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Study of the effect of combined reinforcement and modification of epoxy resin with rubbers on the impact strength of carbon fiber-reinforced plastic

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The effect of modified epoxy resin with polyurethane, silicone, and butadiene rubber and combined reinforcement with fiberglass and Kevlar on the strength properties of carbon fiber-reinforced plastic was studied. It was found that when rubber is introduced into the composition of the epoxy resin, polyurethane, and combined reinforcement according to the carbon fiber/Kevlar scheme in a ratio of 10:10 leads to an increase in impact strength by 42% to 273 kJ/m^2 and compressive strength increases by 35% to 575 MPa.

Keywords: elastomers; rubber; carbon fiber-reinforced plastic; strength; impact strength

Introduction

The creation of composite materials – carbon fiber with the necessary set of properties depends on a large number of physical and mechanical factors. The main role belongs to the polymer matrix as a basis for reinforced plastics. Namely, for carbon fiber-reinforced plastic (CFRP), epoxy resins are used, which, when cured, form rigid mesh structures. Today, the modern industry requires a variety of epoxy binders with a wide range of properties [1–3]. However, polymers based on unmodified epoxy oligomers often have low and unstable performance and physical and mechanical characteristics. The issue is solved by various methods of modifying epoxy resin and carbon fiber (components) of carbon fiber. The increase in the strength of carbon fiber is carried out by modification methods by introducing special additives into the resin: rubbers, plasticizers, carbon nanotubes, and combined reinforcement of carbon fibers (hybrid) [4–7]. Rubbers are most often used as modifying additives. The use of rubbers makes it possible to change the structure of the epoxy resin, which largely determines the strength characteristics of carbon fiber. Among rubbers, polyurethane, silicone, and butadiene rubbers are of particular interest [2, 8–11].

One of the main ways to increase the strength and toughness of carbon fiber is to combine reinforcement with fibers that have good elasticity and are most resistant to impact. As is known, composite materials made of carbon fibers have several advantages in terms of strength indicators for tension, compression, and elastic modulus, compared with other polymer composites reinforced with glass, aramid, para-aramid (Armos, Rusar, Twaron, Kevlar), polyester, basalt, boron, polyethylene fibers. However, in some cases, the use of composites reinforced only with carbon fiber is not rational due to such disadvantages of carbon fiber as relatively low shear strength and impact strength. Therefore, the most promising are polymer composites that combine reinforcing fibers of different natures [12].

Composites containing two or more different reinforcing materials bonded in the same matrix are commonly known as hybrid composites. In most cases, the goal of hybridization is to produce new material while retaining the advantages of its constituents and overcoming some of its disadvantages. Depending on the requirement for a hybrid composite, many combinations of reinforcement are possible. One of them is the hybridization of carbon fiber with fiberglass and/or aramid fabric.

The combination of glass and carbon fiber in a polymer composite can provide optimal mechanical properties [12-15]. Rajpurohit et. al. [16] studied the fracture behavior of hybrid composites. Two-fiber hybrids have been studied. When combined with a fiberglass laminate with carbon fiber, the composites were found to be effective in improving the toughness of the material. As the percentage of fiberglass increased, the maximum load capacity and impact energy absorbed by the material increased. The results on the study of mechanical properties of hybrid plastic, which is a composite material and contains reinforcement with both carbon fibers and fiberglass are reported in [17, 18]. The study found that the compressive strength of hybrid composites exceeds that of pure carbon fiber reinforced plastic by 8%. In [19] the effect of carbon fiber and carbon-aramid fiber reinforcement on the properties of epoxy resin has been studied. The resulting samples were tested for compressive strength/tensile strength, shear, and impact strength. The mechanical tests carried out showed that the presence of aramid fiber in the hybrid carbon-aramid composite resulted in a significant improvement in toughness of up to 37.9% compared to the conventional carbon composite. In addition, aramid fiber had a positive effect on the compressive strength with an increase of up to 19.5%, respectively, the presence of aramid fiber has a more ductile response compared to carbon reinforcement [20].

On the other hand, an increase in the mechanical properties of carbon fiber can be achieved by modifying epoxy resin (ER). In [21–25], the strength of ER and modified carbon plastics has been changed using tricresyl phosphate, rubber, and other additives. The rubbers have been added to the glass-like matrix, typically in an amount of 5–20% of the total mass of the epoxy oligomer, as reported in [26]. In [27] the influence of organosilicon rubber on the properties of epoxy resin has been studied. The physical and mechanical properties of the binders depending on the amount of rubber of 5%, 10%, 15% have been studied. It has been shown that at introduction of 10% SKTN-A silicon rubber in the composition increased the adhesion strength and impact viscosity on 50–60%. A rubber-modified epoxy composite with very high impact strength (21.1 kJ/m^2) and heat resistance (178 $^\circ$C) was developed in [23]. In [25] CFRPs made from ER modified with reactive liquid rubber and silicon dioxide nanoparticles were prepared and investigated. Modification of the matrix led to the production of strong composites with increased interlaminar fracture toughness until 1.299 kJ/m^2.

Thus, the use of two or more different reinforcing fibers and the modification of ER is a promising method for influencing the mechanical properties of CFRP. However, to achieve high levels of functional properties, it is necessary to understand how the modification of the ER, along with the combination of reinforcement, affects the structure and mechanical properties. The purpose of the work is to study the effect of combined reinforcement and modification of ER with polyurethane, silicone, and butadiene rubbers on the mechanical properties of CFRP.

Materials and Methods

Epoxy resin Etal Inject-T, three types of fabric, and three types of rubber were used as the object of study. Their characteristics are detailed below.

Hot curing Etal-Inject-T epoxy compound is used for the manufacture of heat-resistant glass and carbon fiber products. Curing temperature 4 hours at 150 $^\circ$C + 1 hour at 180 $^\circ$C. Carbon fiber Twill 3K-1200-200 (3K is the number of roving threads, K=1000; 1200 is the width of the roll in mm; 200 is the surface density in g/m^2) is an organic material that is characterized by high tension, chemical inertness, and low specific gravity. Aramid (Kevlar) plain for ballistic protection 460 g/m^2 is a heat-resistant durable synthetic fiber. Fiberglass Ortex 360–300 g/m^2 provides fabric compatibility with polyester, vinyl ester, epoxy, and phenolic resins. Silagerm 5045 polyurethane rubber is a two-component polyurethane-based compound for the manufacture of products with better physical properties and characteristics. Silicone rubber SKTN-A is a low molecular weight silicone rubber based on OH polymer. Butadiene rubber SKD, which is a product of butadiene polymerization in solution.

For the manufacture of CFRP samples, the method of manual molding with mechanical pressing and the vacuum infusion method was used. The manual molding technology is economical, does not require much labor, and the material consumption is less than with other molding methods. The essence of the method is that carbon fiber or other reinforcing materials are stacked in layers in a glass mold, where each layer is impregnated with resin. Vacuum infusion differs from manual molding in that after laying dry fabrics in a vacuum tool, the tool must be carefully sealed and the binder must be fed into the tool. Vacuum infusion technology improves manufacturability, and product quality and reduces the presence of pores.

Modification of ER Etal-Inject-T with elastomers (rubbers): polyurethane rubber (PR), silicone rubber (SR), and butadiene rubber (BR) in an amount of 10 wt.%. Fiberglass and Kevlar were used to reinforce the carbon fiber. Accordingly, two compositions were prepared for the experiments: carbon fiber/fiberglass and carbon fiber/Kevlar. The stacking of layers in each composition was carried out in a ratio of 1:1 and 2:1 and consisted of 20 layers by the scheme in Figure 1. The so-obtained samples were designated as follows: C-N1, f-N2, where N1 and N2 are number of layers, C – carbon fiber, f – fabric type (A for aramid and F for fiberglass).

To determine the compressive strength of CFRP, specimens were tested on an Instron Universal Testing Machine. Samples for compression tests made of CFRP are made by GOST 33519–2015 with dimensions: length l =80 $\pm$ 2 mm; width b =10 $\pm$ 0.5 mm, thickness h =4 $\pm$ 0.2 mm. Composite materials like CFRP often have anisotropic properties, meaning that their mechanical properties can differ depending on direction. During our research, we positioned the specimens in the testing machine to apply force along the composite's layup plane, which is considered the weaker direction. This ensures that the material will have the correct impact under the actual service conditions. Testing along the paving plane in the paving direction can provide information on the strength of the material that is most relevant to its actual use. The impact strength of CFRP is determined by the Charpy method according to GOST 4647–2015. This International Standard applies to plastics and specifies a method for the determination of impact strength on specimens with and without a notch.

Results and discussion

The results of testing the strength and impact strength of CFRP reinforced plastics obtained by manual molding are shown in Figure 2 and Figure 3. As can be seen, all modifications of the ER with combined reinforcement lead to an increase in performance compared to the original sample. One of the reasons for the hardening of CFRP is the wetting effect due to rubber, which leads to the formation of microcracks instead of ordinary cracks, as well as the creation of multiple overstress centers, leading to the appearance of a large number of microcracks [28].

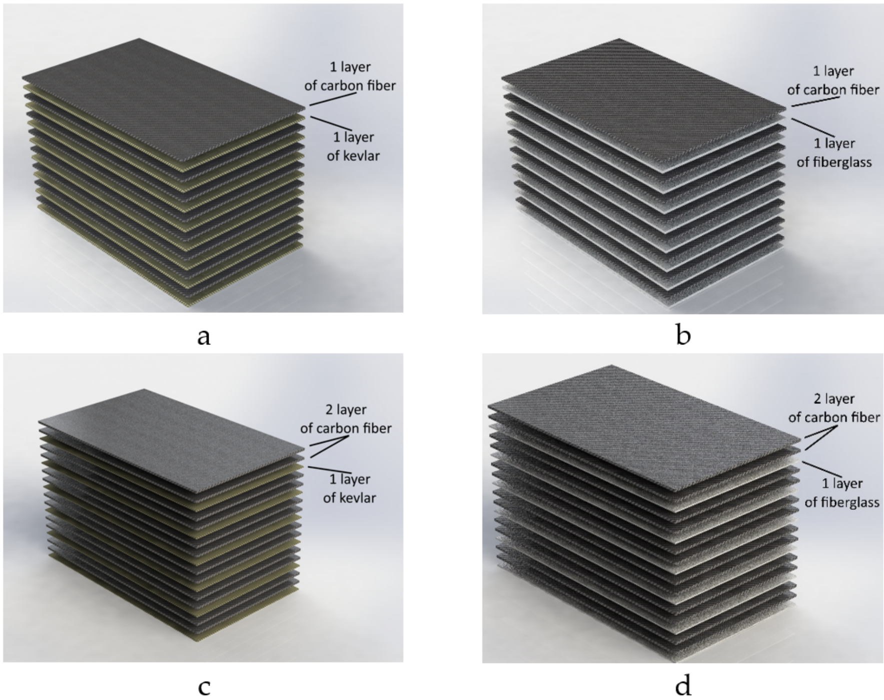

Figure 1. Scheme of stacking fabrics at different ratios of layers: *a* – the ratio of carbon fiber/Kevlar is 10:10; *b* – the ratio of carbon fiber/fiberglass is 10:10; *c* – the ratio of carbon fiber/Kevlar is 14:6; *d* – the ratio of carbon fiber/fiberglass is 14:6.

When using PR as an ER modifier and Kevlar as a reinforcement, it was revealed that the best impact strength result was obtained when reinforcing carbon fiber with 10 layers of Kevlar in a ratio of 10:10. The impact strength of the CFRP increased by 34% (258 kJ/m^2) and the compressive strength increased by 30% (552 MPa) compared to the unmodified sample.

When modified with SR in the amount of 10 wt.% and reinforcing material – Kevlar, the best result in terms of impact strength was obtained when reinforcing carbon fiber with 10 layers of Kevlar in a ratio of 10:10. The impact strength of CFRP increased by 21% (232 kJ/m^2) and compressive strength by 27% (541 MPa) compared to the unmodified sample.

According to the results of strength tests using the manual molding method for the BR modifier in an amount of 10 wt.% of the ER and Kevlar hybrid reinforcement, the compressive strength increased by 24%, and the impact strength by 15% compared to unmodified carbon fiber.

Thus, among the carbon fiber reinforced plastics produced by manual molding, the best indicators of mechanical characteristics were CFRP, modified with PR (C-10,A-10).

For high-quality adhesion of the matrix, minimizing the appearance of air pores, and improving the strength characteristics of CFRP in this series of experiments, the vacuum infusion method was used. The results of the influence of combined reinforcement and modification of ER with rubbers on the impact strength and strength of CFRP during vacuum molding are shown in Figure 4 and Figure 5. Changing the molding method from manual to vacuum made it possible to improve the strength and impact strength of the samples.

Modification of ER with rubbers PR, SR, and BR at best led to an increase in

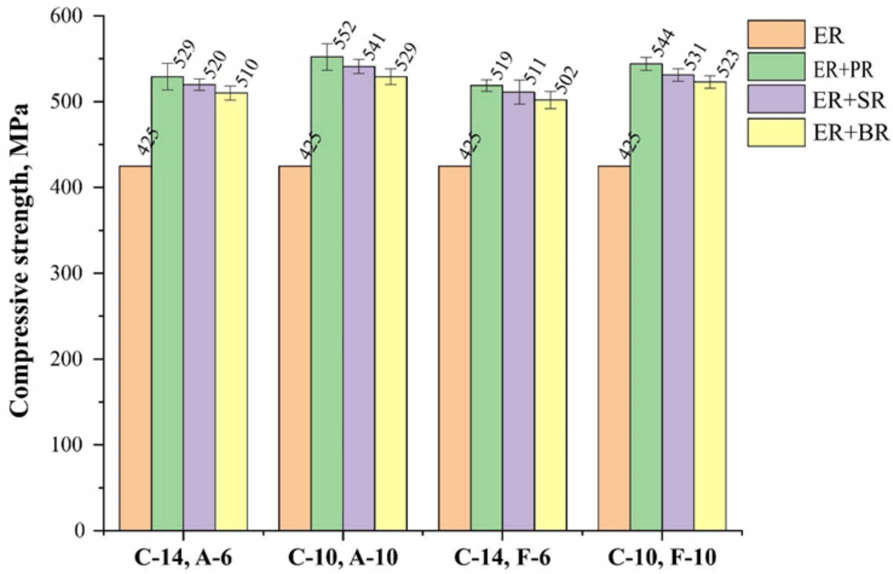

Figure 2. The effect of combined reinforcement and modification of ER with rubbers during manual molding on the strength of CFRP.

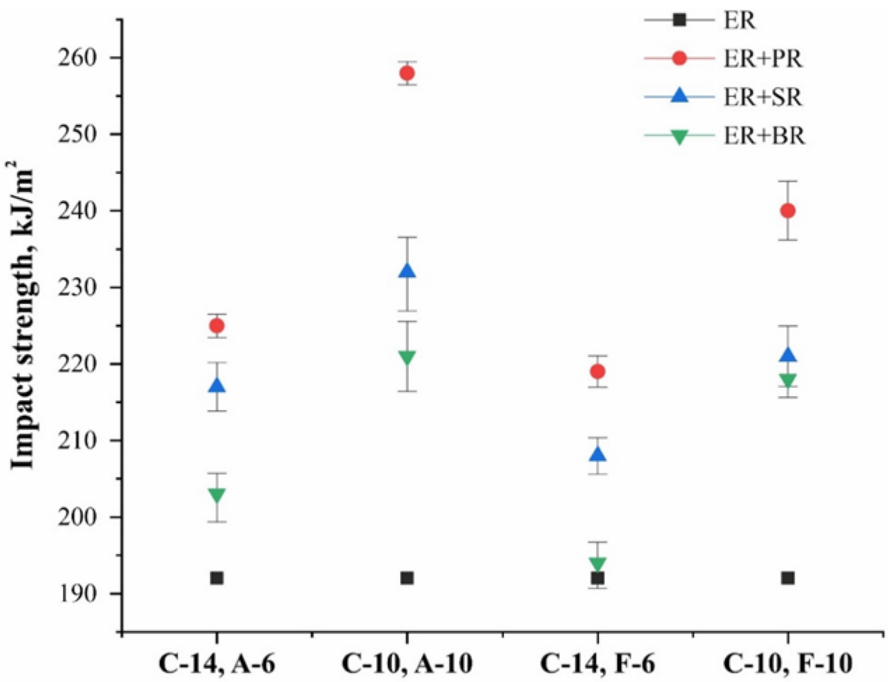

Figure 3. The effect of combined reinforcement and modification of ER with rubbers during manual molding on the impact strength of CFRP.

strength to 575, 562, 549 MPa. In the case of vacuum forming, as well as in hand forming, the 10:10 carbon/kevlar combination proved to be the most preferred in terms of strength and impact strength. The impact strength of unmodified carbon fiber was 192 kJ/m^2. The viscosity of CFRP significantly increases with the introduction of three types of rubber up to a maximum value of 273 kJ/m^2. A possible reason for the increase in CFRP is phase dispersion, in which rubbers can form as microscopic particles in the CFRP matrix, resulting in a more uniform load distribution. This can contribute to a more efficient redistribution of stresses, which ultimately increases the strength of CFRP. Also, rubbers can improve the adhesion between carbon fiber and Kevlar, which increases the adhesion between the layers of the material and prevents the formation of cracks [29].

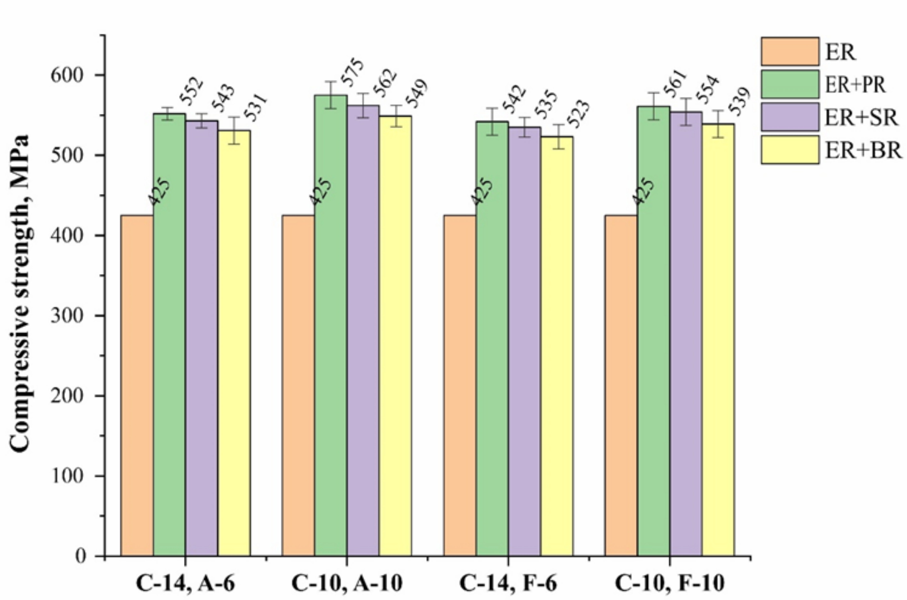

Figure 4. The effect of combined reinforcement and modification of ER with rubbers during vacuum infusion on the strength of CFRP.

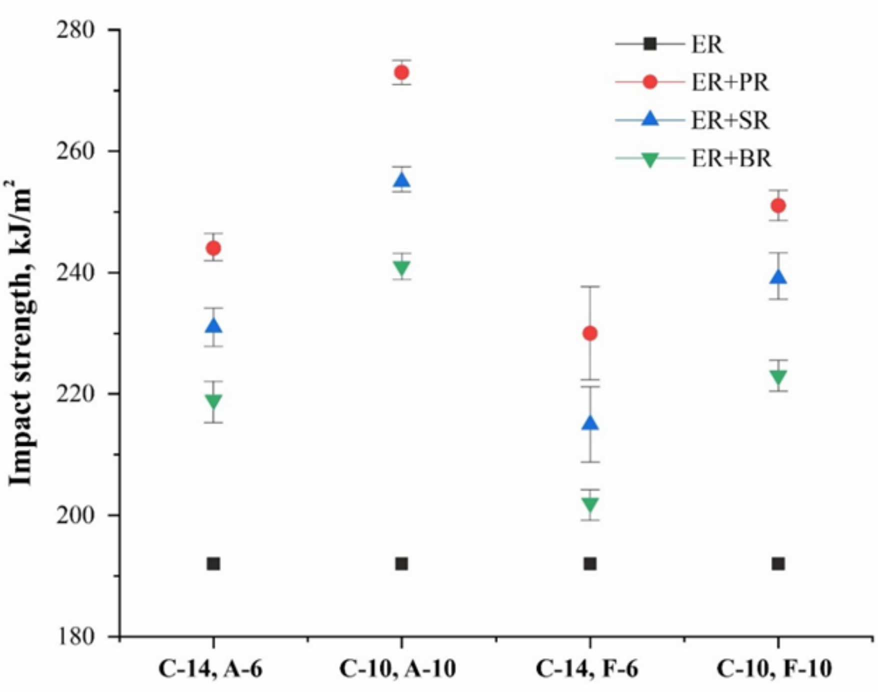

Figure 5. The effect of combined reinforcement and modification of ER with rubbers during vacuum infusion on the impact strength of CFRP.

According to the research results, it was revealed that the introduction of polyurethane rubber into the composition of the ER and combined reinforcement according to the scheme carbon fiber/Kevlar 10:10 leads to an increase in impact strength. Based on the conducted studies, it was revealed that polyurethane rubber and combined reinforcement in the carbon fiber/Kevlar scheme (10:10) are the most promising as an oligomer modifying the epoxy component. Therefore, for greater clarity, the following Figure 6 shows a comparison of the best designs C-10, A-10 for two methods of composite formation.

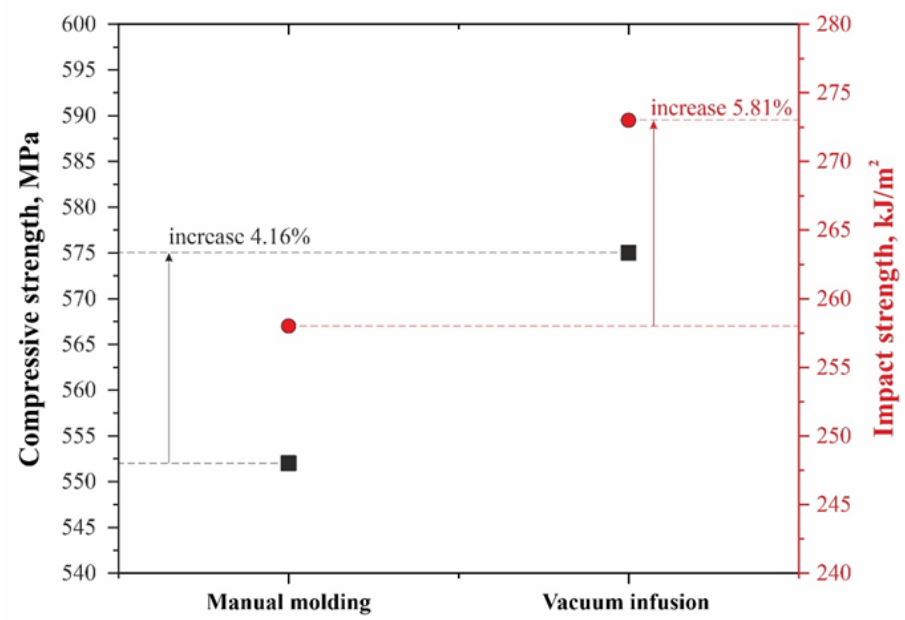

Figure 6. Characteristics of the composite obtained by vacuum infusion and manual molding methods.

Conclusion

In the course of this study, various methods for obtaining CFRP were studied. To create CFRP samples, manual molding methods with mechanical pressing and vacuum infusion were used. Modification of the Etal-Inject-T epoxy resin was carried out by introducing rubbers, such as polyurethane, silicone, and butadiene rubber, in amount of 10 wt.%. For the combined reinforcement of carbon fiber, Kevlar and fiberglass were used. As a result of hand molding studies using polyurethane rubber as an epoxy binder modifier, it was found that the optimal impact strength characteristics were achieved when carbon fiber was reinforced with 10 layers of Kevlar in a ratio of 10:10. The impact strength of CFRP increased by 34% (258 kJ/m^2), and the compressive strength increased by 30% (552 MPa) compared to the unmodified sample The best performance was achieved using the carbon fiber/Kevlar scheme with polyurethane rubber modified epoxy resin in vacuum infusion. This method resulted in a significant 42% increase in impact toughness and 35% increase in compressive strength.

In conclusion, this study confirms that the modification of an epoxy binder using various rubber modifiers and combined reinforcement contributes to a significant improvement in the mechanical characteristics of CFRP, which may have important practical applications in the field of creating strong and lightweight composite materials.

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