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Development of Composites Based on Thermosetting Polymers (Epoxy, Polyester and Vinylester Resins) with Cenosphere Fillers in Different Concentrations – 5 wt.%, 10 wt.% and 15 wt.%

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***Abstract*—Experiments were conducted to develop lightweight composites based on thermosetting polymers with cenosphere fillers. Cenospheres are rich in Al and Si particles and are known as low-density aluminosilicate CFA microspheres. The production of a polymer-based structural material (composite) with reduced weight and good mechanical properties, in which a waste product polluting the environment is used as a filler, is a current engineering task. Epoxy, polyester and vinylester resins were used, as well as different filler concentrations (cenospheres – 5 wt.%, 10 wt.% and 15 wt.%). The mechanical properties of the developed composites were studied. It was found that the use of cenospheres positively affects the mechanical properties of the vinylester resin, while in the case of other thermosetting plastics an increase in some of the mechanical characteristics was registered.**

Keywords*—thermosetting polymers, cenospheres, mechanical properties

I. Introduction

Composite materials are the basis for the development of new efficient products. In industry, polymer-based composites and light alloys are widely used due to their strength and low density. The combination of reduced weight and good mechanical properties make composites the preferred materials for various engineering solutions. They have found wide application in automotive, aerospace, marine and civil engineering.

During the production of electricity from thermal power plants, huge amounts of coal fly ash are generated annually which leads to environmental pollution. Coal fly ash contains heavy metals that can be released upon contact with water [1], minerals such as magnetite, calcite, mullite [2], as well as silica and cristobalite [3]. The furnaces in thermal power plants operate at high temperatures and therefore structural differences are observed in the fly ash particles: plerospheres, ferrospheres and cenospheres (CS) [4]. Plerospheres are particles containing gases, minerals and many smaller particles [5], ferrospheres are spherical particles rich in iron [6], and cenospheres are rich in aluminium and silicon [3]. These Al and Si-rich particles are known as aluminosilicate CFA microspheres [3].

Cenospheres are hollow particles with a spherical shape and their size ranges from 5-500 µm [7], with an average size in the range of 30-350 µm [8, 9]. They have

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found application in various fields: construction [8], building materials [10], paints and coatings [11], ceramics [12], plastics [13], polymer fillers [14], etc. In [15] the structural, thermal, mechanical and dynamic properties of polypropylene-based composites with cenosphere fillers were investigated. The development of syntactic foam based on high-density polyethylene (HDPE) and cenosphere fillers was presented in [16]. The dielectric properties of a composite of low-density polyethylene (LDPE) and cenospheres were investigated in [17].

The purpose of this study is to study the possibility of developing composites based on thermosetting polymers and cenosphere fillers.

II. Materials and methods

Epoxy, polyester and vinylester resins were used to conduct the experiments. The epoxy system "LETOXIT" is based on epoxy resin for lamination "LETOXIT" PR 227 and hardener "LETOXIT" EM 315. Due to its low viscosity and low surface tension, the used resin system wets the laminating fabrics well, and therefore is used for the production of laminates with glass fibers, carbon or aramid fibers. The manufacturer's prescription was followed for the preparation of the resin system, namely 100:38 mass proportions when mixing (resin:hardener). The polyester resin used is DISTITRON 429 BSX25Q, the hardener is MEKR50 in an amount of 2 wt.%, and the amount of accelerator MEKR used is also 2 wt.%. The vinyl ester resin is DISTITRON VE 100, the hardener is MEKR50 and again the amount used is 2 wt.%.

Cenospheres in amounts of 5 wt.%, 10 wt.% and 15 wt.% with sizes in the range of 50-250 µm, with the main fraction having sizes of 100 µm (Fig. 1), were used to develop the composites.

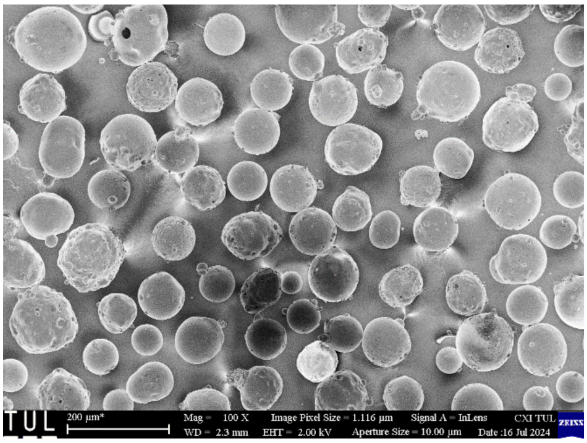

Fig. 1. Image of the cenospheres used.

The composition of the ceramic shell of the cenospheres used was also investigated (Fig. 2 and Fig. 3).

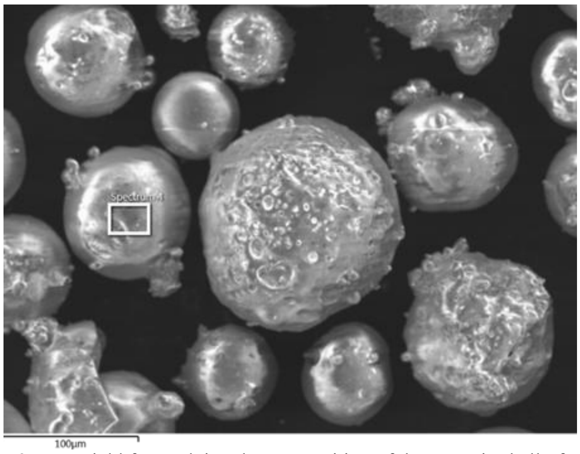

Fig. 2. Field for studying the composition of the ceramic shell of cenospheres

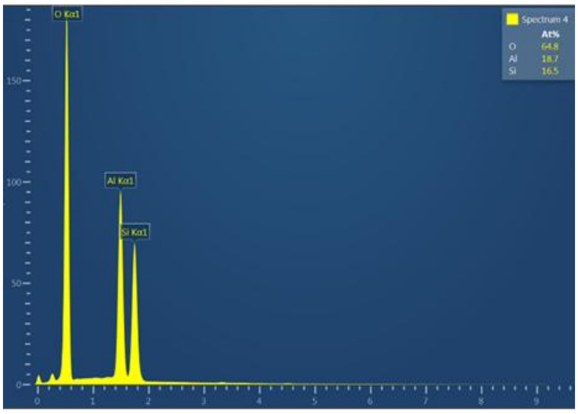

Fig. 3. Chemical composition of the ceramic shell of the used cenospheres.

The results show that the ceramic shell of the used cenospheres consists of silicon-16 at.%, aluminium - 18.7 at.% and oxygen - 64.8 at.%.

The investigated composites based on the thermosetting polymers and aluminosilicate spheres were produced by mechanical stirring at a working temperature of 25ºC (according to the manufacturers' instructions for working with the epoxy resin system). Test specimens were gravity cast for tensile testing and impact toughness testing. Before conducting the mechanical tests, the samples based on epoxy resin were subjected to secondary polymerization at a temperature of 55ºC for 12h. (resin manufacturer's instructions).

The uniaxial tensile test was performed on a Zwick Roell Z050 testing machine, the impact toughness test was performed with an Izod GT-7045-HM device under the following conditions: Izod Impact Test, 1J (10.20 kg.cm), Method - reversed notch, Speed 3.46 m/sec., and the hardness of the composites was measured using the Shore method (scale D - HSD). This method for determining hardness is elastic-dynamic, i.e. the tip is lowered from a certain height and the height of the rebound is used to determine the hardness of the material, using a table showing the value of the hardness versus the rebound value. The measurement was performed using an EQUOTIP hardness tester.

III. Results and discussion

The results of the mechanical tests conducted on the epoxy resin-based composites are shown in Table 1.

TABLE 1 RESULTS OF MECHANICAL TESTS OF EPOXY RESIN-BASED COMPOSITE

Cenospheres [wt. %]	**Rm [MPa]**	**A [%]**	**HSD**	**E/A (kg.cm/cm^2)**
0 wt.%	49	0.8	76.7	1.60
5 wt.%	35.3	0.7	84.7	0.89
10 wt.%	29.2	0.8	87.1	0.88
15 wt.%	26.2	1.7	82.3	0.96

The results obtained show that the pure resin system has the highest values of tensile strength and impact toughness. The addition of 5 wt.% filler leads to a decrease in tensile strength and impact toughness, the elongation at break is like that of the pure polymer, but the hardness of the composite is increased. The highest hardness was measured in the composite with a filler of 10 wt.%, but the tendency to decrease the Rm value is also maintained in it. The impact toughness and the elongation at break are with values identical to the same characteristics as in the composite with 5 wt.% cenospheres. The use of 15 wt.% aluminosilicate spheres as a filler leads to a decrease in the Rm values, the hardness also decreases, but its values are higher compared to those of the pure polymer. A significant increase in the elongation was registered and a tendency to increase the impact toughness value is observed.

The results of the mechanical tests conducted on the polyester resin-based composites are shown in Table 2.

TABLE 2 RESULTS OF MECHANICAL TESTS OF A POLYESTER-BASED COMPOSITE

Cenospheres [wt. %]	**Rm [MPa]**	**A [%]**	**HSD**	**E/A (kg.cm/cm^2)**
0 wt.%	47.8	0.0	79.8	0.32
5 wt.%	29.3	0.0	81.7	0.30
10 wt.%	27.1	0.0	64.8	0.31
15 wt.%	19.8	0.0	55.9	0.26

The results show that the addition of cenospheres to the polyester resin matrix leads to a decrease in both tensile strength and hardness, with only a slight increase in the hardness of the composite being recorded when 5 wt.% filler is used. When 5 wt.% and 10 wt.% cenospheres are used, the impact toughness values of the composite are comparable to the value of the pure resin, and with an increase in the percentage of filler (15 wt.%) the impact toughness decreases.

The results of the mechanical tests conducted on vinylester resin-based composites are shown in Table 3.

TABLE 3 RESULTS OF MECHANICAL TESTS OF A VINYLESTER-BASED COMPOSITE

Cenospheres [wt. %]	**Rm [MPa]**	**A [%]**	**HSD**	**E/A (kg.cm/cm^2)**
0 wt.%	20.5	0.0	75.7	0.375
5 wt.%	33.3	1.3	71.1	0.506
10 wt.%	26	1.7	43.5	0.537
15 wt.%	20	0.7	40	0.443

The results of the studies of composites based on vinylester resin with aluminosilicate sphere filler show a tendency of decreasing hardness values with increasing percentage of filler in the composite. In contrast to the studies with epoxy and polyester resin, with vinylester resin an increase in tensile strength, elongation at break and impact toughness of the composites was registered compared to the same parameters of the pure resin. Only with the composite with 15 wt.% filler was registered a comparable tensile strength compared to the pure polymer.

The experimental results obtained for the studied composites based on the thermosetting polymers with cenosphere fillers are not unambiguous. In the composites with a polyester resin matrix, the amounts of filler used do not positively affect the studied properties. Although the tensile strength and impact toughness of the composites based on epoxy resin are reduced compared to the pure polymer, the filler has a positive effect on the plasticity of the material. The most noticeable positive effect of the different concentrations of cenospheres is registered in the composites based on vinylester resin. A decrease in the hardness of the composites compared to the pure polymer is registered, but the tensile strength and impact toughness are increased. The amounts of filler used have contributed to imparting plasticity to a hard and brittle material. Given that epoxy and vinyl ester resins are commonly used for the production of laminates, expanding the scope of research with reinforcing fibers such as glass fibers, carbon or aramid fibers is appropriate.

IV. Conclusions

Lightweight composites based on thermosetting polymers (epoxy, polyester and vinylester resins) with cenosphere filler in different concentrations – 5 wt.%, 10 wt.% and 15 wt.% have been developed. It has been established that the filler used has a positive effect on the mechanical properties of the vinylester resin, while in the case of other thermosetting plastics an increase in some of the mechanical characteristics has been registered. The obtained results are a prerequisite for expanding the scope of research with the presented composites in order to develop laminates with reinforcing fibers such as glass fibers, carbon or aramid fibers.

The development of lightweight composites with a thermoplastic polymer matrix and aluminosilicate microspheres as filler is the subject of future research.

The development of composites with a polymer matrix and cenosphere filler is a current engineering task. Obtaining new structural composite materials, in which the filler is a waste product and an environmental pollutant (cenospheres), which can be used in mechanical engineering, automotive engineering, aerospace, marine and civil engineering will also have a positive environmental effect.

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