Present Status of Biocomposite Materials

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Since the complete replacement of fossil-based products is not feasible due to cost and performance, bio-based composite materials may have bio-based polymers, bio-based reinforcement, and fillers, or both, while having other ingredients from fossil fuel sources. As for reinforcement, nature can offer wooden and non-wooden fibers (bast, leaf, seed, core, grass, and reed) as composite reinforcements. Cellulosic non-wooden fibers offer tensile strengths ranging from 80 MPa for sisal to 938 MPa for ramie. Applications of biocomposites in structures and infrastructures have proven useful in terms of their moderate mechanical properties, lower cost, availability, biodegradability, and environmental considerations.. Hemp has been used in composite materials. However, it presents challenges; hemp fibers contain a high level of moisture, as plant-based sources are hydrophilic in nature. Its mechanical, thermal, and physical properties, such as tensile strength, wettability, flammability, and swelling, vary. These properties change with changes in plant anatomy, fiber processing conditions, growth conditions, and experimental methods.

Keywords: hemp fiber ; biofillers ; acoustic properties ; Biocarbon fillers ; Hemp composite ; Mechanical properties ; Thermal conductivity ; Polymer composite ; Sustainable material ; Green products

1. Biofillers in Biocomposite Materials

Plant-sourced materials are renewable, sustainable, and abundantly available in nature. Apart from the applications of virgin bio-based polymers (PLA, PHAs, Bio-PBS, epoxy from lignin, and epoxied oil) and biodegradable petroleum-based (PCL, PBS and PBAT) polymers, scholars are interested in composites with bio-fibers and biofillers and emphasize the importance of their physical properties; the aim is to tackle environmental challenges as well as material failures due to the high moisture content, the hydrophilicity of bio-fibers, true and artificial variability in properties such as tensile strength, wettability, flammability, and swelling due to plant anatomy, fiber-processing conditions, and growth conditions. Plant-based fillers can be in powdered form or granular form, and short fibers or continuous fibers. A schematic diagram of the hand layup technique to prepare polymer–hemp composites with biocarbon fillers is shown in **Figure 1**.

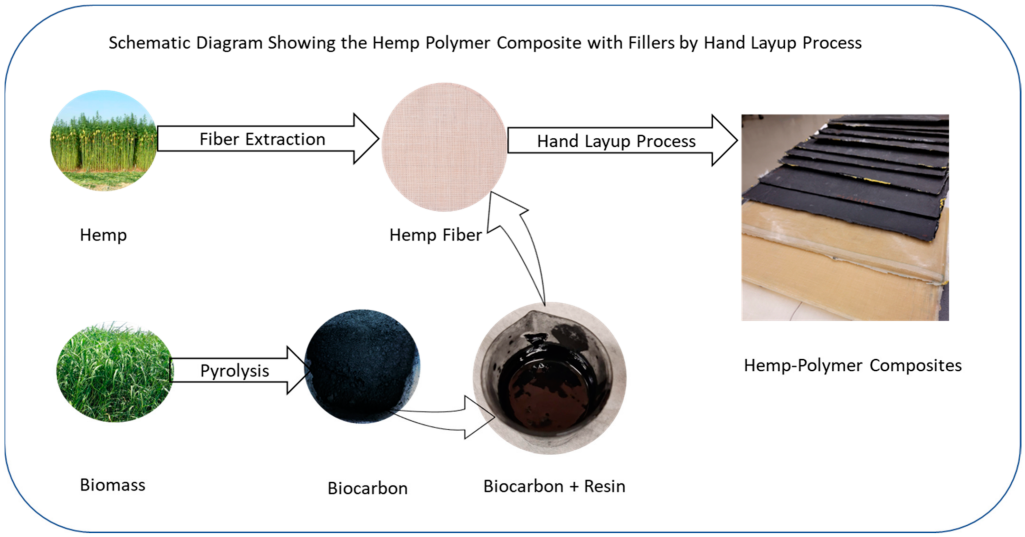

Figure 1. Showing the schematic of the preparation of hemp-polymer composites with fillers by hand layup technique.

Sun W. et al. studied the potential application of hemp-derived biocarbon in supercapacitor cells. An impressive specific capacitance of 160 F/g and an energy density of 19.8 Wh/kg at a power density of 21 kW/kg were reported [1]. Another attempt to enhance the electrical properties of an epoxy resin composite with the addition of biochar obtained from coffee waste was performed by Mauro Giorcelli and Mattio Bartoli [2]. Even though its performance was not comparable to that of the carbon-black-filled composites, it was found that the biochar from the coffee waste improved the electrical conductivity

of the polymer composite materials when compared with the composites without fillers [2]. In a similar study, Nan N. et al. [3] demonstrated that the biocarbon in polyvinyl alcohol lowers its tensile strength and storage modulus below the composite's glass transition temperature. The authors claimed that there is a potential for the replacement of carbon nanotubes and graphene as a filler in electrical applications of polymers. Organic fillers sourced from various plant (rice husk, walnut shell, coconut shell) fibers in the form of particulate fillers in bio-epoxy resin and hardener have been tested by Chandramohan D. and Presin Kumar A.J. [4]. The authors studied the mechanical properties as well as the effect of water on the mechanical strength of the composite samples. The hybrid composite samples showed a better flexural strength under wet conditions as compared to that of the dry samples. The walnut- and coconut-containing samples showed the least water absorption and a superior tensile strength (68.8 MPa), flexural strength (14.9 MPa), and shear strength (81.92 MPa). The value for the elongation at break (21.82%), the energy absorbed during the impact test (20.9 MPa), and the breaking load in the tensile and flexural tests of the composite samples with the walnut and coconut shells were also better than those of the composites with rice husk and coconut-shell fillers as well as rice husk and walnut-shell fillers. Abdellatef Y. et al. added hemp with a size of less than 6.3 mm to concrete to produce hempcrete and study its water-buffering capacity, as well as its mechanical and thermal behaviours [5]. The specific heat capacity of the hempcrete block was in the range of 1365 J/kg K to 1508 J/kg K. The results suggested its potential use for filling in walls. Thus, organic fillers in composite materials can help optimize their mechanical, electrical, thermal, and physical properties [6].

2. Role of Fiber in Resultant Material Properties

During the developmental phase, the desired properties of a composite material have been found to be dependent upon the type [7][8][9], orientation [10][11][12][13][14][15][16], size [17][18][19][20][21], and properties of the matrix as well as the reinforcement. Hemp fibers in composite materials come with different physical properties (aspect ratios), as well as mechanical, thermal, and chemical properties [22][23][24][25][26][27]. These properties are crucial, as the strength and properties of a fiber-reinforced composite depend on the alignment and size of these fibers. The fibers are aligned in the direction of the load so that a partial load is carried by the fiber which adds to the tensile strength and stiffness of the material in the principal stress direction [28][29][30][31]. This shows that the change in the fiber alignment can influence not only the strength and stiffness but also the impact toughness and shear modulus of the final material. In addition to that, the average strain sustained by the matrix and fiber in a continuous-fiber composite is similar, as they are collinear [28]. In contrast, the stress is unevenly distributed from fiber to fiber in short- and various-sized fiber composites [32][33][34][35][36]. Moreover, the stress concentration closer to the fiber ends in the composite results in an incomplete (semi-developed) stress distribution profile which weakens the composite materials due to the poor load-bearing capacity. The shear stress transfer mechanism is defined by the shear lag theory [37][38][39] which assumes the fibers are in tension and the matrix is in shear stress when a force is applied to the material. As a result, the stress rises from zero at the ends of the fiber to the maximum value at the midpoint (such as the load distribution in a simply supported beam). However, short fiber composites can have similar mechanical properties to those of continuous fiber composites if the short fibers are properly aligned, are well bonded with the matrix, and are longer than the critical length [34][40]. Another benefit of employing short fibers in composites is that they offer complex shapes through continuous or semi-continuous production processes, saving production costs and time [41][42][43][44][45]. This explains why fiber length is another important physical property of the fiber that affects the materials' properties. The properties of matrices and production techniques [46][47][48][49][50] are other factors that govern fiber selection for specific applications of composite materials. Once the material design based on the tensile behaviour of the fiber/matrix is accomplished, other factors that play significant roles in material characterization and classification include: the materials' heat resistance behaviour, production costs [51], tool wear [52][53], product density [54][55], and the compatibility between tools and equipment. For instance, the varying thermal properties of the fiber and the matrix can cause internal stresses due to thermal cycles sustained during material processing and production [56].

3. Hemp and Biocomposite Materials for Acoustic Properties

Due to their porous nature, the plant-based ingredients used in biomaterials improve the sound absorptivity by adding and changing the biofiller content in the matrices of biocomposite materials [57][58]. Jiayi Guo (2016) [59] studied hemp residue with plastic polymers with different compositions and varying sound frequencies and found that the increased amount of hemp enhances the damping ability of the resulting material and hence the sound absorption due to its high-porosity nature. There have been other sound absorption studies of bio-based products, including the following: luffa fiber was studied by Hasan Koruk, Garip Genc (2019) [60]; PLA was studied by Yao R et al. (2016) [61] and Mosanenzadeh SG et al. (2014) [62]; pinewood fiber, rice straw, and pulp were studied by DT Liu et al. (2012) [63]; wool in composites was studied by Merve Kucuk and Yasemin Korkmaz (2012) [64]; poplar wood was studied by Limin Peng et al. (2014) [65]; nano clay was studied by R. Gayathri (2013) [66]; coir was studied by Zulkifli R et al. (2008) [67]; tea leaf fiber was studied by Seçkin

Çelebi and Haluk Küçük (2012) [68]; meranti wood dust was studied by Sa'adon S and Rus AZM (2014) [69]; hemp, flax, beech, pine, and rapeseed straw were used to make lignocellulosic materials by Ewa Markiewicz, Dominik Paukszta and Sławomir Borysiak (2012) [70]; corn cobs, sunflower stems, and sheep wool were studied by Irina Oancea (2018) [71]; and hemp and kenaf fiber were studied by B. Yeşim Buyukakinci, Nihal Sokmen, and Haluk Kucuk (2011) [72]. In all these reinforcements, the matrices were plastic. From these findings, it was understood that the increase in the fiber matrix would increase the coefficient of the sound absorption. It has been reported that musical instruments made from glass fiber and from hemp perform in a similar fashion. Increasing the thickness of the natural fibers and introducing the fillers have been shown to reduce the noise reduction coefficient. Similarly, the noise reduction coefficient was found to increase with the reduction in filler sizes. These findings are derived from the summary in **Table 1**.

Hui Z. and Fan X. (2009) studied the sound-absorption properties of hemp fibrous assembly absorbers. They found that the sound absorption increased with increases in the thickness, bulk density, and air gap of the assembly as expected, while the sound absorption decreased by increasing the fiber diameter. When compared with wool, cotton, and acrylic fibers, hemp had an excellent performance due to its larger fiber diameter [73]. Buksnowitz C. et al. (2010) reinforced epoxy polymers with hemp and glass fibers to compare their sound absorptivity and found that the logarithmic acoustic damping for hemp was 0.0032 as compared to 0.0317 for glass fiber, indicating hemp composite's better sound absorption [74]. The acoustical properties of hemp-based materials were further studied by Jalil M. et al. (2014) using longitudinal, flexural free vibration, and forced vibration methods. Hemp-reinforced polyester composites demonstrated a significantly low acoustic conversion efficiency compared with that of carbon fiber polyester and glass fiber polyester composites. The acoustic conversion efficiency is a parameter used to evaluate the acoustic performance of a material based on its acoustic coefficient and sound quality factor. With such properties, hemp composites have applications in sound absorption while mineral-based composites are suitable for musical instruments [75]. Moreover, hemp shiv was solely studied by Gle P. et al. (2012) for its acoustical properties, showing a transmission loss lower than 10 dB with a material thickness of 5 cm [76] as compared to 43 dB with a 31 cm thickness when used in structures as hemp concrete bricks [77]. Furthermore, hemp concrete (two kinds of clay and hemp shiv) was examined for its acoustic performance in buildings, showing promising sound absorption and transmission loss [78]. Hemp has been gaining more attention as a building material recently. Fernea R. et al. (2019) performed another experiment to study hemp cement's acoustic properties and found that hemp fiber is a better thermal insulator than hemp shiv in all frequencies ranging between 250 and 4000 Hz. The sound absorption coefficient increased with the increase in sound frequency. With a multilayered composition, the concrete showed impressive sound absorption of up to 90% at a frequency range of 0 to 500 Hz [79].

Table 1. Biofiber-reinforced material for acoustic properties.

Ref.	Materials	Method	Findings
[59]	**Hemp crop residue + Reclaimed crushed tire CT) + LDPE/PP.**	**The impedance tube method according to ASTM E1050-12.**	**The maximum sound absorption value of 0.68 at 1650 Hz for a 80% hemp hurd, 10% coarsely crushed tire, and 10% PP composite was found. A comparison showed whole stalk hemp (WSH) composites are better at damping acoustics than the rest; this could be due to the high porosity of WSH composites. The 30% WSH, 60% CT, and 10% LLDPE composite had the highest sound-absorption coefficient (α).**
[60]	**Luffa fibers w/o surface treatment + epoxy resin.**	**Absorptivity was measured using the impedance tube as per ASTM E1050-12 and the transmission loss levels as per ASTM E2611-17.**	**The value of α changed with the change in the fiber-matrix volume fraction. α decreased when the volume fraction of resin was further increased after a specific fiber-matrix ratio. Similarly, the transmission loss increased by increasing the matrix fraction. The transmission loss value of luffa composite with a 1.5 matrix volume fraction was found to be similar to that of a cement and glass plate.**
[80]	**Pinecone char (PCC)/China Poplar char (CPC) + Epoxy resin (ER) (10, 20 30 wt.%) + Poly pox Hardener 043 + (2,4,6-tris) dimethyl amino-methyl phenol catalyst.**	**Velocity was calculated from the equation of motion, and the acoustic impedance was calculated as $Z = \rho.V_L$, in which ρ is the material density and V_L is the longitudinal wave velocity.**	**The V_L of the ER/char composites was higher than that of pure ER. The V_L of the ER/CPC composites ranged from 2754 to 2811 m/s^2, while that of the ER/PCC composites ranged from 2726 to 2798 m/s^2. The biochar increased the acoustic impedance in all composites as compared to the pure ER. The velocities showed a linear increment with an increasing biochar concentration (PCC and CPC) in the ER/BC composites up to 30% due to the increased filler density and reduced inter-atomic spacing among the fillers.**

Ref.	Materials	Method	Findings
[75]	Isophthalic unsaturated polyester resin + Methyl ethyl ketone peroxide (1 wt.%) and cobalt octoate (0.9 wt.%) + (Carbon fiber/Glass fiber/Hemp fiber).	Longitudinal and flexural free vibration tests were performed to analyze the acoustic response using the fast Fourier transform (FFT) method on MATLAB. A standard water absorption test was performed.	The glass-fiber-reinforced composites showed an acoustic performance similar to that of walnut wood. The carbon fiber-reinforced composite showed improved acoustical properties. The surrounding atmosphere's water content had a neglible effect on the quality of sound from instruments made from carbon fiber and glass fiber composites.
[63]	Pinewood fibers/Rice straw pulp + Polyurethane (PU) + Acetone/Acetic ether.	The impedance tube method was used according to ISO E10534-2. Sound frequencies from 90 to 7000 Hz were analyzed.	Void volume is a critical factor in damping sound. Wood fiber and straw fiber biocomposites have good sound-absorbing properties due to their better sound-absorbing behaviour over a wide frequency range (250 to 7000 Hz). The increase in fiber thickness lowered the value of the sound absorption coefficient (α). The compact-structure wood-fiber biocomposite offered a higher value of α compared to that of the straw fiber biocomposite of a similar thickness.
[81]	Polyethylene terephthalate (PET)/Lightweight microfibers/Blown plastic fibers/Glassfiber/blends of cotton or plastic fibers (shoddy) + Polyester/Polypropylene.	The sound absorptivity of the composites was determined according to ASTM E1050.	α was indirectly proportional to the fiber diameter, and the absorptivity increased with the increase in the specific flow resistance per unit thickness of the sample up to 1000; beyond this resistance value, α started to decrease. The tortuosity mainly influenced the location of the peaks, and the porosity and flow resistivity affected the size of the waves. Having a higher fiber surface area and a lower fiber size increases the value of α. Less-dense materials absorbed the sound of low frequencies (500 Hz), and highly dense composites absorbed waves above 2000 Hz. The air gap increased α for medium and higher frequencies. Attaching thin films such as PVC increased the α for low- and mid-frequency sounds.
[82]	Biochar + (sand/coarse aggregate + cement powder in 3:1) + water.	The sound absorption coefficient (α) was determined using a Kundt tube as per ISO 10534-2. The noise reduction coefficient (NRC) was calculated as an average value.	The higher sound energy dissipation within the interconnected pore networks in the concrete by adding biochar caused higher sound absorption coefficients. Biochar showed similar effects to that of the activated carbon. Due to the high surface area and porosity of the activated carbon, the concrete with a higher amount of biochar resembled the concrete with a lower amount of activated carbon. The noise reduction did not change with the change in the carbon filler amount in the samples whereas it substantially affected the sound absorptivity of the final material.
[64]	7:3 wool and bicomponent (polyester Core with Co-polyester Sheath polyester: 7:3 cotton and polyester. 7:3 acrylic–cotton–polyester and polypropylene. 9:1 polyester and low melt polyester. 7:3 polyester and polyamide. Polyester only. meta-aramid only.	Sound absorption was measured at frequencies between 50 Hz and 6.4 kHz according to ISO 10534-2 and ASTM 1050-98 standards.	The nonwoven composite from a cotton and polyester mixture was better than a wool and bicomponent polyester composite in terms of sound absorptivity. Adding acrylic and polypropylene fibers into the mixture improved the absorptivity of sounds with low- to mid-range frequencies. The composite with microfibers was found to perform better in sound absorption due to its low weight and high thickness.
[65]	Poplar wood fiber + Polyester fiber (PET) in 3:1 + Isocyanate adhesive (solid content), resin, foaming agent in 50:6:4.	Sound absorptivity was measured by the impedance tubes method in the frequency range of 50–6400 Hz for every 4 Hz.	The airflow resistivity of the wood fiber/polyester fiber composite up to a certain value was inversely proportional to its sound absorptivity. When the airflow resistivity was further reduced below the optimum value (1.98×10^5 Pa·s/m^2), the value of α decreased. Additionally, the value of α at low frequencies increased with the increase in the cavity width.
[67]	Coir + polyester.	The noise absorption coefficient (NAC) was measured by the reverberation room method, and the transmission loss index was measured as per ISO 717-1.	The coir fiber with a perforated panel had a higher NAC at 500 to 2500 Hz, and beyond that, the coir fiber without a board had a higher coefficient. The coir fiber as a reinforcement in polyester increased the sound absorption coefficient and transmission loss index value of the composite material.

Ref.	Materials	Method	Findings
[68]	(Polyol + isocyanate 1:1) + Tea leaf fiber waste	The sound absorptivity of the material was measured at 50 to 6300 Hz; the waves were based on a two-microphone transfer-function method according to ISO 10534-2 and ASTM E1050-98.	Soft foam was found to absorb low- and mid-to-high-range frequencies of sound better than rigid foam; the maximum absorption was found to be at higher frequency ranges. Adding tea leaf fibers into the soft foam increased the sound absorption coefficient by 50%. Adding tea leaf fibers into the rigid form improved its sound-absorbing property in all sound frequencies.
[69]	(Treated rubber/Meranti wood dust) + (Polyol + Isocyanate) polymer foam.	The sound absorption coefficient and the normal specific acoustic impedance ratios of materials as per ASTM E1050 at a frequency range of 100 to 6000 Hz were studied.	The filler loading concentration and particle size in polymer foam influenced the α. The frequency absorption level increased from 2800 Hz to 3700 Hz from light to heavy filler loading composites. The noise reduction coefficient (NRC) was inversely proportional to the wood particle size. The higher the pore size, the lower the NRC.
[70]	Isotactic polypropylene (PP) + lignocellulosic materials derived from hemp, flax, beech, pine, rapeseed straw was used as fillers.	The acoustic standing wave method was applied to determine a material's sound-absorption power at 1000, 1800, 3000, 4000, 5000, and 6300 Hz.	With a hemp filler, the value of the coefficient (α) increased rapidly up to about 25% when the frequency was increased from 3000 to 6300 Hz. For other biofillers, a higher absorptivity was observed at the frequencies of 3000 Hz to 4000 Hz. The inclusion of a biofiller in pure polypropylene increased the absorption of sound above 3000 Hz by about one-fifth.
[71]	Concrete samples: polystyrene granules, polyethylene terephthalate (PET) granules, treated corn cobs, and sunflower stems, and small balls made of sheep wool.	The acoustic absorption was calculated based on the acoustic interferometer technique (Kundt tube). The effect of the thickness was studied.	The sound-absorption coefficients of the composites were considerably higher than the values for conventional concrete. Among the 40 mm samples, the corn-cob composite was the best sound-absorbing material with a noise-reduction coefficient (NRC) of 0.193. For the 80 mm specimens, it was the PET concrete with an NRC of 0.285. The NRC increased by about 1.5 times when doubling the thickness of the sample. Additionally, density and the porosity were the other influencing factors on the sound absorptivity.
[72]	PU (Polyol + isocyanate at 1:1) + Cotton/wool/bamboo (approximately 1 mm long and weight ratios 4%, 8% and 12% for each).	The materials' sound absorptivity was measured as per ISO 10534-2 and ASTM E1050-98 standards.	PU containing 12% cotton fibers resulted in a value of almost 0.8 for sound absorptivity above 2 kHz, which is four times that of pure PU foam. A composition of 4% wool fibers in PU foam offered better sound absorptivity compared to that of virgin PU. PU foam and wool fiber mixed composites result in a maximal sound absorption in most frequencies. Cotton-fiber-blended PU foam absorbs more sound than wool fiber mixed composites. PU containing 4% bamboo fiber foam showed a sound absorptivity of 0.7, which is higher than that of pure PU in the same frequency range. PU with a bamboo fiber absorbed sound more efficiently as compared to the PU with a wool fiber composite. The sound absorptivity was directly proportional to the cotton content in the PU, and indirectly proportional to the bamboo and wool content in the PU.

4. Hemp and Biocomposite Materials for Thermal Properties

The thermal conductivity (TC) of a material can be determined by applying Fourier's law of heat conduction, which states that the rate of heat transfer through a material is proportional to the negative temperature gradient and the surface area of the material. In polymer composite laminates, heat transfer by conduction is influenced by the type of plastic used, in addition to its surface area and temperature gradient. Moreover, TC is an anisotropic property [83], which means that the value also depends upon the direction in which it is measured, as shown in **Figure 2** below. In **Figure 2**, T_1 and T_2 are two end temperatures along the x direction of the composite materials with fillers. (T_1 is higher than T_2 so that the heat flows from the left towards the right side.) K_x, K_y, and K_z are the effective thermal conductivities of the resultant material along the z, y, and z directions, respectively. The filler type, shape, orientation, and content are other factors contributing to the change in TC [84]. The TC of semi-crystalline polymers decreases with the increase in the temperature difference from room temperature up to the melting point, due to the increase in specific heat with the rise in temperature. It suddenly rises in the melting zone, reaches the maximum value, and then declines. In amorphous polymers, the thermal conductivity that changes with the temperature depends upon the glass transition temperature, showing two different TC

change patterns before the glass transition temperature (rubbery state), and after the glass transition temperature (glassy state) [85].

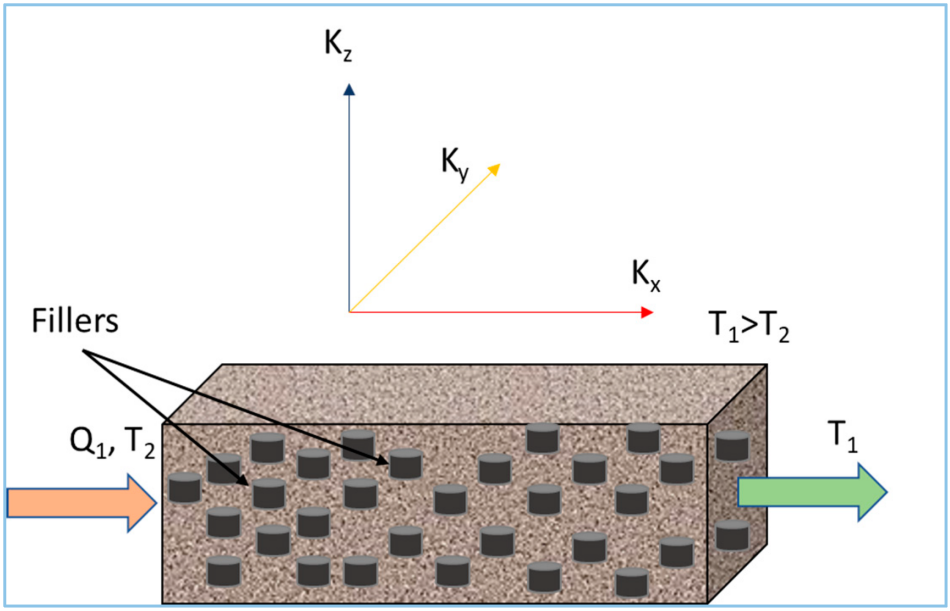

Figure 2. Showing three thermal conductivities as anisotropic properties along x, y, and z directions.

Researchers have been able to determine the thermal conductivity of a material for a long time now. Progressively, the techniques and processes to find the thermal conductivity have changed with the availability of computational tools. In one of these studies, WD Kingery (1955) [86] identified that the presence of oxides (fused silica, Al_2O_3, MgO, BeO) with a higher thermal conductivity causes the normal conductivity and temperature relationship in a material to deviate. This was explained to be the cause of the radiant heat transfer, porosity, emissivity, and electronic conductivity of the specimen. Salgado-Delgado R. et al. (2016) [87] studied the effect of sugarcane charcoal in Portland cement and found that the increased particle loading and increased particle size both reduced the material's thermal conductivity. This has been attributed to the decrease in the contact area with the increase in pore size and air volume due to the increase in the loading and particle size of the filler. Additionally, the presence of oxides in the composite materials also contributed to its reduced thermal conductivity. Pine wood in polyethylene was studied by Bourai K., Riedl B., and Rodrigue D. (2013) [88], and the results showed a compromised thermal conductivity when the wood concentration was increased. The weak bonding between the wood and polyethylene (one hydrophilic and another hydrophobic) interface, resulting in voids and gaps, contributes to this divergence in thermal conductivity (k). Mathematical models have been studied to analyze the thermal properties of the materials.

Moreover, due to their excellent heat-insulating behaviour and light weight, hemp fibers have been consumed and studied for a long time. Hempcrete is the result of using hemp in concrete to reinforce structures and buildings. Researchers have studied its mechanical and thermal properties [89], physical and structural properties [90], its microstructure and its strength [91], the impact of hysteresis and temperature on it [92], its modification to improve its water-repellent behaviour [93], its heat-conducting properties as a wall [94], and its numerical modelling [95]. Various models have been implemented to study hemp composite materials' thermal conductivity. The main matrix in hemp composites is the thermoplastic polymer.

5. Hemp Composites for Mechanical Properties

Hemp fibers have been studied for their superior mechanical properties, demonstrated by their high tensile strength and tensile modulus. Shahzad A. (2013) [96], in his study of hemp fiber's physical and mechanical properties, found out the tensile strength was 277 ± 199 MPa, modulus was 9.5 ± 5.8 GPa, and failure strain was 2.3 ± 0.8%. Pickering et al. (2007) [97] found the strengths of hemp fibers with different growing periods. The result showed that the unretted fibers with a 114-day growing period have a tensile strength above 800 MPa. The fibers' mechanical properties were greatly affected by the growing time and the selection between the retted and unretted fibers. The linearly related stress–strain curves for the hemp fibers showed a huge deviation in the tensile properties among the fibers, which is one of the challenges that need to be addressed when compared to uniform synthetic fibers, which offer consistent mechanical properties. On a positive note, these fibers can be used as reinforcements to form strong composite materials.

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