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INFLUENCE OF SONICATION ON GRAPHITE AND GLASSY CARBON PARTICLE SIZE DISTRIBUTION IN EPOXY MATRIX AND MECHANICAL PROPERTIES OF PRODUCED COMPOSITES

In the following study the influence of the ultrasonic treatment of graphite and glassy carbon powder reinforcement on epoxy composites was examined. Sonication treatment was applied to ethanol dispersions of graphite and glassy carbon respectively. After ultrasound treatment the dispersions were dried at the temperature of 70°C. Subsequently, the graphite and glassy carbon powders were mechanically extracted. The produced powders were then analyzed – the grain size distributions of the pre- and post-treatment powders were compared. The results show that the grain sizes of the sonicated graphite decreased, while the glassy carbon particles were not significantly influenced. Epoxy resin composites were made with the pre- and post-treatment powders as reinforcement. The mechanical properties of the prepared composites were examined using a Brinell hardness tester and a tensile tester. The results show slight changes in the mechanical properties of the composites reinforced with the sonicated powders in comparison to the non-sonicated powders and the neat resin samples.

Keywords: epoxy composite, glassy carbon, sonication, powder treatment, mechanical properties

INTRODUCTION

Ultrasonics are commonly used in many branches of research and industry. The most common application of ultrasonic waves can be found in ultrasonic washers and ultrasonic non-destructive material testing; however, there is a far greater, vast potential of ultrasonic radiation applications. Sonication processes use ultrasonic waves to influence materials, often in the form of powder dispersions in liquids, to change their properties, particle size and/or to cause reactions in the said materials. Particularly important is the process of ultrasonic exfoliation of graphene flakes from graphite. By using sonication, it is possible to break graphite powder through the prolonged influence of ultrasonic irradiation into a dispersion of graphene nanoplates in liquid. The liquids used in exfoliation include but are not limited to: ethanol [1], water [2, 3], DMF [4, 5], NMP [4, 6], as well as different acids and salts [4, 7, 8]. Other parameters also have a direct influence on the sonication process such as the addition of other materials into the dispersion. For example, Navik and others studied the addition of curcumin, which promoted graphene exfoliation [9]. The parameters of sonication such as time and energy have been thoroughly studied and optimized [4-7]. The effects of ultrasonic radiation on the erosion of graphite flakes were also subject to study [10, 11]. Other processes combine ultrasonics with more steps to optimize the manufacturing process and increase the quality of the obtained material [12]. Sonication can also be used in composite preparation [13, 14], in supporting catalytic reactions [15, 16] and for oxidation reactions [17]. Beyond typical sonication, ultrasonics can support processes like ultrasonic spray atomization [18] and ultrasonic assisted flotation [19]. Another important branch of ultrasonics dependent processes is sonochemical synthesis. An example of a sonochemically synthesized material can be SbSI (antimony sulfoiodide) piezoelectric nanowires used in research, often as part of a composite sensor for strain measurement [20, 21], energy harvester [21] or acoustic power sensor for ultrasonic reactors [22]. The amount of research and common usage of ultrasonics to support material manufacturing and processing show the viability and capabilities of the sonication route for application in material modification.

Glassy carbon is an allotrope of carbon characterized by its amorphous structure, its 2D planar structural elements and high proportion of sp^2 hybridization among its atoms in ambient conditions [23]. Most of the research and application for this material can be found in electrochemistry and catalysis [23]; however, glassy carbon powder can find more diverse applications due to its unique properties. Specifically, the usage of glassy carbon in composites for tribological applications and in hybrid composites has been exten-

sively researched recently [24-27]. The cited literature shows that glassy carbon powder can work as a self-lubricating agent within the composite structure during friction. Research into the ultrasonic treatment of glassy carbon particles has already been done by Levêque et al., who proved that sonication treatment can provide small amounts of glassy carbon nanoparticles dispersed in a supernatant [28].

Epoxy composites reinforced with carbon particles are examined in many different ways for their potential applications in tribological and mechanical applications. Nevertheless, it is very important to provide a satisfactory and repeatable mechanical performance, which often is hard to obtain in composites with non-homogenous reinforcement [29]. The unpredictability of the reinforcement distribution may cause undesirable anisotropy [30]. A successful and significant particle size decrease by applying sonication could provide a simple and viable method of increasing the quality of reinforcing carbon powders and lead to enhanced mechanical properties of their composites. Well-dispersed graphite can be used as relatively cheap and effective reinforcement for epoxy composites. The additional use of CNTs can increase the desired properties exponentially, though with a significant increase in the price of the material [31-34].

MATERIALS AND METHODOLOGY

The materials used were commercially available graphite powder (Biomus, Poland), glassy carbon foams made by Prof. Jerzy Myalski (Silesian University of Technology, Poland) by means of polyphenol resin pyrolysis, and Epidian 62 epoxy resin with the Z-1 curing agent (Ciech, Poland) used in 10 to 1 proportions. The glassy carbon foams were milled in a Fritsch Pulverisette 6 (Fritsch, Germany) high-energy planetary ball mill. Ethanol dispersions were made in glass beakers with 0.5 liter of 96% ethanol (Pol-Aura, Poland) and 15 g of each powder. The sonication processes were carried out on a VCX750 ultrasonic reactor (Sonics & Materials, Inc., USA) with 40% amplitude in 30 second cycles (10 seconds active, a 20-second pause) for 8 hours. The pre- and post-treatment powder particle size distribution was examined with a Malvern Mastersizer 3000 laser diffraction particle size analyzer (Malvern PANalytical, UK). Composite samples were made out of epoxy resin with an addition of 20% weight pre- and post-treatment graphite and glassy carbon powders, respectively. Additional neat resin samples were made for comparison with the prepared composites. Mechanical properties testing (static compression test) was carried out utilizing an Instron 4469 tensile tester (Instron, USA) with a 5 kN load and a loading velocity of 5 mm/min on cylindrical samples (18 mm diameter, 18 mm height). The hardness was measured using a Brinell hardness tester HK460 (Heckert, Germany) under a 365 N load on cylindrical samples (30 mm diameter, 10 mm height).

RESULTS AND DISCUSSION

The particle size distribution measurement results are presented below. The charts show the averages of the obtained results. Figure 1 shows the grain size distribution of the graphite powders – pre-treatment (non-sonicated) shown on top, and post-treatment (sonicated) shown below the first one. Figure 2 presents the grain size distribution of the glassy carbon powders – pre-treatment (non-sonicated) shown on top, and post-treatment (sonicated) shown below the first one. The x-axis (labeled Dx) represents the total percentage of powder particles that are of the diameter seen on the y-axis and smaller (i.e. for the non-sonicated graphite (Fig. 1, top chart), 90% of the particles (Dx = 90%) are 86 μm in diameter and smaller).

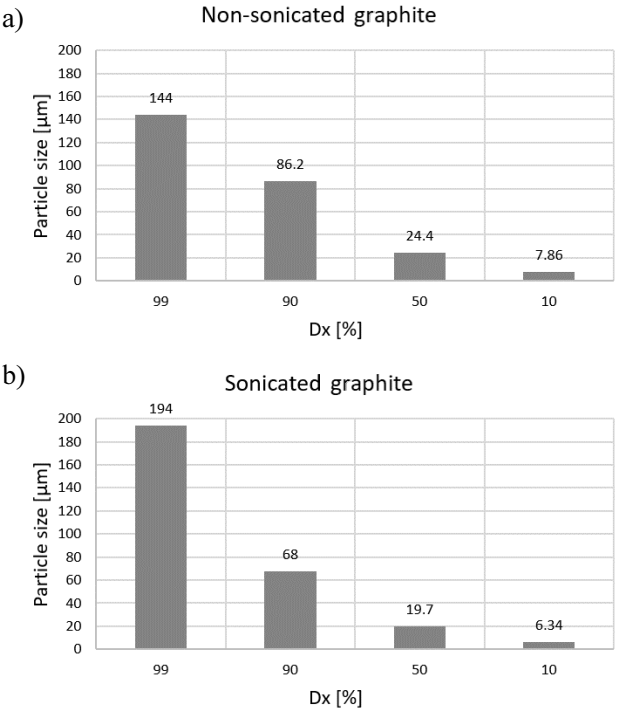

Fig. 1. Measured particle size distributions of graphite powder: a) pre-treatment (non-sonicated graphite), b) post-treatment (sonicated graphite)

As can be seen in the charts presented in Figure 1, the graphite powder particle size distribution changes significantly after the sonication process. While 9% of the largest particles (difference between Dx99 and Dx90) have a greater diameter for the non-sonicated graphite than for the sonicated one, most of the particles decreased in size after the sonication treatment. 90% of the particles are 21.1% smaller in diameter for the sonicated graphite in comparison to the non-sonicated graphite, 50% of the particles of sonicated graphite are about 19.1% smaller than their non-

sonicated counterparts, and 10% of the smallest particles of the sonicated graphite are 19.3% smaller than 10% of the smallest particles of non-sonicated graphite. Overall, the results show that the employed sonication decreases the sizes of 90% of the graphite particles by about 20% in comparison to the non-treated powder.

The glassy carbon particle size distribution shown on the charts above (Fig. 2) does not change significantly after sonication treatment. If anything, the measured particle sizes show a small increase for Dx90, Dx50 and Dx10 – increases by 5.9, 7.3 and 14.2%, respectively. The standard deviation of the performed measurements is below 1% of their value, which means that the increase does not result from measurement inaccuracy or error; however, differences of this scale for particles in the range of ~20 µm to 2.5 µm most likely will not result in a noticeable change in the mechanical parameters of the type of composites researched further in this paper.

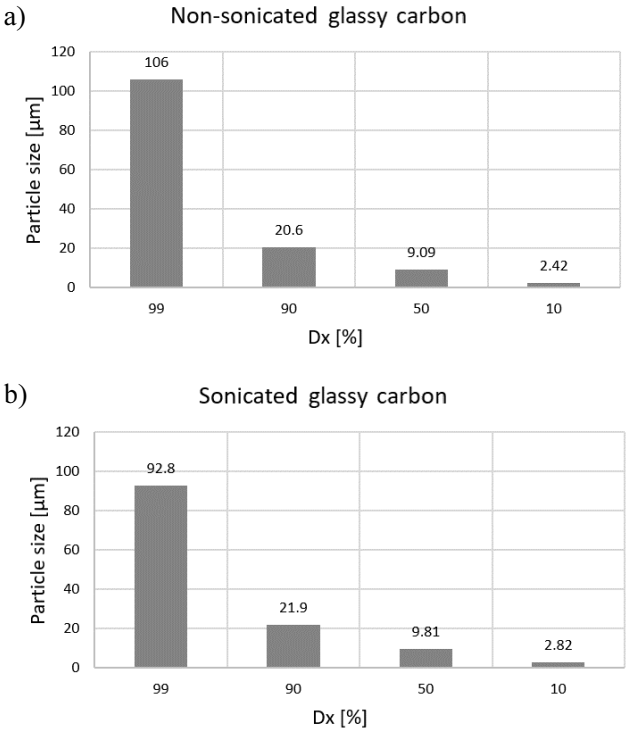

Fig. 2. Measured particle size distributions of glassy carbon powder: a) pre-treatment (non-sonicated glassy carbon) b) post-treatment (sonicated glassy carbon)

The static compression test was chosen due to the type of forces affecting elements working in tribological applications. The static compression test curves for each investigated material are presented in Figures 3 and 4.

The values of the compressive strength for the examined materials are presented in Table 1.

As can be seen from the figures and table, the addition of the examined powders to the resin used in the research resulted in a slight decrease in the compressive strength of all the composites, except for the sonicated glassy carbon, in which an increase in the compressive strength value of 0.22 MPa was noted. While the mechanical parameters were lowered by the addition of the researched powders, the materials used in this research could still prove viable for use in tribological applications owing to the fact that the presence of carbon powders acting as a self-lubricating agent might decrease the wear of the element. The influence of sonication on the examined powders seems inconclusive for the static compression test of the composites – the non-sonicated graphite composite is significantly stronger than the sonicated one, while the reverse is true for the researched glassy carbon composites.

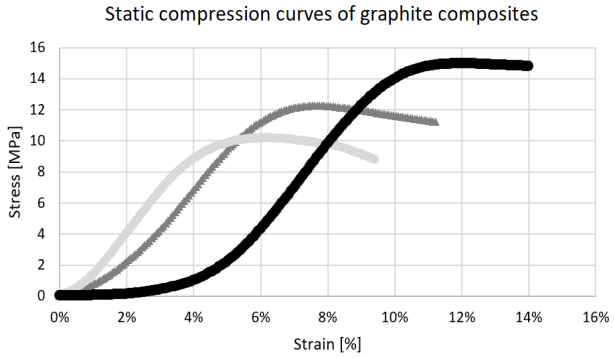

Fig. 3. Static compression test curves for graphite composites in comparison to neat resin (light gray curve – sonicated graphite, dark gray curve – non-sonicated graphite, black curve – neat resin)

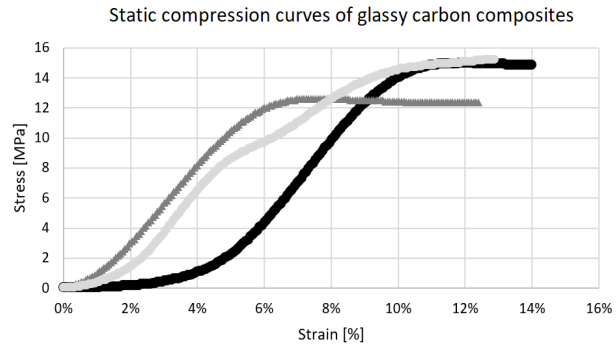

Fig. 4. Static compression test curves for glassy carbon composites in comparison to neat resin (light gray curve – sonicated glassy carbon, dark gray curve – non-sonicated glassy carbon, black curve – neat resin)

TABLE 1. **Results of static compressive test**

Material	Compressive strength [MPa]	Compressive strain [%]
Non-sonicated graphite	12.28±0.1	7.7
Sonicated graphite	10.19±0.1	6.1
Non-sonicated glassy carbon	12.60±0.1	7.5
Sonicated glassy carbon	15.21±0.1	12.9
Neat resin	14.99±0.1	12.1

Hardness testing was carried out on a Brinell hardness tester. The results are presented in Figure 5.

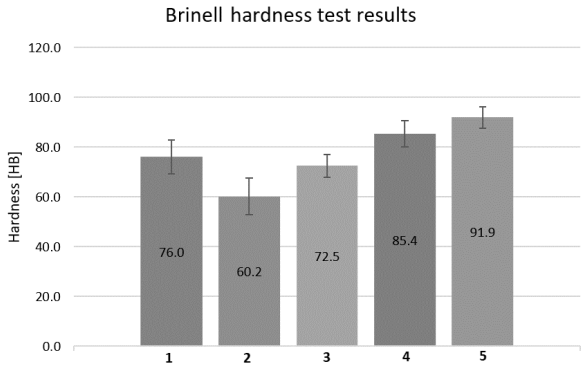

Fig. 5. Comparison of measured Brinell hardness values for examined materials: 1 – non-sonicated graphite, 2 – sonicated graphite, 3 – non-sonicated glassy carbon, 4 – sonicated glassy carbon, 5 – neat resin

The results of the hardness measurements indicate effects similar to the ones noted in the static compression test. The neat resin exhibits the highest hardness among the studied samples, with the HB values decreasing for the composite samples. The sonicated glassy carbon has the highest hardness among the examined composites, while the relations between the sonicated and non-sonicated powder composite samples are parallel to the static compression test, with higher measured values for the non-sonicated graphite composite than for the sonicated one, and the reverse for the glassy carbon composites.

CONCLUSIONS

The sonication process has a significant influence on the particle size and the particle size distribution for graphite powder. This results most likely from the graphite structure and its tendency to exfoliate. While the conducted treatment does not provide changes in a size range comparable to the processes employed for graphene exfoliation, it is still a change significant enough to influence the mechanical parameters of the composite materials. For the initially milled glassy carbon, sonication has little to no influence on the particle sizes or their distribution. The energy applied in the sonication process by cavitation does not seem to have a significant effect on glassy carbon, at least at the energy levels and in the time frames used in this research. Despite this, the composites made out of sonicated glassy carbon seem to have the highest mechanical properties among the researched composites – it seems to be a better solid state lubricant than graphite. The addition of 20% weight carbon powders to the resin used in this research decreased its mechanical properties; nonetheless, its tribological parameters might be positively influenced by carbon powders working similar to self-lubricating agents within the composite structure. Further research should be conducted to verify the viability of using the examined powders as fillers in tribological composites.

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