

Review Article

Mechanical Properties of Natural Fibre Reinforced Geopolymer Composites: A Review

Noor Abbas Al-Ghazali¹, Farah Nora Aznieta Abdul Aziz^{1*}, Khalina Abdan² and Noor Azline Mohd Nasir¹

¹*Housing Research Centre (HRC), Department of Civil Engineering, Faculty of Engineering, Universiti Putra Malaysia, 43400 UPM, Serdang, Selangor, Malaysia*

²*Institute of Tropical Forestry and Forest Product (INTROP), Universiti Putra Malaysia, 43400 UPM, Serdang, Selangor, Malaysia*

ABSTRACT

The cement production consumes many natural resources and energy, pollutes the environment, and cannot meet the current building materials' green and sustainable development requirements. Therefore, geopolymers have gained popularity as cement replacements in recent years. Geopolymers have promising characteristics such as low energy consumption and carbon footprint, valuable compressive strength, fire resistance, flame resistance and good durability. However, these materials suffer from low tensile and flexural strength. Hence, fibres are added to overcome these issues and enhance their toughness index. Natural fibres are biodegradable, low-cost, renewable materials and widely available in many countries. This article reviewed previous Natural Fibre Reinforced Geopolymer Composites (NFRGC) studies, focusing on compressive strength, tensile and flexural strengths, and toughness. In addition, the available literature on the effect of the treatment methods of natural fibres on the mechanical properties of NFRGC

has been addressed. The findings indicate that adding the appropriate type and content of natural fibres to geopolymer composites can enhance their mechanical properties. However, more attention should be paid to the effects of the pre-treatment of natural fibres on the performance of NFRGC.

Keywords: Fibres, geopolymer composites, mechanical properties, natural fibres

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E-mail addresses:

na706050@gmail.com (Noor Abbas Al-Ghazali)

farah@upm.edu.my (Farah Nora Aznieta Abdul Aziz)

khalina@upm.edu.my (Khalina Abdan)

nazline@upm.edu.my (Noor Azline Mohd Nasir)

*Corresponding author

INTRODUCTION

Davidovits introduced the concept of geopolymers in 1978. These inorganic polymers are produced by the chemical reaction between an aluminosilicate source with an alkaline activator through the geopolymerisation process that results in polymeric chains and cross-linked networks consisting of Si-O-Al-O bonds of comparable or greater strength than that of Ordinary Portland Cement-based composites (OPCC) (Ganesh & Muthukannan, 2021; Farhan et al., 2018). A literature review showed that Geopolymer Composites (GPC) have excellent mechanical strength and durability. These outstanding attributes include low porosity (Fang et al., 2018), high early strength (Jawahar et al., 2016), and high performance in sulphate and acid environment (Gopalakrishnan & Chinnaraju, 2019), and elevated temperature resistance (Yasaswini & Rao, 2020). These features make GPC a potential candidate for various industrial applications (Zhang et al., 2020). However, both the OPCC and GPC are brittle materials with low tensile strength. Reinforcing these materials with different kinds of fibres, such as synthetic, steel, or natural fibres, is one of the most common methods to overcome these weaknesses. Besides, many researchers claimed that geopolymer composites had greater material greenness than traditional cement and that fibre reinforcing can improve their ductility and durability (Bellum, 2021).

The use of natural fibres as a replacement for conventional fibre reinforcements in composites has gained popularity in recent years. The production of economically viable, ecologically sustainable, and healthful products based on natural sources is gaining popularity. Using natural fibres as reinforcements in geopolymer composites is an attractive alternative for the building industry. Several studies on the strength and behaviour of geopolymer composites reinforced with natural fibres have been published in recent years. Since natural fibres are abundant in many developing countries, more in-depth studies should be conducted on the various issues related to their utilisation.

The Natural Fibre-Reinforced Geopolymer Composites (NFRGC) contain two main parts, as shown in Figure 1. The first part is the geopolymer matrix, which comprises two main components (the binder and the alkaline activator). For the binder, different aluminosilicates sources have been used by researchers to produce geopolymer binders. These sources are derived either from a natural source such as kaolinite, albite, clays or from industrial by-products like Ground Granulated Blast Slag (GGBS) (Gupta, 2021; Fang et al., 2018), Fly Ash (FA) (Abdulmuttaleb et al., 2022; Chen et al., 2022; Chindaprasirt et al., 2021), Palm Oil Fuel Ash (POFA) (Ayub et al., 2021), Rice Husk Ash (RHA) (Abbass & Singh, 2021) and Silica Fume (SF) (Liang et al., 2021). The other primary component of geopolymer is the alkaline activator, which is essential to creating Al and Si crystals. The most popular alkali activator is sodium silicate or potassium silicate combined with sodium hydroxide or potassium hydroxide. However, a single alkali activator can also be utilised in geopolymer composites.

On the other hand, the second part of NFRGC is the Natural Fibres (NF), which can be derived from three different sources (plant, animal, and mineral source). Compared to animal and mineral fibres, plant fibres are more favourable due to the difficulties of collecting the fibres from animals. In addition, these fibres contain a high amount of protein in their structure. While for the mineral fibres, fibres should undergo several processes before being included in the geopolymer. The plant fibres can be derived from different parts of the plant, such as the bast, stalk, leaf, seeds, grass, and fruit. This article reviews previous studies on the mechanical properties of NFRGC with a focus on compressive, tensile, and flexural strengths and toughness.

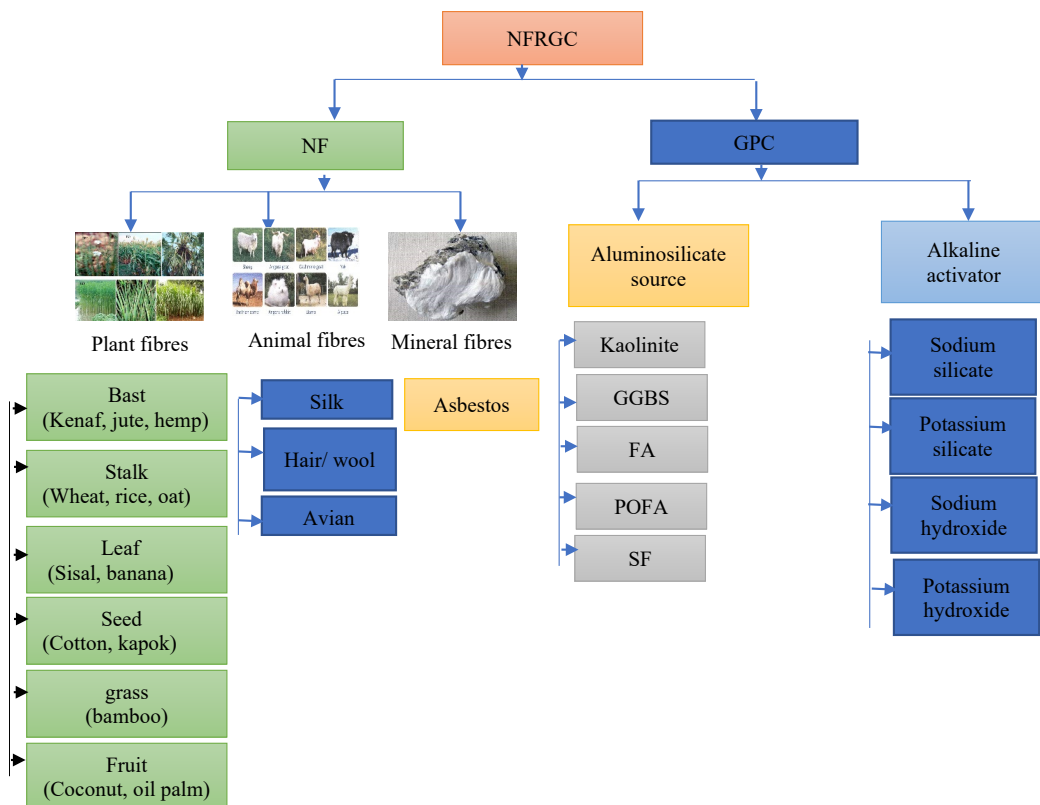


Figure 1. NFRGC components

MECHANICAL PROPERTIES OF NFRGC

Compressive Strength

The inclusion of NF can highly affect the compressive strength of geopolymers, and this effect is dependent on both the matrix and fibre type, properties, content and length.

Korniejenko et al. (2016) examined the performance of the NFRGC prepared with different types of NF and how adding 1% by weight of the composite fibres will affect the strength properties of geopolymer. The results showed that NFRGC containing coconut fibres, sisal, or cotton fibres achieved higher compressive strength than the neat geopolymer. Only the NFRGC prepared with raffia fibres showed lower strength than the latter unreinforced composite due to the weak interfacial bonding between raffia fibres and the geopolymer matrix. According to Yanou et al. (2021), the increase in the sugarcane bagasse fibres (SBF) content resulted in a reduction in the compressive strength of NFRGC. The findings revealed that the inclusion of SBF in percentages of 1.5%, 3%, 4.5%, 6% and 7.5% caused reductions of 18.3%, 31.8%, 43.1%, 65.9% and 71.8% in comparison with the neat geopolymer, respectively. The authors attributed the strength reduction to the high void content in the NFRGC mixtures, which created weaker zones within the geopolymer matrix.

On the contrary, Silva et al. (2020) reported that the compressive strength of NFRGC containing jute or sisal fibres was 64% and 76% higher, respectively, compared to the neat geopolymer. Besides, the test data showed that the strength of NFRGC tends to increase with the fibres content, up to a limit where a reduction was observed, and the type of fibre determines the optimum content and strength. Similarly, Alomayri and Low (2013) reported that increasing the cotton fibre percentage from 0.3% to 0.5% by weight in the NFRGC increased the compressive strength by 65%, while further adding cotton fibre (beyond 0.5%) caused a strength reduction. The improved compressive strength for the NFRGC containing the appropriate content of fibres is because a great amount of stress has transferred from the matrix to the fibres, which causes a higher load carried by the fibres. Besides, the good distribution of NF in the matrix enhanced the interfacial bonding between the fibre and the matrix.

On the other hand, the strength reduction with higher fibres could be attributed to the increased porosity due to the agglomeration of the fibres. In a different study, Alomayri et al. (2014) studied the performance of NFRGC prepared with cotton fibre in the fabric form with various contents and orientations. The findings revealed that NFRGC prepared with horizontally oriented fabric exhibited higher compressive strength than composites prepared with vertically oriented fabric. The horizontally oriented fabrics can absorb and transfer applied loads uniformly over the cross-section. Also, the horizontally oriented fabric-matrix interface is not subjected to shear loadings, which decreases the risk of fabrics detachment or delamination at high loads. Besides, increasing the fabric layers for both types increased the strength of NFRGC. Likewise, Assaedi et al. (2015) observed that the inclusion of the flax fabric significantly increased the strength properties of geopolymer, and increasing the fibre dosage caused a higher strength improvement. The compressive strength of plain geopolymer increased from 19 MPa to 91 MPa for NFRGC containing 4.1% by weight flax fibres. This improvement could be attributed to the ability of the fibres to absorb and transfer the stress from the matrix.

Zulfiati and Idris (2019) examined the influence of the length and content of NF and the alkalinity of the geopolymer on the performance of NFRGC. The authors incorporated three different lengths of pineapple leaf fibre (10 mm, 20 mm, and 30 mm) with a content of 0.25 and 0.5% to FA-based GPC activated by sodium silicate and sodium hydroxide of two different concentrations (14 M and 16 M). The findings indicated that the length of the fibre showed a very significant impact on the strength of the composite, and for both fibre percentages, NFRGC containing pineapple fibres of 30 mm length achieved better strength than the composites reinforced by fibres of 10 mm and 20 mm lengths, as shown in Figure 2. NFRGC containing 0.5% fibres of 30 mm length and activated with sodium hydroxide of 16 M achieved the highest compressive strength of 41.4 MPa. It could be attributed to the fact that fibres of 30 mm in length were more effective in withstanding and transferring the applied stress to other composite portions (Islam & Ju, 2018). Furthermore, the NFRGC activated by higher sodium hydroxide concentration has good performance because the matrix binding process is faster by increasing the concentration. Therefore, it causes the polymerisation process to be more maximal with increasing time, resulting in a stronger bond between the fibre. Hence, the matrix, and the composite's performance is highly dependent on interfacial bonding.

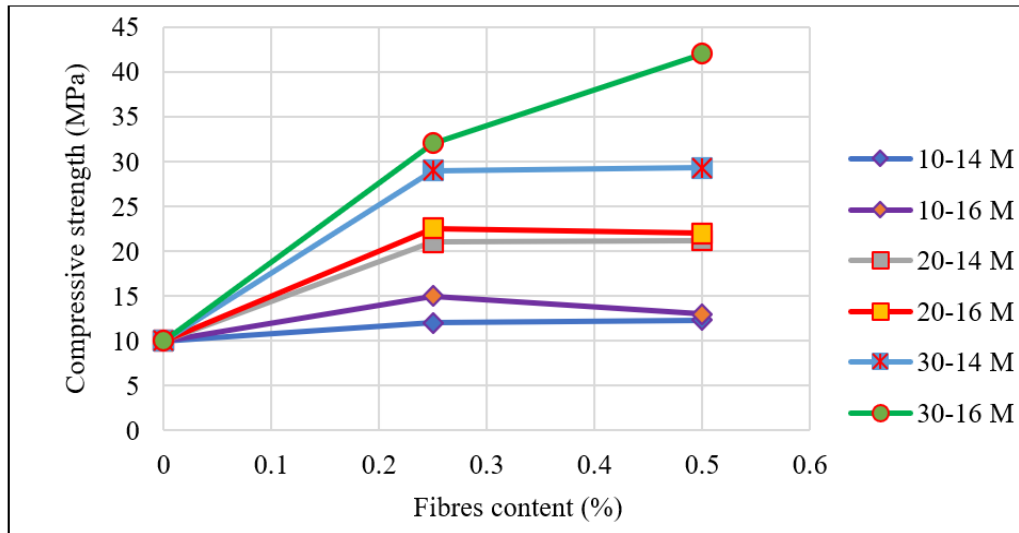


Figure 2. Impact of the length and content of NF on the compressive strength of NFRGC (Zulfiati & Idris, 2019)

In summary, NFRGC achieved a compressive strength of about 20% to almost 55% higher than the plain geopolymer mixes, as shown in Figure 3. Besides, in all the works related to NFRGC, there was an optimum content for fibre inclusion depending on the

matrix and fibre type and properties. Therefore, a preliminary study should be conducted before applying a large scale. Another note is that NFRGC containing the NF in the fabric form exhibited higher strength improvement than short fibres due to the better distribution of the continuous fibres in the geopolymer composites. Besides, their alignment in the direction of tension effectively bridges the cracks, resulting in higher stress transfer at the fibre-matrix interface.

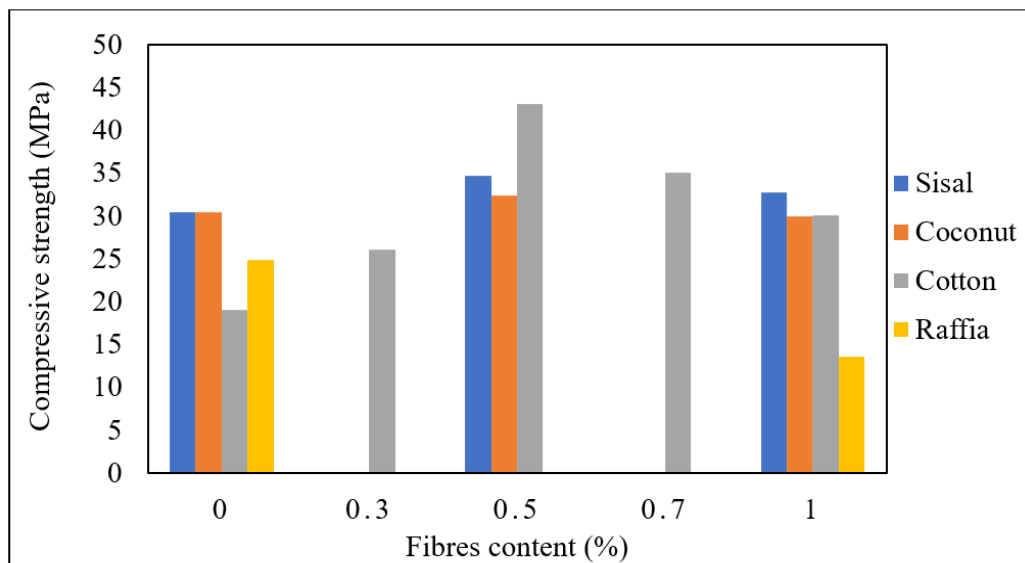


Figure 3. Variation of compressive strength of NFRGC prepared with different types of NF (Alomayri & Low, 2013; Korniejenko et al., 2016; Wongs et al., 2020)

Flexural and Splitting Tensile Strengths

Wongs et al. (2020) compared the performance of NFRGC prepared with two different types of NF and geopolymer composites prepared with synthetic fibres. The NFRGC was prepared with 0.5%–1% sisal fibre or coconut fibre, while the other composite was prepared with 0.5%–1% glass fibre. The results showed that the NFRGC and the composites prepared with the glass fibres achieved higher flexural and tensile strengths than the plain geopolymer. Besides, the tensile and flexural strengths of the fibrous composites tended to increase with increasing the content of the fibre. For example, NFRGC achieved a flexural strength of about 5.3–6.6 MPa, which was higher than the flexural strength of normal GPC (3.1 MPa), while GPC containing glass fibres achieved a flexural strength of about 3.1–3.7 MPa. The strength improvement could be attributed to the fibre’s high tensile strength and elasticity modulus. That stress in the matrix could be transferred to the fibre via the matrix-fibre interface.

Silva et al. (2020) examined the performance of NFRGC containing sisal fibres with an aspect ratio of 73 and content of 0.5%–3%, or jute fibres with an aspect ratio of 189 and content of 0.5%–2%. The results revealed that NFRGC showed higher tensile and flexural strength than the plain geopolymer without fibres. Besides, increasing fibre dosage until an optimum value increased the strength of the composites, as presented in Figure 4. The highest tensile strength was achieved by the NFRGC prepared with 1.5% of jute fibres (1.6 MPa) or 2.5% sisal fibres (2.3 MPa). At the same time, the highest flexural strength was achieved by the NFRGC prepared with the highest fibre percentage. For NFRGC containing jute fibres, the maximum flexural strength of 3.3 MPa was achieved with 2% fibre content. While for NFRGC prepared with sisal fibres, the highest flexural strength was obtained with the addition of 3% fibre content. Similarly, Alshaaer (2021) reported that the flexural strength of NFRGC containing jute fibres was four times higher than the strength of the geopolymer matrix without fibres. The ultimate flexural stress of the plain geopolymer was 3 MPa. However, NFRGC prepared with 15% by weight jute fibres achieved ultimate flexural stress of about 12 MPa.

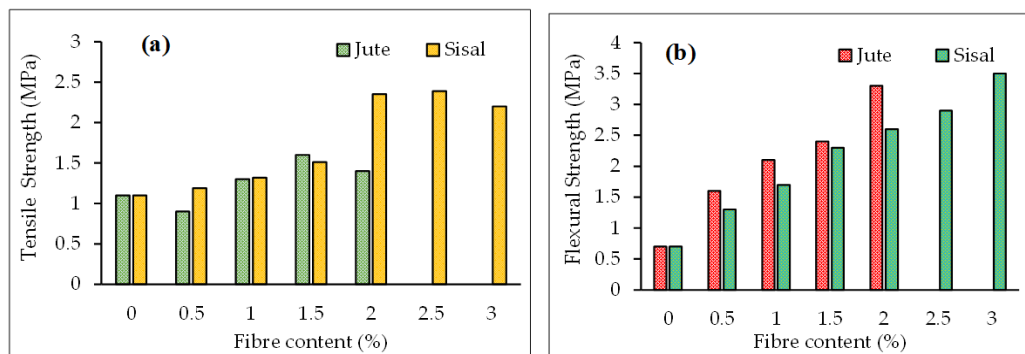


Figure 4. Strength performance of NFRGC containing jute or sisal fibres: (a) splitting tensile strength; (b) flexural strength (Silva et al., 2020)

Kavipriya et al. (2021) incorporated 0.25%–1% of sisal fibres to FA-based GPC activated by sodium silicate and sodium hydroxide solution. To reduce the self-weight of geopolymer concrete, the authors replaced the coarse aggregates with bamboo sticks at different proportions. The findings showed that the inclusion of sisal fibres was an effective solution to overcome the strength reduction due to the incorporation of bamboo sticks. Besides, increasing the dosage of the fibre from 0.25% to 0.75% increased the flexural strength of NFRGC. However, beyond 1% fibre content, a reduction in flexural strength was observed because incorporating more fibres negatively affects the geopolymerisation process, thus decreasing the strength. In another study, Abbass et al. (2021) observed that

the splitting tensile strength of NFRGC containing coconut fibres tends to increase first with the fibre content and then decrease. The gain in strength could be attributed to the ductile nature of the fibres; however, the reduction is owing to the increased porosity and low density of the fibres. The NF also can enhance the toughness of the composites and alter the failure mode from the brittle form to the ductile, as illustrated in Figure 5.

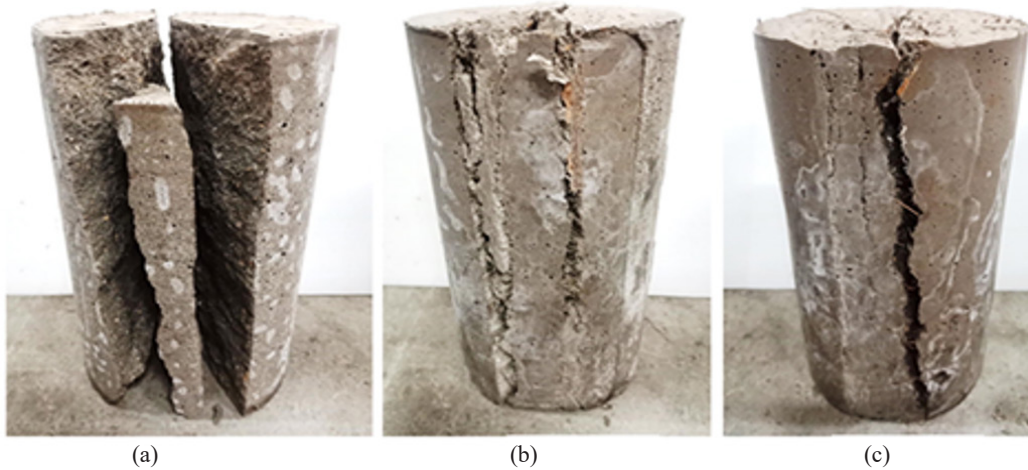


Figure 5. The failure mode of geopolymer composites; (a) plain GPC, (b) NFRGC containing 1% sisal fibre, (c) NFRGC containing 1% coconut fibre (Wongsa et al., 2020)

Assaedi et al. (2016) examined the impact of different nanoclay platelets contents on the mechanical properties of NFRGC containing flax fibres. The content of nanoclay platelets ranged from 1% to 3% by weight, while the content of flax fibre was 4.1% by weight. The results showed that NFRGC containing 2% by weight nanoclay achieved the highest flexural strength and toughness. The improvement in NFRGC strength with the inclusion of nanoclay could be related to the reduction of porosity and increased density of the composites. However, adverse mechanical properties were observed when the nanoclay content exceeded 2% by weight. In another study, Assaedi et al. (2019) observed that incorporating nano-silica particles was also effective in increasing the flexural strength of NFRGC prepared with flax fibre. After four weeks, the flexural strength of NFRGC containing nano-silica particles increased by 32.4% compared to NFRGC without nano-silica. This enhancement could be ascribed to reducing the unreacted fly ash particles, voids, and porosity, which led to a stronger bond between the geopolymer paste and the NF. However, after the ageing period of 32 weeks, all NFRGC exhibited a reduction in strength. This reduction was due to the weakening of the hemicellulose and lignin in the alkaline environment, which caused fibres brittleness (Filho et al., 2013). However, this

reduction was highly reduced for NFRGC containing nano-silica particles because nano-silica particles consumed some alkaline solutions, thus lowering the matrix's alkalinity. At the same time, the extra silica accelerated the geopolymerisation, thus increasing the amount of geopolymer gel in the composites. Consequently, the matrix's density increased, and the matrix-fibres bonding enhanced. The Scanning Electron Microscope (SEM) images of the fracture surface of the composites after 32 weeks indicated that the NF present in the NFRGC without nanoparticles had shown degradation signs and separation of the small fibrils, as obvious in Figures 6(a) and 6(b). On the other hand, NF embedded in the NFRGC containing nanoparticles did not show any notable degradation signs [Figures 6(c) & 6(d)].

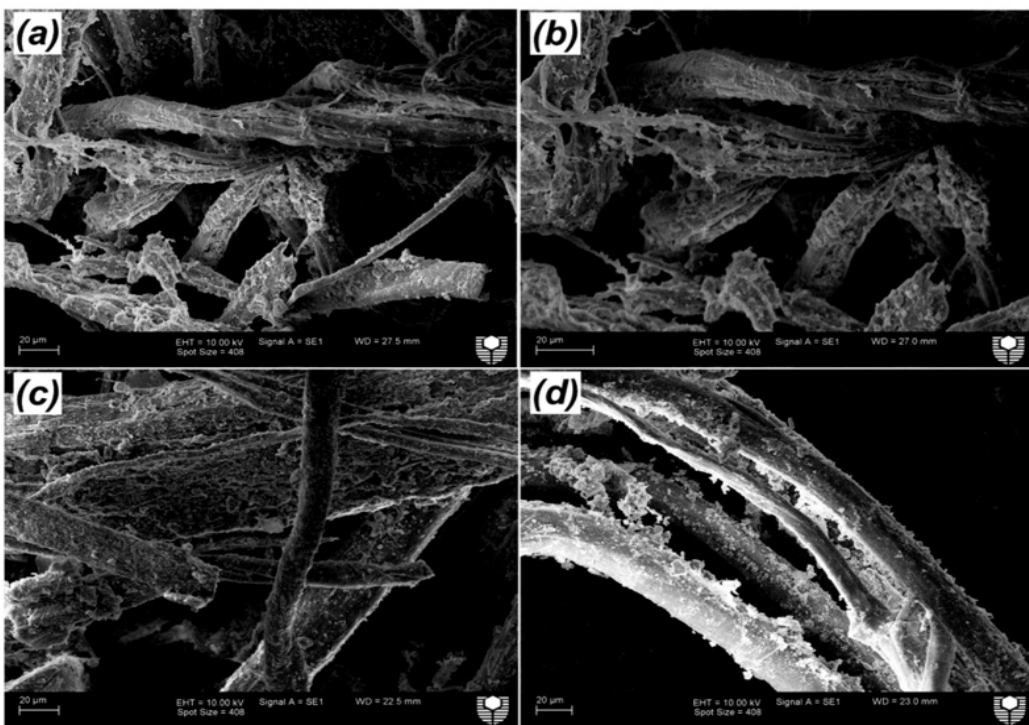


Figure 6. SEM images of flax fibre after 32 weeks in; (a) and (b) NFRGC without nano-silica particles, (c) and (d) NFRGC with nano-silica particles (Assaedi et al., 2019)

Natural Fibre Treatment and its Impact on the Mechanical Properties of NFRGC

Although NF has numerous advantages as reinforcing materials, such as low cost, lightweight, biodegradability, recyclability, and good mechanical properties, some obstacles like moisture absorption, biodegradation, and organic impurities should be addressed before the inclusion of these fibres in the geopolymer composites. The treatment or modification is usually done on natural fibres before embedment in composites. Waxes, oil, and other

undesirable components will be eliminated during the treatment process, allowing for better adhesion between natural fibres and the matrix. Many studies have revealed that suitable treatment can strengthen the interfacial bonding between the geopolymer and NF. The impact of NF pre-treatment on the mechanical characteristics of NFRGC is discussed in further depth in the following sections. The discussion will focus more on alkali-treated fibres, as this is one of the oldest and most well-known ways of modifying NF. Only a few studies have been conducted on the effects of other modifications methods.

The Impact of Treatment on the Compressive Strength

Maichin et al. (2020) studied the performance of NFRGC containing untreated or treated hemp fibres. The fibres were treated by soaking the hemp fibre in a 5 M sodium hydroxide (NaOH) solution for 48 hours before adding it to the FA-based GPC. The authors found that NFRGC achieved lower compressive strength than the plain geopolymer without fibres. There was no significant difference between the compressive strength values of NFRGC prepared with treated or untreated fibres. However, the highest deflection of about 0.64 mm was achieved by the NFRGC prepared with treated fibres compared to 0.41 mm and 0.51 mm deflection values of plain geopolymer and NFRGC prepared with untreated fibres. The low deflection of NFRGC prepared with untreated fibres could be attributed to impurities and waxes on the fibre's surface, which decreased the bonding strength between the fibre and the matrix.

Similarly, Chen et al. (2014) treated sweet sorghum fibre with a 2 M NaOH solution for 12 hours to reinforce FA-based GPC. The findings revealed that incorporating treated fibres into geopolymer slightly decreased the unconfined compressive strength. However, the post-peak toughness of the geopolymer matrix improved considerably with the addition of sweet sorghum fibres up to 2%, then declined slightly but remained much greater than that of the plain geopolymer matrix. Likewise, Pauline and Angelo (2018) treated abaca fibre with a 6% by weight NaOH solution to reinforce foamed geopolymer composite. The authors found that the compressive strength of NFRGC prepared with treated abaca fibres increased from 19.6 to 36.8 MPa. The alkali treatment improved not only the strength but also the fibre-matrix adhesion.

Zhang et al. (2021) studied the performance of NFRGC prepared with three different types of kenaf fibres: untreated kenaf fibres (KF), alkali-treated fibres (TKF) and CaCl_2 -treated kenaf fibre (TKF- CaCl_2). The fibres were incorporated at three different contents (5%, 10% and 15% by weight). The results showed that the compressive strength of NFRGC is affected by the content of the fibre, as shown in Figure 7(a). Increasing the fibre content from 5% to 15% by weight caused a reduction of about 10.7%, 6.1%, and 10.6% for NFRGC prepared with untreated, alkali-treated, and CaCl_2 -treated kenaf fibre, respectively. It is owing to the reduction in the proportion of geopolymer in the NFRGC containing higher

fibre content and increased porosity (Abbas et al., 2022). After compressive strength testing, images of destroyed specimens revealed a sudden fracture of the neat geopolymer [Figure 7(b)]. However, the NFRGC samples prepared with high TKF-CaCl₂ content retained their original shapes after the maximum load [Figures 7(c)–7(e)] due to the ability of the treated fibres to control the cracks propagation in the geopolymer composites.

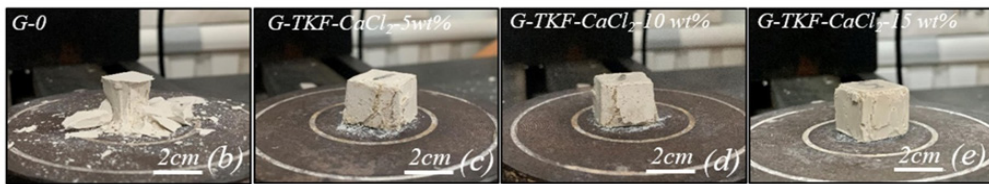
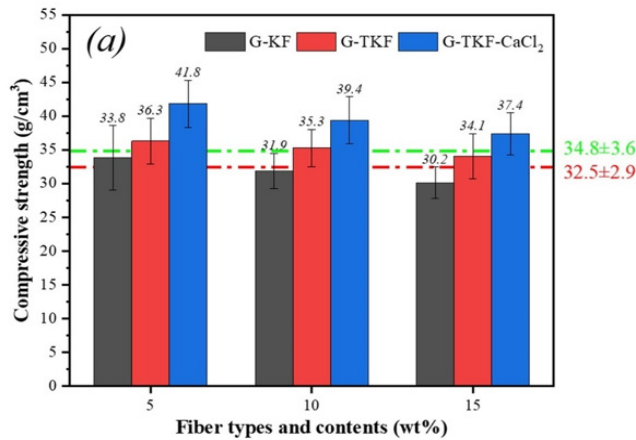


Figure 7. Compressive strength of (a) the NFRGC prepared with different types and contents of kenaf fibre; (b) failure mode of the plain geopolymer without fibres, (c) NFRGC prepared with 5% by weight TKF-CaCl₂; (d) NFRGC prepared with 10% by weight TKF-CaCl₂; and (e) NFRGC prepared with 15% by weight TKF-CaCl₂ (Zhang et al., 2021)

The Impact of Treatment on the Flexural and Splitting Tensile Strengths

Malenab et al. (2017) pre-treated abaca fibre using different types of chemical treatments: (i) alkali treatment by soaking the fibres in 6% by weight NaOH solution for 48 hours and (ii) aluminium sulphate treatment by soaking the fibres in 10% by weight of Al₂(SO₄)₃ solution. The findings revealed that treating the fibre with aluminium sulphate was efficient in forming AlOH₃ deposits on the surface of the NF, which increased the surface roughness for a better fibre-matrix interface. Then, they synthesized NFRGC by using untreated and pre-treated abaca fibre. The results showed that NFRGC prepared with treated fibres

exhibited lower compressive than that prepared with untreated fibre. However, the flexural strength was increased by 33% compared to the NFRGC with untreated fibres.

Furthermore, the SEM images indicated that NFRGC reinforced with untreated fibres displayed visible gaps between the fibre and matrix, indicating poor interfacial bonding [Figures 8(a) & 8(c)]. On the other hand, the NFRGC containing pre-treated fibres displayed narrower gaps, and the presence of the geopolymer products (zeolite particles) on the treated fibre surface referred to the strong interaction between the fibre, and the geopolymer paste [Figures 8(b) & 8(d)]. In another study, Chen et al. (2014) reported that NFRGC containing 2% alkali-treated sweet sorghum fibres exhibited higher tensile and flexural strength with about 36% to 39%, respectively, compared to the plain geopolymer.

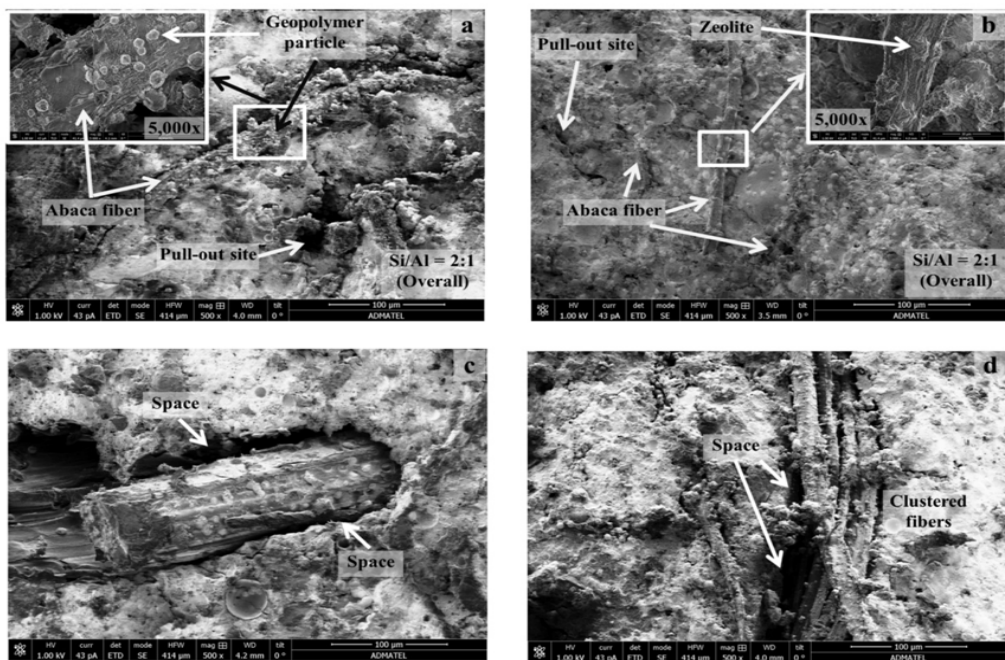


Figure 8. SEM images of NFRGC fractured surfaces: (a) NFRGC prepared with untreated abaca fibre; (b) NFRGC prepared with pre-treated fibres; (c) interface between geopolymer paste and untreated fibres and (d) interface between geopolymer paste and pre-treated abaca fibres (Malenab et al., 2017)

Researchers also investigated other modification methods. Findings by Alzeer and MacKenzie (2012) showed that the chemical modification of the wool fibre surface was very effective in reducing the lipids and fatty acids, and the treatment with formaldehyde enhanced the tensile strength and alkali resistance of the wool fibre. The flexural strength of the NFRGC containing formaldehyde-treated wool fibres was increased by 40% compared to the plain geopolymer composite. In another study, Suwan et al. (2022) reported that the

alkalinity of the GPC medium could act as an alkaline treatment reagent for the reinforced NF. The authors examined the impact of the alkaline environment in the geopolymer system on the modification process of the embedded fibres and the relationship between these factors and the properties of NFRGC containing hemp fibres. The untreated (UT) and pre-treated fibres (T) were examined in parallel to evaluate the effect of surface treatment of NF after being incorporated into the GPC mixes. The fibres were added at a content of 0.5% to FA-based GPC activated by sodium silicate and sodium hydroxide solution with different concentrations (5 M, 8 M, and 10 M). The findings indicated that the flexural strength of the NFRGC containing untreated fibres (5MUT, 8MUT, and 10MUT) appeared to be higher than that of treated fibres (5MT, 8MT and 10MT), as shown in Figure 9. It could be attributed to the unexpected decomposition of hemp fibre in the high alkalinity of the geopolymer system due to the double alkalisations processes of (a) pre-treatment process and (b) self-treatment process, respectively (Maichin et al., 2020).

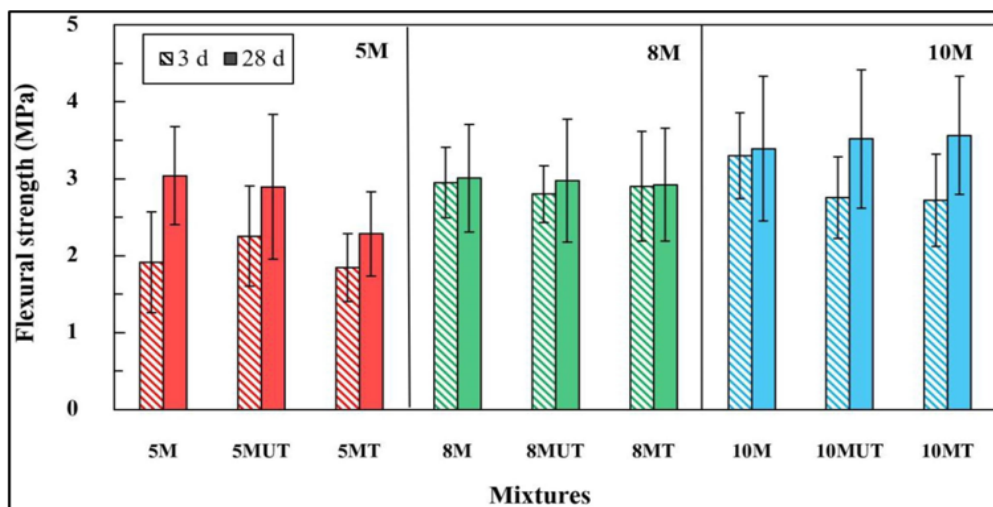


Figure 9. Flexural strength of NFRGC containing untreated (UT) and alkali-treated hemp fibres (T) in various NaOH concentrations (Suwan et al., 2022)

CONCLUSION

This paper presented a review of the published articles on the mechanical properties of NFRGC; the compressive strength, flexural and splitting tensile strengths of these composites were discussed. The following conclusions are drawn:

- NF are low-cost and ecologically sustainable alternatives to conventional fibres for improving the mechanical characteristics of geopolymer composites.

- Among NF, sisal, hemp, abaca, jute, cotton, flax, and coconut fibres are the most common fibres used for fibre-reinforced geopolymer materials due to their high specific strength and modulus.
- Literature showed that the mechanical characteristics of NFRGC are strongly influenced by the geopolymer matrix, NF type, content, and length of the fibres. However, the NFRGC containing the NF in the fabric form showed better strength improvement than the short fibres due to the better distribution of the continuous fibres in the geopolymer composites. Besides, their alignment in the direction of tension can effectively bridge the cracks, resulting in higher stress transfer at the fibre-matrix interface.
- The incorporation of nanoparticles can enhance the mechanical strength of NFRGC due to their ability to lower the geopolymer matrix's alkalinity by consuming some of the alkaline solutions, which can be used as a valuable technique to address the NF durability issues.
- The moisture absorption capability of NF, which harms the mechanical and durability characteristics of reinforced geopolymer composites, is an obstacle to their usage. However, as highlighted in this review, there are viable scenarios such as the pre-treatment or using the alkalinity of GPC medium as an alkaline treatment reagent for the reinforced NF, which can be used to minimise the water-absorbing chemical components of NF, thus improving their performance.
- In general, future studies are required to understand the processes of NF and NFRGC degradation. Furthermore, suitable and novel surface modification methods for NF should be developed to enhance their composites' moisture resistance, mechanical characteristics, and durability.

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