fibers

Review

Carbon Fiber-Reinforced Geopolymer Composites: A Review

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Abstract: The article summarizes the state of the art in carbon-reinforced geopolymers. It takes into consideration various types of matrices and types of carbon fibers (CFs). The article shows the growing importance of this composite in the investigation conducted in recent years. Today, it is one of the most promising modern research areas, taking into account the decrease in the prices of CFs and their appearance on the market waste-based CFs, as well as research on new methods of producing CFs from sustainable precursors. The research methods applied in the article are critical analyses of the literature. The results of the literature analysis are discussed in a comparative context, including production methods and the influence of CFs on geopolymer properties. The potential applications for carbon fiber-reinforced geopolymer composites are shown. Additionally, the current research challenges for geopolymer composites reinforced by CFs are presented.

Keywords: carbon fiber; geopolymer; geopolymer composite

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1. Introduction

Different types of chemical fibers, such as steel fibers, fiberglass, carbon fibers (CFs), and others, are currently the most commonly applied additives to various types of composites, including concrete-like materials dedicated to the construction industry [1–3]. The addition of these fibers is primarily aimed at enhancing mechanical properties, in particular bending strength, and additionally decreasing the propagation of microcracks in the material [4,5]. Other anticipated benefits, depending on the type of fiber used, are increased fire resistance, reduction in thermal conductivity, improved capacity for electricity conduction, or other desirable features for a particular application [6,7]. It should be noted that chemical fibers usually have better repeatability than natural fibers thanks to their higher strength properties [5]. However, its use does not bring so many benefits to the environment in comparison to natural fibers [7,8]. Above all, as a rule, their resources of traditional feedstocks for the production of synthetic fibers, such as crude oil, are limited, although modern technologies allow for the development and obtaining of these kinds of materials from renewable sources. Contemporary biobased polymers can be obtained, among others, from starch or biobased succinic acid [9,10]. Similarly, these kinds of new technologies are also developed for CFs [11,12].

The growing use of CFs is linked to their numerous advantages. They have a low density, high tensile strength, great Young's modulus, abrasion resistance, low friction, high fatigue and creep strength, great chemical resistance, good electrical conductivity, good dimensional stability, vibration damping, non-melting properties, and low absorption of X-rays [13,14]. The basic physical and mechanical properties of CFs are strongly connected with the form of fiber. A comparison between different forms of CFs is presented in Table 1 [13–17].

Table 1. Basic properties of carbon fibers [14–17].

Type of Fiber	Density [g/cm^3]	Young's Modulus [GPa]	Tensile Strength [MPa]	Elongation [%]
PAN-based carbon fiber	1.6–2.0	230–500	2500–7000	0.6–2.5
Pitch-based carbon fiber	ca. 1.5	30–935	500–3800	0.3–1.5
Carbon nanotube	1.2–1.3	1000–1800	11,000–63,000	5.7–7

The main barrier to the industrial applications of these fibers is their high price. However, it should be noted that new technologies are expected to enter the market and the related decline in their prices [14]. Other disadvantages of CFs include the predisposition to oxidize in an oxygen environment under high temperature (in non-oxidizing atmospheres; at temperatures up to 2000 °C, CFs do not lose their properties). However, it should also be stressed that the oxidation of a CF is catalyzed by an alkaline environment [13]. This feature makes the geopolymer matrix, having an alkaline pH and a favorable environment for the manufacture of composites, which can also work at high temperatures.

CFs were used for the first time as filaments for electric light bulbs. They were founded by Thomas Edison in early 1880. Today, carbon fibers are used as reinforcements in the production of modern composite materials based on polymers, metals, and ceramics [13,18]. They are used, inter alia, in the aviation and space industry, construction (including areas threatened by earthquakes, where they are used as reinforcements of light structures), the energy industry (including blades of wind power plants), the medical industry (reinforcing polymer orthopedic splints), and as elements of sports equipment, including yachts [13,18,19].

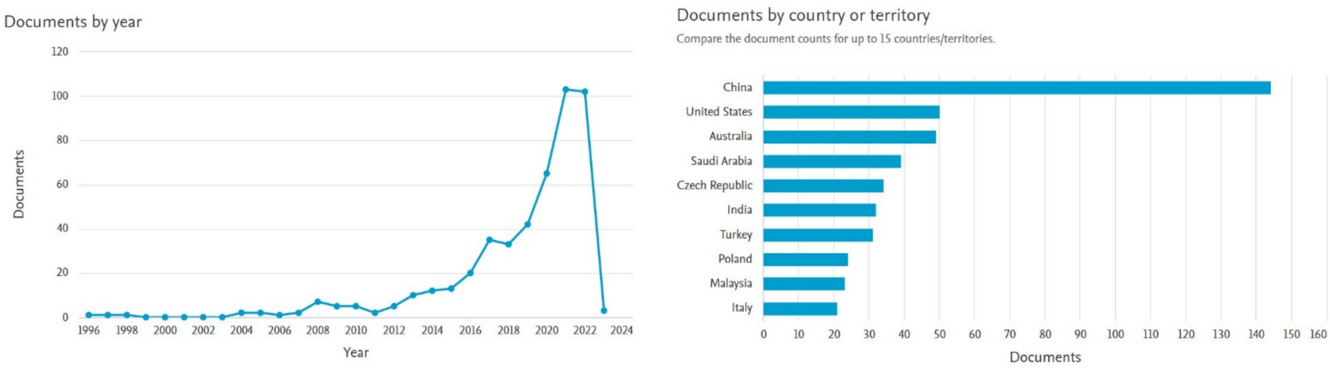
A large amount of research has been carried out with the addition of CFs to geopolymers because of the promising properties of the obtained composites. CFs are produced mainly by the pyrolysis of polyacrylonitrile (it is estimated that 90% of carbon fibers in the world are produced on the basis of PAN), and their properties are primarily influenced by the parameters of the manufacturing process used [16,17]. It consists almost entirely of stretched carbon structures, chemically similar to graphite [20]. In addition to the classification of CFs according to their origin, they can be also divided into several groups because of their mechanical characteristics. The main background for this classification is modulus of elasticity and tensile strength [16,17]. According to the modulus of elasticity, we can classify the CF into four groups: ultra-high modulus (>500 GPa), high modulus (>300 GPa), intermediate modulus (<300 GPa), and low modulus (<100 GPa). This classification does not include carbon nanotubes, because for the nanotubes these values are much higher. The second classification is tensile strength. For this value as a high result, consider 3000 MPa for traditional CF and a minimum of 7000 MPa for nanotubes [17]. It is worth paying attention to the fact that the origin, including the method of manufacturing and mechanical properties, is quite often connected [17,21].

It is worth noticing that a lot of contemporary research is dedicated to the development of new methods of producing CFs using sustainable feedstocks, especially the lignin precursor [11,12,20]. The estimations show that lignin-based CFs can cut in half the CO_2 footprint of CF production [22,23]. Among the natural sources that have been studied as a precursor for CFs, lignin and lignin blends seem to be the most promising [20,23]. The obtained CFs show in this case much better properties than those obtained from other natural sources, such as pitch and cellulose (Viscose Rayon); however, they are still lower than for CFs obtained from fossil fuels [23,24]. One of the problems that must be solved is the proper homogenization of the CF and the elimination of impurities, such as sulfur [24,25].

The article reviews up-to-date literature in the area of carbon-reinforced geopolymers, taking into consideration new possibilities for market development given by reducing the prices of CFs, entrance into the market, waste-based CFs, and developing new methods of CF manufacturing from sustainable precursors. The opportunities, as well as challenges for geopolymer composites reinforced by CFs, are discussed, including the most promising opportunities for the application of this material for advanced products in different industries.

2. Research Methodology

The study was carried out using scientific article databases such as ScienceDirect, Scopus, and Google Scholar. The starting point was research in the Scopus database using the term "geopolymer composite" and "carbon fiber" combined together. The research results show 472 documents (Figure 1a).



(**a**) (**b**)

Figure 1. Results of the analysis in the Scopus database. (**a**) Published documents by year and (**b**) published documents by country [26].

The first papers were published in this area in 1996. The rapidly growing research in this area started in 2016 and especially in the last 3 years and is characterized by a lot of new research and publications (Figure 1a) [26]. It is connected with factors such as:

- Increasing the number of investigations on geopolymers as a material suitable for suitable development and circular economy; including material with CFs that came from recycling.
- The decreasing price of CFs and appearance on the market of waste CFs and the research on new methods for producing CFs from sustainable precursors.
- Increasing the number of investigations on geopolymers as materials for advanced applications, including research about conductivity.

The authors in the review are focused on the newest publication, especially the last 3 years; however, some oldest publications that are important on the topic have also been analyzed. The leading country that provided research in this area is China, but research is also provided in America, Asia, and Europe (Figure 1b) [26]. It shows that this topic is important for many countries. The publications were also analyzed according to type. It shows that 8.3% of the publications have a review character. Among them, there has been a lack of publications dedicated only to carbon fiber-reinforced geopolymer composites in recent years. In most of the reviews, this kind of composite is only a small part of the overall investigation.

3. Types of Carbon Fibers Used in the Geopolymer Matrix

CFs can be applied in different forms of occurrence in the geopolymer matrix. The most popular seem to be short CFs because they are easy to use for different applications. Most manufacturing methods do not require significant changes to implement the short fibers, despite the application of long fibers or textiles [14,27]. However, several research works involved short fibers; a significant part of them concerns work made with the addition of long fiber [28,29] fabrics as composite elements [30,31], carbon fiber felt, or similar forms [32,33], as well as the addition of other forms of carbon, i.e., graphene [34,35], graphite [36], or waste CF used in the manufacture of aviation products [37]. The last three additives were introduced most frequently into geopolymers in the form of particles.

3.1. Short and Long Carbon Fibers

Theoretically, the difference between short and long CFs can be determined by a mathematical calculation based on fiber tensile strength, fiber diameter, and interfacial shear strength. According to this model, the proper length of the fiber will improve flexural strength and fracture properties. It implies significant strengthening and toughening effects [38]. In practice, many authors prefer the experimental approach due to some problems with finding appropriate input data for the theoretical model, especially due to the changeable properties of the matrix used, which are typical for cementitious materials [15,38].

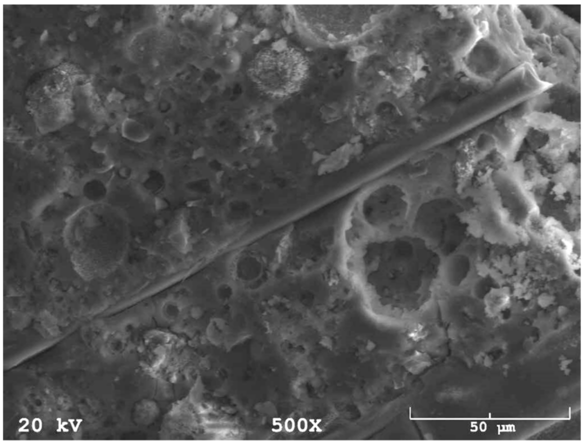
The typical fibers used have a form of chopped short fibers with a length of 3 to 60 mm and a diameter from a few µm or long fibers, such as roving applied throughout the entire length of the samples or the investigated element (Figure 2) [15,39,40].

(**a**) (**b**)

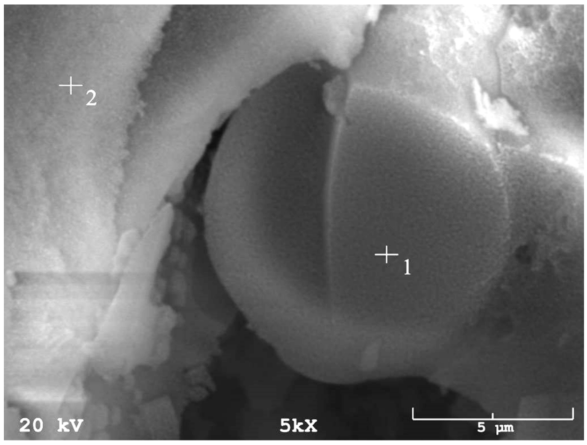
Figure 2. Different forms of carbon fibers. (**a**) Short carbon fibers [39] and (**b**) long carbon fibers (roving) [40].

The amount of short CF in geopolymer composites is usually 1–2 wt.% of the geopolymer mass, or even less, up to 1.0 vol.% [6,15,39]. This amount effectively increases the mechanical properties of the composite, as well as its ductility [6,15]. The addition of a larger amount of CF usually causes problems with the processing properties of the composites, especially in the case of short fibers [39]. In the case of long fibers, it is possible to increase the used amount of fibers; however, the investigations show that the optimal amount is between 0.1–2.0 wt.% and depends on the type of geopolymer matrix used [6,15,40].

The CF usually has good coherence with different types of geopolymer matrices. The microscope investigation shows a lack of decohesion between the CF and the geopolymer matrix (Figure 3a,b). Additionally, energy-dispersive X-ray spectroscopy (EDS) analysis confirms coherence between the CF and matrix material (Figure 3c,d). The EDS analysis carried out at point 1 concerning the fiber material covered with the geopolymer and the analysis carried out at point 2 concerning the geopolymer matrix material (Figure 3b) shows similar results that are probably the effect of covering the CF by a thin layer of matrix material [14].



(**a**)



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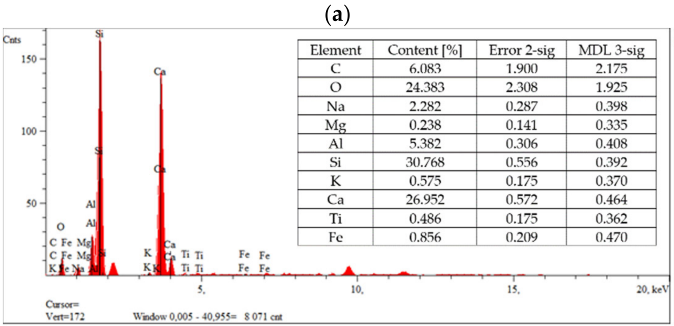
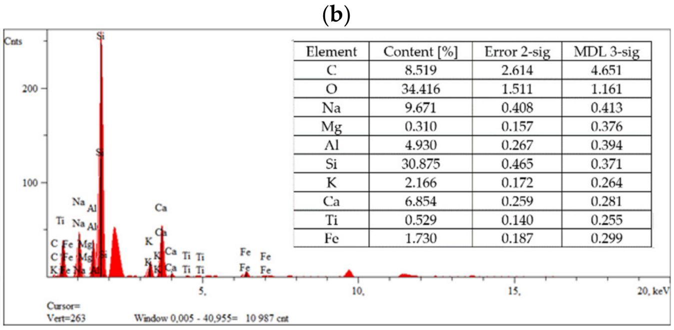


Element	Content [%]	Error 2-sig	MDL 3-sig
C	6.083	1.900	2.175
O	24.383	2.308	1.925
Na	2.282	0.287	0.398
Mg	0.238	0.141	0.335
Al	5.382	0.306	0.408
Si	30.768	0.556	0.392
K	0.575	0.175	0.370
Ca	26.952	0.572	0.464
Ti	0.486	0.175	0.362
Fe	0.856	0.209	0.470

(**c**)



Element	Content [%]	Error 2-sig	MDL 3-sig
C	8.519	2.614	4.651
O	34.416	1.511	1.161
Na	9.671	0.408	0.413
Mg	0.310	0.157	0.376
Al	4.930	0.267	0.394
Si	30.875	0.465	0.371
K	2.166	0.172	0.264
Ca	6.854	0.259	0.281
Ti	0.529	0.140	0.255
Fe	1.730	0.187	0.299

(**d**)

Figure 3. Scanning microscopy of short fibers in the geopolymer matrix. (**a**) Description of what is contained in the first panel; (**b**) description of what is contained in the second panel; (**c**) EDS analysis for point 1; (**d**) EDS analysis for point 2 [14].

It is also worth noticing that it is possible to improve the adhesion of CFs to the geopolymer matrix by using plasma pretreatment of the fibers, which may both change their surface topography, making their surface rougher, and depositing new functional groups on their surface, allowing them to bond with the matrix more effectively [41,42]. The mechanism of surface modification depends on the gas used in the plasma generator. For example, argon plasma makes the surface rougher due to plasma etching and improves the interlaminar strength by increasing the adhesion of fibers to the geopolymer matrix [41,43].

Recently, some trials were made with sizing methods, which create the physical-chemical link between a surface of CF and a geopolymer matrix [44]. They show that the sizing of a CF has a strong influence on the impregnation of a CF by minerals, as well as affecting the quality of the bonding between a CF and a geopolymer matrix [44]. It is worth noticing that this method was not investigated and it seems to be a perspective for further investigation.

3.2. Textiles, Grids, and Fabrics

Textiles, grids, and fabrics reinforced in a geopolymer matrix are very often applied in the context of improving mechanical properties, including material behavior in elevated temperatures [45,46]. This kind of reinforcement is applied as one or multiple layers of 2D or 3D fabrics or grids [46,47]. These kinds of composites have many advantages; however, they also have some weak points, including high cost and partial carbon oxidation at high temperatures [48,49]. In the case of the design of this type of geopolymer, the main challenge is usually the proper bond between the textile reinforcement and the geopolymer matrix [46,49]. The bonding is usually the weakest element of the composite where the failure mechanism started [46,49].

The reinforcement of carbon textiles compared to other types of textiles generally shows better mechanical properties, especially tensile strength and Young's modulus [31,50]. Additionally, these kinds of composites are investigated as materials against electromagnetic fields that are emitted by electronic and telecommunication devices, such as mobile phones [51]. The research in this area was carried out using the reinforcement of geopolymer composites with carbon or basalt fiber grids and nanoparticles of MgO [52]. The results show the shielding efficiency of the investigated solution with the CF in the 30 MHz–1.5 GHz frequency range [51]

3.3. Carbon Microfibers, Nanofibers, and Carbon Nanotubes

In recent years, micro and nanofibers became more and more popular as an additive to the geopolymer matrix. It is not only the result of their excellent mechanical properties, but also new research, including the development of new methods of biodegradation of these nanomaterials by fungi, bacteria, plants, animals, and microbial enzymes, which make it more environmentally friendly [52]. The micro and nanofibers could have different threads, filaments, whiskers, nanotubes, nanoparticles, and also other graphene-based materials with diameters of about 0.5–1.5 µm or even finer [53,54]. The implementation of this type of reinforcement comparison with traditional CFs helps in the homogenous distribution of the fibers [53,54]. In the case of nanoreinforcement, the most obvious advantages appear to be small amounts of material, usually less than 0.2 wt.%, especially from the point of view of increasing mechanical properties [53,55,56]. As part of geopolymer composites, in addition to improving their mechanical properties [56], carbon nanotubes can lower their porosity and decrease the water absorption rate to improve their durability in humid or aggressive environments [56,57]. It also positively influences crack reduction and prevents any catastrophic fractures, including thermal cracks at elevated temperatures [47,57,58]. They also significantly increase electrical conductivity [59].

Other studies on the addition of microfibers (fibers about 100 µm in length) were carried out in the weight ratio of 0, 5, 10, and 15% to the metakaolin-based geopolymer matrix [58]. The mechanical properties were tested after 28 days at temperatures of 30 °C, 200 °C, 400 °C, and 800 °C. The best results were obtained for the 10% addition of the carbon microfiber, for temperatures of 30 °C and 200 °C. They were for 30 °C—44.2 MPa for the material with a 10% addition of microfibers and 28.4 MPa for the matrix material. By analogy, the compressive strength for a temperature of 200 °C was 48.8 MPa and 36.6 MPa, respectively. The best values were achieved for temperatures of 400 °C and 800 °C for a 15% fiber addition. They were, respectively, 33.5 MPa and 24 MPa, compared to 14.8 MPa and 11.2 MPa for a pure matrix for the same temperatures [58]. Research was also carried out on the addition of carbon nanotubes to geopolymers, both as an additive to metakaolin-based matrices [60,61] and fly ash [56]. The addition of nanotubes enhanced the mechanical properties of the composites.

Other research shows that geopolymer composites with silica-grafted carbon nanotubes can be protective materials against electromagnetic radiation [62,63]. Obtaining effective shielding properties requires the use of a large number of carbon nanotubes—the highest effectiveness was achieved with a 5 vol.% addition of nanomaterial [63].

3.4. Hybrid Reinforcement

Carbon fibers are also used for hybrid reinforcements, i.e., with two different types of fibers. The correct combination of different types of fiber could efficiently improve composite properties [64,65]. In this kind of composition, a synergistic effect occurs by strengthening the influence of one fiber on the matrix on the other one. In the results obtained, the properties of the composite are better than the sum of the application of both fibers separately [65,66].

Yang et al. [64] investigated carbon nanotubes (30–50 nm in diameter) combined with polyamide (PA) fibers, with a length of 5 mm and a diameter of 12 µm. The metakaolin-based geopolymer was activated with phosphoric acid. The fibers were added to the matrix in the following proportions: 0.0, 0.5, 1.0, 1.5, 2.0, and 2.5 wt.% [64]. The behavior of the material without reinforcement was compared with that of PA fiber reinforcement and hybrid reinforcement. The greatest mechanical properties were reached for the hybrid reinforcement. The compressive strength was about 115 MPa for 1.5% of the hybrid reinforcement comparison to the same amount of PA fibers (approximately 90 MPa), which significantly increased. This improvement was even more significant compared to the value obtained for a pure matrix—51.7 MPa [64]. Research also confirmed an increase in bending strength and fracture properties. The composite with a 1.5% addition of hybrid fibers has about 38 MPa of bending strength compared to 27 MPa for the same amount of PA and 9.9 MPa for the reference sample (without reinforcement) [64].

Promising results were also obtained for hybrid steel and carbon reinforcement [65]. Both fibers are also well-known as reinforcements that develop the mechanical properties of the geopolymer matrix [66–68]. The research involves a lightweight geopolymer matrix based on fly ash and microspheres. The results of this investigation show an improvement in the mechanical properties of composites. The highest value of bending strength was obtained for reinforced 1.5 wt.% CFs and 0.5 wt.% steel fibers [67]. Similar works were made for hybrid reinforcement with carbon and SiC fibers. They show an improvement in flexural strength and also Young's modulus for compositions at elevated temperatures [69–71].

4. The Technology of Preparing Carbon Fibers Reinforced with Geopolymer Composites

Casting technology is most often applied to geopolymer manufacturing. It is also the most often applied to composites with carbon fibers in case of using all types of fibers—short, long, textile, etc. [6,37,72]. In this case, different parameters are used for the production of geopolymers, such as temperatures and activators [6,72]. However, it is worth noticing that the addition of CFs does not influence production parameters and usually, the technology does not require significant modifications. A slight exception can be the addition of a significant amount of CFs that can influence the processing properties through the density of the paste. Some research shows that an addition of about 5 wt.% makes processing impossible [39].

The alternative solution for casting technology is additive manufacturing. It is one of the fastest growing manufacturing technologies with increasing importance in different sectors of the economy. Additionally, in the case of geopolymers reinforced by different types of fibers and their application in the construction industry, this technology became more and more important [73,74]. In the case of a CF reinforced by a CF, this technology is still in the development stage—only preliminary research was provided [75,76]. However, this research shows promising results for the technology, such as a positive influence on flexural strength, improved interlayer bonding, reduced cracking propagation and shrinkage; but they have not yet been not tested on the industrial scale [75,76].

CFs were investigated as short and long fibers. The reinforcement of short fiber-printed 3D geopolymer composites by short fibers was studied for the CF with a length of 5 mm, a diameter of 8 μm in an amount of 1 wt.% in a fly ash-based matrix. The addition significantly improves ductility, tensile, and flexural strength, and has a positive influence on compressive strength [27]. Similar results were obtained for long fibers or carbon microcables [77,78]. The development of this technology seems to be a valuable alternative for steel reinforcement in the case of 3D printing.

Other possible application methods of production are different kinds of foaming processes. There are two main technologies for the production of foamed geopolymers, one using aluminum powder and the second using peroxide (H_2O_2) [79]. Both of them also use fibers. There are usually short fibers, less often combining foamed geopolymers with textiles [79,80]. In the case of CFs, research was made on short fibers, especially nanotubes [81–83]. The results show the positive influence of CFs on flexural strength and fracture toughness [81–83]. Additionally, the thermal conductivity of the materials increased approximately two times [83]. In the case of foamed materials, an interesting property is a possibility of using by-products of the secondary aluminum industry as foaming agents [84]. These by-products are considered hazardous waste, and their use as geopolymer foaming agents may significantly reduce the load of these industries on the environment and serve as part of the circular economy. Due to the high aluminum content, by-products may react with alkaline compounds within the geopolymer mix and produce hydrogen, which foams the geopolymer [84]. These seem to be also interesting opportunities for the development of CF reinforcements in this type of material.

5. The Influence of Carbon Fibers on the Properties of Geopolymer Composites

5.1. Processing Properties

CFs have an influence on manufacturing properties, including castability and shrinkage. CFs and various other fibers may be used to lower the dry shrinkage during the geopolymer curing process, including geopolymers cured at room temperature and heat-cured geopolymers. The dry shrinkage is significantly lower for heat-cured geopolymers, as higher temperatures increase the geopolymerization rate. Higher fiber content also reduces dry shrinkage [85]. However, the addition of CFs reduces the workability of different types of geopolymers. The workability decreases making it harder to mix, place, cast, etc., with minimal loss of homogeneity, according to slum cone tests [86]. CFs are also perfectly elastic and are, therefore, less affected by fatigue deformation during shrinkage and other processes (including loading and unloading of the composite) [53]. Research was also conducted on geopolymer composites with carbon nanotube additives. Carbon nanotubes also reduce workability by increasing the viscosity of the geopolymer mix [59].

5.2. Physical and Mechanical Properties

Among the mechanical properties of the geopolymer composites with CFs, compressive strength and flexural strength were the most tested [6,10]. Other investigated properties were connected with density, creep, brittleness, fracture toughness, hardness, specific modulus, tensile strength, etc.

A study with the use of short CFs was carried out with the use of the metakaolin-based geopolymer matrix with the addition of slags [87]. The bending strength of the composites was tested with 1.0% by weight of CFs with a diameter of 10 μm and a length of 7 mm. The results showed the strengthening of the composite. For the pure matrix, the bending strength was 6.9 MPa and for the reinforced material it was 11.7 MPa [87].

A lot of research was dedicated to the possibility of applying CF-reinforced geopolymer composites as a material for usage at elevated temperatures. A study of the high-temperature behavior of geopolymer composites with the addition of short CFs also concerned the mechanical properties [88]. CFs were added in amounts of 0, 0.5, 1, and 1.5 wt.% to a fly ash-based geopolymer matrix. The research was carried out at temperatures 28 °C, 200 °C, 400 °C, 600 °C, and 800 °C. The dimensions of the fibers were approximately 11 μm in diameter and 6 mm in length [88]. The main findings of the provided tests for the compressive strength at 28 °C show a growth of this property for composites containing 1 and 1.5% CF (approx. 31 and approx. 32 MPa, respectively) and a reduction in compressive strength for a material containing 0.5% CF—approx. 27 MPa, in comparison to the pure matrix material—about 29 MPa [88]. At temperatures of 200 °C, 400 °C, and 600 °C, the compressive strength growth in relation to the materials was investigated at an ambient temperature (28 °C). Only at the temperature of 800 °C was there a reduction in compressive strength for composites with 0.5 and 1.5% CF. The best results were realized at the temperature of 200 °C. It was, respectively, 0.5% CF —about 36 MPa, 1% CF—about 40 MPa, and 1.5% CF—about 36 MPa [88]. The investigation positively verifies the possibility of using composites with CFs in products for high temperatures.

Research on the addition of short CFs, taking into account the impact of temperature on the mechanical properties of composites, was also made on a matrix prepared as a mixture of fly ash and metakaolin [89]. CFs with the following dimensions were included in the matrix: a length of 6 mm and a diameter of 7 μm. The following proportion of the additives was used: 0, 0.5, 1, and 2% by weight. Specimens were investigated after 7 days at an ambient temperature and at 500 °C [89]. The outcomes from the provided tests at the ambient temperature indicated a reduction in the value of compressive strength from about 50 MPa for the material without CFs to about 45 MPa for the composite with the 2% CFs. However, at a temperature of 500 °C, the composite with the CF was more durable and reached about 5 MPa compared to about 2 MPa for the pure matrix material [89]. The bending strength was improved with the addition of CFs, for both temperatures. For the material without CFs, it was approximately 5.5 MPa and below 0.1 MPa, respectively, and for composites with the 2% addition of CF, it was 15 MPa and 1 MPa [89].

Other research was performed on the fly ash-based geopolymer reinforced with cut CFs (exact length not specified) with a diameter of 7 µm. CFs were put into the composites in the following weight proportions: 0.0, 0.1, 0.2, 0.3, 0.4, and 0.5%. Two types of samples were tested and cured at 25 °C and 60 °C. The tests were carried out after 7, 14, and 28 days, progressively observing the rise in compressive strength of the composites [90]. After 28 days, the greatest results were achieved for 0.5% CF content. For specimens cured at 25 °C it was about 43 MPa for the composite with a 0.5% CF; for comparison, the matrix material achieved 30 MPa. For specimens cured at 60 °C it was about 58 MPa for the composite with a 0.5% CF, the matrix material made under the same terms achieved 45 Mpa [90]. In addition to the compressive strength tests, tests related to the electrical conductivity of composites were also carried out. Conductivity increased with an increasing amount of carbon fibers [90].

Another study investigated the effect of CFs on flexural strength, fracture work, and Young's modulus based on the volume fraction of CFs in a metakaolin-based geopolymer. The highest values of flexural strength and work of fracture, specifically 91.3 Mpa flexural strength and 6435.3 J/m^2, were measured on a sample containing 3.5 vol.% of short CFs; higher contents led to the deterioration of these properties. Young's modulus peaked at 6 vol.% of the CF, with a value of approximately 20.5 GPa [91].

The overall carbon-based fibers decrease the density of the geopolymer composite, because of the lower density than the density of the matrix [53,67]. Additionally, they are the most suitable reinforcements for the strength geopolymer composite [72]. These types of fibers increased not only the flexural and bending strength but also the compressive strength (Table 2.). This phenomenon is typical for fibers with a high Young's modulus, including steel and CFs, and can be confirmed in previously provided research [53].

Table 2. Mechanical properties of composites—carbon fibers reinforced with geopolymers.

Fiber	Geopolymer Matrix	Compressive Strength (Matrix) [MPa]	Flexural Strength (Matrix) [MPa]	Compressive Strength (Composite) [MPa]	Flexural Strength (Composite) [MPa]	Reference
Short CFs (length 7 mm; 1.0% wt.)	Metakaolin + slag	—	6.9	—	11.7(+69.9%)	[87]
CFs (6 mm length, 11 µm diameter, 0.5% wt.)	Fly ash	29	-	34 (+17%)	-	[88]
Short CFs (length 7 mm; 1.0% wt.)	Fly ash	50	5	46 (−8%)	15 (+200%)	[89]
Carbon microfibers (100 µm length)	Metakaolin + shale clay	28.43	-	38.97 (+37%)	-	[58]
Short CFs (7 mm length, 4.5 vol.%)	Metakaolin	-	16.8	-	96.6 (+475%)	[91]
Short CFs (7 mm length, 6 vol.%)	Metakaolin	-	16.8	-	87.4 (+420%)	[91]
Short CFs (7 mm length, 7.5 vol.%)	Metakaolin	-	16.8	-	42 (+150%)	[91]

It is also worth noticing the mechanism of reinforcement by the long CFs of the geopolymer matrix. CFs work effectively against sample destruction, even after the first appearance of cracking [40,78]. The appearance of the fibers actively works against crack propagation, even in elevated temperatures. Firstly, because of working against microcracking propagation thanks to additional "barriers" in the form of fibers in the material. Secondly, because of the maintenance of the continuity of the matrix material [40,78]. The second process is clearly visible in Figure 4, where the flocks of the fibers are visible in the cracking point for the different types of the geopolymer matrix. They are evidence of working of the fibers against the sample decoherence. After the force was released, the CFs try to return both halves of the samples to the previous shape.

(**a**) (**b**)

Figure 4. Geopolymer samples reinforced by long carbon fibers. (**a**) Metakaolin-based matrix and (**b**) Fly ash-based matrix.

5.3. Isolation Properties—Acoustic and Thermal Isolation

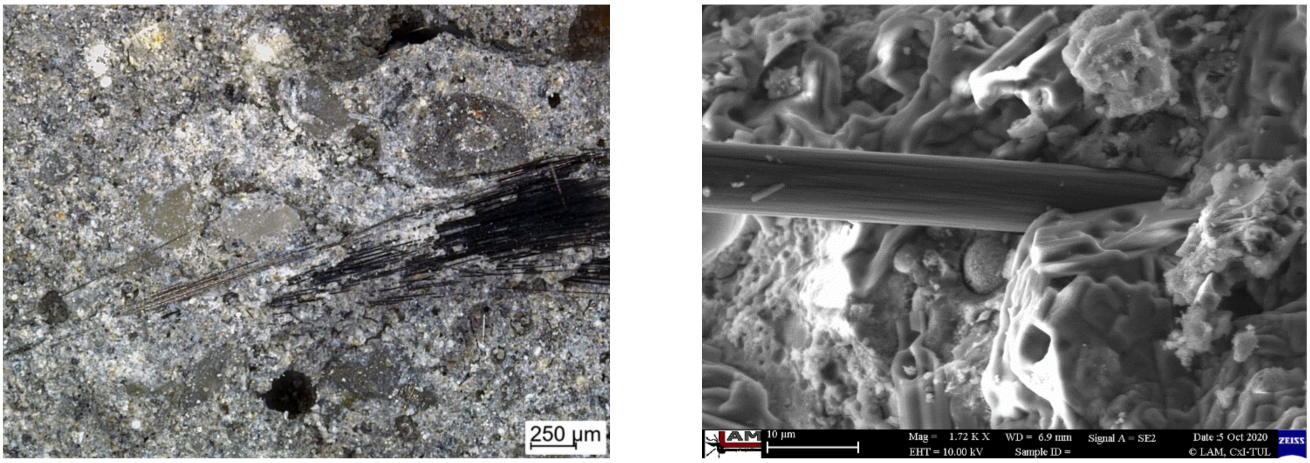
Although the acoustic influence of carbon fibers as additives to geopolymers has not yet been sufficiently investigated, the fibers influence the thermal isolation properties of the geopolymer composite. While geopolymers may be used as an insulating material, especially in foam form, carbon nanofibers increase their thermal conductivity and, therefore, make their thermal isolation properties worse. This effect can be mitigated by foaming the composite, which mitigates the effect of fibers on thermal isolation and allows fibers to serve as a reinforcing additive in foamed geopolymer composite, as its mechanical properties are worse compared to compact geopolymers [84].

5.4. Durability, Creep, and Chemical Properties

Geopolymer composites reinforced with CFs are very durable and may be used to improve the durability of other structures, including deteriorated ones, such as bridges made of reinforcement concrete, even in an aggressive environment [92]. For example, when tested with an accelerated corrosion environment simulated by hydrochloric acid solutions of various concentrations, hybrid geopolymers and epoxy resin-based carbon reinforced composites were able to slow acid penetration into existing reinforced concrete bridge structure by nearly 30% when compared with steel jacketing, making it a potential method of increasing the service life of structures made of concrete and other materials. However, the geopolymer is still susceptible to degradation, reducing its compressive and tensile strength, which are improved by carbon fibers which, therefore, significantly reduce the effect of degradation or at least slow its progress and extend the durability of the geopolymer before degradation becomes critical [92].

Research was also conducted on geopolymer composites with carbon nanotubes. The addition of carbon nanotubes reduces water absorption by lowering the porosity of geopolymers. A lower water absorption rate reduces the susceptibility of the geopolymer composite to various environmental effects, including the absorption of aggressive chemicals and microbial degradation [59].

It is also worth mentioning the durability investigation. Research shows that geopolymer composites with CFs have better durability than glass fibers or basalt fibers [29,93]. These kinds of properties are usually connected with good adhesion and homogeneity of these kinds of composites (Figure 5). Unlike glass fibers that show sliding behavior and detachment with the geopolymer matrix, CFs show good cohesion in elevated temperatures [94,95].



(**a**) (**b**)

Figure 5. Microstructure of geopolymers reinforced by CFs. (**a**) CFs in the geopolymer matrix—the confocal microscope, surface investigation on breakthroughs and (**b**) CFs in the geopolymer matrix—scanning electron microscope (SEM).

There were also some investigations connected with the creep behavior of geopolymer composites reinforced by CFs in the literature [96]. The results show that the specific creep strain of specimens with CFs is better than the plain geopolymer matrix—about 12% [97]. In comparison to other geopolymer composites, this behavior is even better than for other types of fibers [97].

5.5. Electrical and Magnetic Properties

Although pure geopolymers are electrically conductive due to the presence of alkali ions in their structure, their electrical conductivity is low [98,99]. CFs have very high electrical conductivity and can significantly increase the electrical conductivity of the geopolymer composite. Because of this, they are used for the fabrication of conductive composites with geopolymers (as well as other matrices). The increase in conductivity is significant even with very low amounts of CFs [98] or the addition of graphene oxides [100,101].

As mentioned earlier, CFs may also be used as electromagnetic shielding (EMI shielding) in geopolymer composites because their high electrical conductivity allows them to reflect electromagnetic waves. When CFs are evenly distributed in the geopolymer matrix, increasing their content leads to a gradual increase in EMI shielding, although the rate of increase in their shielding capabilities decreases with increasing content, as EMI shielding properties are saturated. The shielding efficiency is also improved when larger carbon fibers are used [98]. They are also effective when using carbon grids (nets made from carbon fibers). When used as EMI shielding, geopolymer composites with dense carbon grids (such as HTC 10/15) are able to completely block Wi-Fi signals at a distance of around 6 m, with a 60% reduction in signal intensity at 2 m [99].

Research was also conducted on geopolymer composites with carbon nanotubes [59,102]. Even small amounts of carbon nanotubes may significantly increase the conductivity of geopolymer composite and lower its electric resistance and impedance. Increasing the number of carbon nanotubes in the geopolymer composite increases the formation of "conductive paths" and lowers the effect of tunneling gaps. However, these effects significantly depend on other factors, such as mixture composition, curing conditions, agglomeration of carbon nanotubes, etc. [59].

5.6. Thermal Properties

In terms of room temperatures, the main aim of the addition of CFs is to increase the mechanical properties and change the behavior of composites in terms of fracture characteristics, avoiding a brittle fracture [103]. CFs can play a similar role at elevated temperatures, especially up to 500 °C [58]. CF additives increase heat conductivity, the specific heat, and the diffusivity. For example, in a study on foaming geopolymers by adding by-products from the secondary aluminum industry, a simple addition of carbon nanofibers increased thermal conductivity by 12.5%, specific heat by 3%, and diffusivity by 9% [84].

CF additives for geopolymer composites improve their properties at high temperatures. Geopolymer composites with CFs retain their mechanical properties even at temperatures up to 700 °C, and fibers also provide an effective crack control mechanism and improve their flexural strength at temperatures up to 500 °C [89]. The explanation for this fact is that in the air atmosphere, CFs are oxidized to CO_2 and at more than 500 °C they lose their properties [38,104]. For the higher temperature, it is recommended to use a SiC fiber which kept 80% of mechanical properties even at 1200 °C in the atmosphere [38,104]. Nevertheless, in protecting the atmosphere, CFs can hold their properties up to 2000 °C [104,105].

5.7. Other Properties

CFs may also be functionalized with antimicrobial agents in order to be used for protection against bacteria and other harmful microorganisms. These antimicrobial agents may include organic compounds, such as antibiotics, including gentamicin [106], or inorganic agents, including nanoparticles of silver or other metals [107]. It is also possible to manufacture them with an additive of silver salts, such as silver nitrate, or to give antimicrobial properties to the fibers themselves [108]. With this functionalization, CFs can be used as antimicrobial agents in composites, including building materials, such as concrete, which may be vulnerable to microbially-induced degradation (MIB), especially in a humid environment, where their surface can be colonized with sulfur-oxidizing or nitrifying bacteria, which lowers their pH and allows colonization by other microorganisms, which then degrades it chemically or mechanically. Although, there are sufficient alternatives, including silver nanoparticles or photocatalytic particles [109].

6. Applications for Carbon Fibers Reinforced with Geopolymer Composites

As geopolymer composites with carbon (and other) fibers have significantly improved mechanical properties, including compressive strength, flexible strength, and tensile strength, one of their main application potentials is as building materials, where they may serve as a replacement for other building materials, especially concretes based on ordinary Portland cement (OPC), as their mechanical properties are superior. However, their use as a building material is hampered by higher prices when compared to concrete and the necessity of using caustic activator solutions, which presents logistical and safety problems. They may also be used to create lightweight construction materials [99] or can be used in the 3D printing of structures or pre-fabricated construction blocks [27]. Due to their mechanical and chemical resistance, geopolymers with CF additives may be used to repair various other structures, including deteriorating concrete structures in aggressive environments [92].

The use of geopolymer composites reinforced by CFs as thermal insulation may be impeded by the high thermal conductivity of CFs, as they also increase the heat conductivity of the geopolymer composite. However, this effect can be mitigated by foaming the geopolymer, since CFs still significantly increase the mechanical properties of the geopolymer foam and mitigate the reduction in mechanical properties caused by foaming, making them suitable for this application, although other types of fibers with lower thermal conductivity may be better for this application [84]. The exemplary product in this area is fire-resistant panels for aircraft cabin interiors [72,110].

COVID-19 accelerated work on the antibacterial properties of different materials, including CF-reinforced geopolymer composites [109,111]. The most promising results were obtained in this case with CFs and the addition of silver nanoparticles or silver nitrate additive [108,109]. This kind of composite gives new possibilities for the application of geopolymer composites, especially for infrastructure where aseptic properties are required, including medical buildings.

There are also some new areas of applications for geopolymers reinforced by CFs, especially in the form of nanofibers or nanoparticles. These types of applications are related to the electrical conductivity and piezoresistivity of this kind of composite [15,56]. The newest research shows that highly electrically conductive composites with nanoadditives can be manufactured by 3D printing technology [74,112]. The new area of investigation is also the use of recycled fibers for the manufacture of biodegradable composite materials, and other environmental applications that are urgently required [109].

7. Conclusions

The review shows the increasing importance and new area of application of CF-reinforced geopolymers. These kinds of composites are investigated for many years, but their applications were limited, especially because of the high price of CFs. Nowadays, this situation is changing, because of the new technology of CF production that allows this fiber to be obtained from recycling as well as from renewable sources.

The paper also indicates the growing importance of different industries. It is possible not only because of the sources of feedstock for CF production but also because of development of the new technologies, especially additive manufacturing. This technology allows, i.a., for easier testing of the new products in a small series.

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