ISSN 2522-9117 (print), ISSN 2786-6149 (online)
"Фундаментальні та прикладні проблеми чорної металургії". 2025. Випуск 39
"Fundamental and applied problems of ferrous metallurgy". 2025. Issue 39

https://doi.org/10.52150/2522-9117-2025-39-17

УДК 621.762.8:004.923

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INFLUENCE OF CARBON FILLER SURFACE MODIFICATION ON INTERFACIAL ADHESION IN THERMOPLASTIC CFRTP COMPOSITES (REVIEW)

Abstract. *Introduction.* Carbon-fiber-reinforced thermoplastic composites (CFRTP) occupy a special place among modern structural materials due to their unique combination of high specific strength, corrosion resistance, and recyclability. *Relevance.* Their use is becoming increasingly significant in the aviation, automotive, energy, and medical industries, where higher requirements are imposed on wear resistance, durability, and process controllability during manufacturing. However, one of the key scientific and technical challenges remains ensuring an adequate level of interfacial adhesion between carbon fiber (CF) and the polymer matrix, which determines the overall set of mechanical properties and the functional reliability of the final material. *Objective:* To analyze the literature and systematize data on modern methods of surface treatment of carbon fiber filler in CFRTP, which are carried out to improve the adhesion between the reinforcing component and the thermoplastic matrix. *Results.* The study outlines the problem of insufficient adhesion due to the difference in material polarity and analyzes modern approaches to CF surface modification: chemical (acid, silane, isocyanate, radiation treatment), physical (plasma, microwave, metal coatings, application of nanostructured coatings), as well as the use of compatibilizers and coupling agents (MAPP, PPEK, copolymers). It is shown that such methods significantly increase adhesion at the fiber–matrix interface, which contributes to the improvement of the mechanical properties of CFRTP composites and expands their potential for engineering applications. The interaction patterns of surface-modified fibers with matrices such as polyamide-6, polyetheretherketone (PEEK), and polypropylene are characterized separately, demonstrating different levels of wettability and thermomechanical compatibility. *Conclusion.* Specialized technological approaches based on physical or chemical methods make it possible to enhance adhesion between the matrix and the filler in CFRTP, which in turn improves the properties of the composite and broadens the scope of its industrial applications.

Keywords: thermoplastic composite, carbon fiber, interface, surface treatment, surface modification, adhesion.

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For citation: Vysnevetskyi, V. V. (2025). Influence of carbon filler surface modification on interfacial adhesion in thermoplastic CFRTP composites (Review). *Fundamental and applied problems of ferrous metallurgy*, 39, 285-295. https://doi.org/10.52150/2522-9117-2025-39-17

Introduction. The development of new durable materials with unique properties is extremely relevant, especially when it comes to materials that meet the needs of industry and contribute to reducing environmental pollution. New materials must also overcome existing challenges, such as high cost, recyclability, ensuring reliability, and reducing energy consumption. One of the priority tasks in creating new products is the simultaneous reduction of weight and increase of operational load [1–2]. A promising lightweight material is carbon fiber (CF), which is distinguished by high strength, thermal stability, and chemical inertness. It is non-toxic, has low density, high wear resistance, corrosion resistance, recyclability, and an optimal strength-to-weight ratio. CF also possesses unique thermal, mechanical, and electrical characteristics. The production of this fiber is carried out through the carbonization of base materials such as synthetic polymers (polyacrylonitrile, phenolic resin), involving stages of oxidation and heat treatment under load at high temperatures with control of the final fiber properties.

Raising the carbonization temperature (up to 2500 °C) makes it possible to obtain CF with an increased carbon content. Products made from polymer composites using CF are widely applied due to their high mechanical, thermal, electrical, and tribological properties. The fields of application include wind energy, aviation, automotive engineering, industrial and social infrastructure, the marine industry, construction, and sporting goods [2, 3]. CF is classified depending on the base material, processing temperature, and the length or orientation of the fiber. Such classification allows for the selection of different configurations for specific applications. The results of various studies have confirmed the influence of CF orientation on the properties of polymer composites [4, 5, 6].

In recent decades, there has been a growing interest in carbon fiber-reinforced polymer composites (CFRP), particularly in thermosetting polymers such as epoxy and polyester resins. However, thermosetting composites are generally not recyclable, which creates environmental and economic challenges in large-scale production.

Thermoplastics, in contrast to thermosetting polymers, are generally recyclable and can be easily adapted to specific requirements. They can be combined with unidirectional, discrete (short and long), or continuous CF to produce composites with tailored properties in one or multiple directions. Thermoplastics are divided into: general-purpose – polylactic acid (PLA), polyethylene (PE), polypropylene (PP), polystyrene (PS), acrylonitrile butadiene styrene (ABS); and high-performance or engineering plastics – polyamide (PA), polyethylene terephthalate (PET), polycarbonate (PC), polyether ether ketone (PEEK), polyetherimide (PEI), polyethersulfone (PES), polyphenylene sulfide (PPS).

Thanks to the simplicity of manufacturing processes, safe chemical compositions, higher recyclability, and the potential for mass production, thermoplastics offer significant advantages over thermosetting resins. At the same time, the final composite materials demonstrate properties that exceed the performance of the individual components. Carbon-fiber-reinforced thermoplastics (CFRTP) allow for a weight reduction of approximately 50% compared to steel and 20% compared to aluminum [7]. Thermoplastics can be combined with unidirectional CF, discrete fibers (short and long), or continuous fibers (CCF) to create composites with service and performance characteristics tailored to the required level in one or multiple directions.

CFRTP is manufactured by conventional forming methods such as injection molding, rotational molding, extrusion, vacuum forming, and compression molding. Although CFRTP has attracted the attention of numerous researchers due to its mechanical and thermal properties, recyclability, flexibility, short production time, and environmental safety, it is still under development for certain industries and requires overcoming the problem of high production costs. During manufacturing processes, multiple factors must be considered to ensure the high quality of the final products while maintaining production efficiency. Synthesized CF possesses a smooth and non-polar (hydrophobic) surface, whereas polymers are generally polar (hydrophilic). Therefore, surface pretreatment of CF is required for the reinforcement process.

The treatment involves the creation of functional groups on the CF surface in order to ensure proper interfacial adhesion between the polymer and CF, which is crucial for the development of high-performance composites; this plays a decisive role in their practical application. In studies [7–8], the importance of a strong bond between the reinforcing fiber and the matrix was emphasized for obtaining composites with superior technical properties (Fig. 1).

The manufactured CF must be wear-resistant, capable of withstanding loads without cracking, and able to operate reliably under various conditions, such as elevated temperatures and humidity.

Objective: analysis of literature sources and systematization of data on

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modern methods of surface treatment of carbon fiber filler in CFRTP, which are carried out to improve the adhesion of the reinforcing component with the thermoplastic matrix.

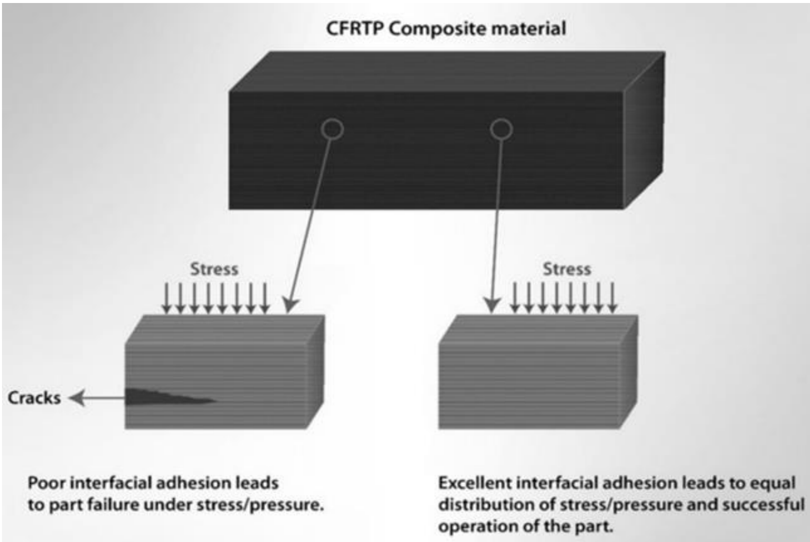

Figure 1 – Schematic diagram of poor and excellent interfacial adhesion between the polymer matrix and the reinforcing fiber [8].

Materials and Methods of Research. The presented work is based on a comprehensive bibliographic review of modern scientific and technical publications concerning the features and methods of fiber surface modification, which enhance adhesion to the matrix and ensure the improvement of the mechanical properties and durability of CFRTP.

Research results and discussion. Composites based on CF reinforced with thermoplastic polymers (CFRTP) attract considerable scientific and technical interest due to their favorable performance characteristics, which determine their potential for widespread implementation in various industrial sectors. These characteristics can be purposefully modified by optimizing the choice of materials and manufacturing techniques. Particular attention should be paid to parameters such as fiber length, their orientation in the polymer matrix, and the surface condition of CF (Fig. 2). Each decision made at the production stage has a direct impact on the final properties of the composite, which can either limit or expand the possibilities of using the material in specific industrial applications.

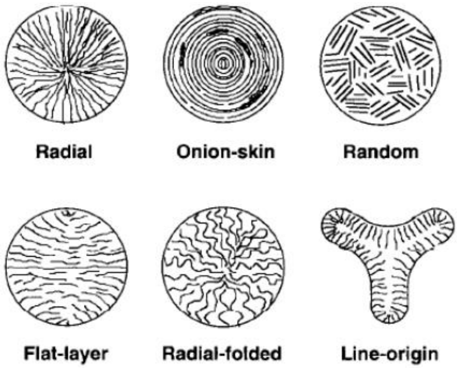

Figure 2 – Microstructure of carbon fibers [9].

The interfacial property is a key factor, since with strong adhesion the load is effectively transferred from the matrix to the CF without causing damage to the product. The bond between the reinforcing fibers and the thermoplastic matrix is often insufficient due to differences in their polarity: thermoplastics are predominantly polar, whereas CF is non-polar. To eliminate the negative effect of these features, various CF surface treatment methods are studied and applied (Fig. 3), including both chemical and physical techniques [3, 5, 7, 8, 10, 11]. The regulation of interfacial properties leads to a significant improvement in adhesion between CF and thermoplastic polymers. This is attributed to the control of surface interactions between the reinforcing fibers and the polymer matrix [12].

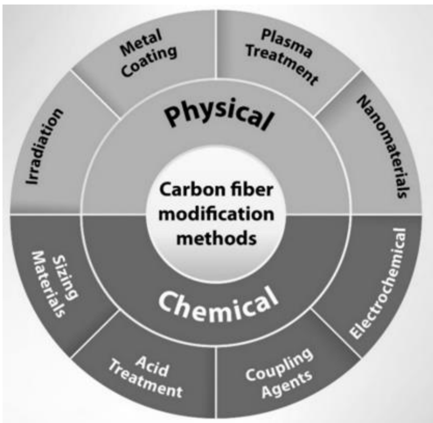

Figure 3 – Standard CF surface treatment methods [8].

Substances known as coupling agents and compatibilizers enhance the interfacial adhesion strength between the reinforcing filler and the polymer matrix in composites. In [10], it was investigated how three types of maleic anhydride-grafted polypropylene (MAPP), differing in chain length and anhydride group concentration, affect the interfacial adhesion properties in composites reinforced with recycled carbon fiber (RCF). It was found that the degree of compatibility is largely determined by the molecular weight and the amount of anhydride groups in the composition of the coupling agent.

A study [9] was conducted in which a silane coupling agent was applied to polypropylene-based composites with CF, and it was found that surface treatment of the fiber with this agent significantly affects the interaction between the fiber and the matrix, as well as the overall properties of the material. Similar studies were carried out in other works [13], where the influence of surface characteristics of short CF and the concentration of modified homopolypropylene on the adhesion between reinforcing fibers and the matrix was analyzed. In the first experiment of the study, a bifunctional group was used to modify long CF, which resulted in improved mechanical strength of polypropylene-based composites reinforced with long CF. In the second, an aminated polyphenylene sulfide composition was used as a compatibilizer in the composites, showing that amination enhances compatibility between the fiber and the matrix and strengthens adhesion at their interface. Additionally, three different polyethylene copolymers were applied as compatibilizers in high-density polyethylene composites reinforced with CF. As a result, it was found that interfacial adhesion depends on the type of copolymer, although the addition of all tested compatibilizers improved the properties compared to the case when they were absent.

Several studies have investigated the influence of different sizing materials (polyurethane, polyamide, polyimide, phenoxy) on the properties of polyamide-based composites reinforced with CF [14–16]. The characteristics of composite materials largely depend on the choice of substances used for CF treatment. In [15], an isocyanate-modified epoxy emulsion and a silane coupling agent were applied for the preparation of CF in PA6-based composites. The use of coupling agents and compatibilizers significantly improves the quality of bonding between the fibers and the matrix, although in certain cases, such as with UHMWPE composites (Ultra-High Molecular Weight Polyethylene), such improvement is not observed.

In the study [17], composites based on PPEK and CF were synthesized, using PPEK as a sizing material for CF at three different concentrations (0.1, 0.5, and 1 wt.%). It was established that sized CF exhibited better compatibility with PPEK compared to untreated fibers, which was confirmed by contact angle measurements.

Acid treatments have also been applied to improve the interfacial properties of CFRTP composites. In the study [18], chemical interaction between HDPE and CF was reported without the use of an additional coupling agent. It was found that a significant increase in fracture strain, flexural modulus, and flexural strength was observed with an increasing number of CF layers in the composites. The reason for the better compatibility of nitric acid–treated fiber with the HDPE matrix was that the hydrophilic carboxyl group of benzoic acid reacted with the hydroxyl groups of the treated fiber. This contributed to improved interaction with the polyethylene matrix, which in turn enhanced the interfacial interaction between UHMWPE and CF after acid treatment. In the study [19], CF was treated with nitric acid and then incorporated into polyoxymethylene composites. After such treatment, an increase in the number of active functional groups, higher surface roughness, and a greater fraction of amorphous carbon were observed on the fiber surface.

Studies [11, 20] conducted by different research groups demonstrate the effectiveness of plasma treatment in improving the properties of CF-based composites. These investigations revealed that plasma surface modification of CF increases the number of functional groups, which positively influences the interfacial adhesion in PC/CF composites. Oxygen and helium treatments under atmospheric pressure enhance the interfacial properties of PA6/CF composites by increasing oxygen concentration, roughness, and surface energy of CF. Microwave plasma treatment of CF improves the mechanical bonding with the cyclic butylene terephthalate (CBT) matrix in CFRTP composites. Research on the treatment of RCF in dry air and CO_2 highlighted the importance of gas type and treatment duration in achieving optimal adhesion.

The study [21] established that radiation exposure positively influences the adhesion between CF and high-density polyethylene (HDPE), while also altering the surface properties of HDPE/CF composites. This improvement is achieved through two key effects of irradiation: first, the cross-linking of the thermoplastic resin, which enhances load transfer to CF, and second, the formation of surface functional groups that facilitate fiber–matrix interfacial interactions.

The application of surface coatings is also employed to enhance adhesion between different phases. For instance, nickel plating of CF has been shown to improve their properties and interfacial adhesion, as reported in study [22]. However, this technique may lead to corrosion due to the electrical conductivity and chemical characteristics of the CF surface. In contrast, in study [6], a nickel coating was applied to investigate the orientation of CF in thermoplastic composites.

To strengthen the bonding between CF and thermoplastic matrices, surface modification of CF through nanoparticle grafting is applied. As reported in study [23], a significant improvement in the interfacial properties of PES/SCF composites was observed after CF treatment with graphene oxide. The deposition of carbon nanotubes and graphene oxide onto CF was investigated. It was demonstrated that this treatment enhanced adhesion, surface roughness, and wettability. These modifications facilitate the formation of stronger bonding with the PI matrix through the creation of hydrogen bonds and mechanical interlocking.

Conclusions

The progress in the development of carbon fiber-reinforced thermoplastic polymers (CFRTP) has been examined with the aim of improving service and performance properties. The high efficiency of CFRTP holds strong potential for future research, as the integration of CF as a reinforcing element provides significant improvements in composite characteristics compared to pure thermoplastic matrices, thereby opening new prospects for the application of CFRTP in various industrial fields.

The quality of composites is shaped by a variety of interrelated factors. These include the manufacturing method, the adjustment of technological parameters, the selection of the thermoplastic matrix, the type and spatial orientation of CF, their concentration, size, and surface treatment methods. These elements directly influence the bonding strength between fibers and the polymer, as well as how well the fibers are distributed within the matrix. As a result, these interactions determine the mechanical properties and operational performance of the final material.

To achieve better adhesion between CF and the thermoplastic matrix, surface modification of CF is required. The application of chemical or physical modification methods strengthens the bond between the filler and the matrix, which has a positive effect on the mechanical properties of the composite compared to composites with untreated fibers.

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ВПЛИВ МОДИФІКАЦІЇ ПОВЕРХНІ ВУГЛЕЦЕВОГО НАПОВНЮВАЧА НА МІЖФАЗНУ АДГЕЗІЮ В ТЕРМОПЛАСТИЧНИХ КОМПОЗИТАХ CFRTP. (ОГЛЯД)

Анотація. *Вступ.* Вуглецево-волоконні композити з термопластичними матрицями (CFRTP) посідають особливе місце серед сучасних конструкційних матеріалів завдяки унікальному поєднанню високої питомої міцності, корозійної стійкості та можливості вторинної переробки. *Актуальність.* Їх використання набуває актуальності в авіаційній, автомобільній, енергетичній та медичній галузях, де висуваються підвищені вимоги до зносостійкості, довговічності та технологічної керованості процесів виготовлення. Проте однією з ключових науково-технічних проблем залишається забезпечення належного рівня міжфазної адгезії між вуглецевим волокном (CF) і полімерною матрицею, що визначає комплекс механічних характеристик та функціональну надійність готового матеріалу. *Мета:* аналіз літературних джерел та систематизація даних щодо сучасних методів обробки поверхні вуглецевого волокна-наповнювача CFRTP, яку проводять для покращення адгезії армуючого компонента з матричним термопластиком. *Результати.* У роботі окреслено проблему недостатньої адгезії через різницю у полярності матеріалів та проаналізовано сучасні підходи до модифікації поверхні CF: хімічні (кислотна, силанова, ізоціанатна, радіаційна обробка), фізичні (плазмова, мікрохвильова, покриття металами, застосування наноструктурних покриттів), а також використання сумісників та агентів зв'язування (MAPP, PPEK, кополімери). Показано, що такі методи суттєво підвищують адгезію на межі волокно–матриця, що сприяє покращенню механічних властивостей CFRTP-композитів і розширює їхні можливості для інженерних застосувань. Окремо охарактеризовано закономірності взаємодії поверхнево модифікованих волокон із такими матрицями, як поліамід-6, поліефірефіркетон (PEEK) та поліпропілен, які демонструють різний рівень змочуваності та термомеханічної сумісності. *Висновок.* Спеціалізовані технологічні підходи на основі фізичних або хімічних методів дозволяють підвищити адгезію між матрицею та наповнювачем в CFRTP, що дозволить покращити властивості композиту та розширить можливості його промислового застосування.

Ключові слова: термопластичний композит, вуглецеве волокно, межа, поверхнева обробка, модифікація поверхні, адгезія.

Посилання для цитування: Вишневецький В. В. Вплив модифікації поверхні вуглецевого наповнювача на міжфазну адгезію в термопластичних композитах CFRTP. (Огляд). *Фундаментальні та прикладні проблеми чорної металургії.* 2025. Вип. 39. С. 285-295. https://doi.org/10.52150/2522-9117-2025-39-17

Рукопис надійшов до редакції / Received 06.08.2025
Рекомендовано до друку / Accepted 21.10.2025