bond is not required. An example can be utilization of the material for floors where the resistance of epoxides is an advantage (Mleziva, 1993). The material can also be used for filling larger cracks and for shaping of imbalances. The performed experiments confirmed the assumption of authors Xue Qunji and Wang Qihua (1997) that filler application in epoxy resins considerably influences the final properties. The rubber particles used considerably decreased the shear strength of the overlapped adherents compared to the experiment of Valasek et al. (2013) who used particles of the size 27.6 µm. According to Ku et al. (2010), inorganic microparticles can improve the tensile strength in some cases, however, this is not the case in this experiment, where the boundary of the particles and the resin was prone to creation of cracks and it came to decreasing the strength. Optimization of mechanical properties would be possible in combination with another filler (e.g. with fibres), as Subramaniyan et al. (2012) state. It is true, the mechanical properties are low, but the described way of utilization is material utilization of the waste also generated in the production and liquidation of agricultural machines. The limits of the material with EPDM waste rubber of the middle particle size higher than 1,000 μm can be summed up in the following way:

- the inclusion of 20% of EPDM particles led to the decrease of the lap-shear strength of 33% for Eco Epoxy and of 39% for Glue Epox Rapid,
- the inclusion of 20% of EPDM particles led to the decrease of the tensile strength of 84% for Eco Epoxy and of 81% for Glue Epox Rapid.

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