Review

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Advancement in hemp fibre polymer composites: a comprehensive review

https://doi.org/10.1515/polyeng-2022-0033
Received February 15, 2022; accepted March 9, 2022;
published online May 9, 2022

Abstract: Natural fibres as reinforcement for composite materials have witnessed a resurgence of interest in the past few years, largely due to ecological concerns, legislative directives and technological advancements. Hemp is one of the most popular natural fibres used as reinforcement in polymers owing to its superior mechanical properties. At present, hemp fibres have attracted the global interest of design engineers for developing composites having extensive applications in automobiles, electrical, construction and packaging industries. Although several literatures explore different aspects of hemp fibre reinforced composites, there is no proper literature that summarizes the surface treatment, processing techniques, mechanical performance and hybridization of hemp fibre composites. This review is envisioned to put forth a comprehensive summary of the research work published in the field of hemp fibre reinforced composites with special reference to the structure of hemp fibres, different methods for surface modification and processing techniques to fabricate the composites based on thermoplastic, thermoset and biopolymers. The paper also focuses on the effects of surface treatment on the mechanical performance of the composites.

Keywords: hemp; mechanical properties; natural fibres; polymer composite; surface treatment.

1 Introduction

Polymer composites are hybrid materials fabricated by the judicious combination of polymer matrix with different types of fibrous and non-fibrous reinforcement that exhibit synergistic properties. However, the major environmental issues associated with the colossal use of polymers and their composites are: the exploitation of rapidly diminishing nonrenewable petroleum resources, emission of significant amount of greenhouse gases into the environment and their poor biodegradability [1]. These ecological concerns and public awareness have brought about a paradigm shift towards the use of environment friendly and sustainable materials that are biodegradable. Furthermore, in response to stringent environmental policies pertaining to greenhouse gas emissions, industries have started exploring new composite materials based on bio-materials derived from renewable resources [2]. In this context, the use of natural fibres as potential reinforcement in composites has emerged as a field of substantial interest over the past few years, both in terms of their fundamental research and industrial applications.

There are a wide variety of natural fibres that are obtained from plants, animals and minerals as presented in Table 1. Amongst them, lignocellulosic fibres extracted from diversified parts of the plants such as leaves, bast fibre, seeds and straw offer unprecedented advantages over inorganic fibres [3]. These fibres are rich in cellulose, hemicelluloses, pectin, lignin and waxes, as illustrated in Table 2. Amongst them, hemp, jute, kenaf, flax and sisal are some of the fibres effectively employed as reinforcements in thermoplastic and thermoset matrices. As natural fibres are cost-effective, light-weight with relatively good mechanical properties, especially tensile and flexural properties, non-abrasive to processing equipments and CO_2 neutral, their composites are poised to replace conventional fibres such as glass fibre and carbon fibre reinforced materials in engineering, automobile, aerospace and construction applications [8]. Joshi et al. did a comparative study on the life cycle assessment of glass fibre and natural fibre reinforced composites based on three extant [9]. They found that natural fibre reinforced composites are superior to glass fibre reinforced composites due to the following reasons: (1) production of natural fibres had lower environmental impacts, (2) natural fibre composites had higher fibre content for equivalent performance, that reduced the volume and weight fraction of more polluting base polymer (3) the light-weight of natural fibre components improved

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Table 1: Classification of natural fibres.

Natural fibre	Source	Examples
Mineral	–	Wollastonite, asbestos, basalt and fibrous brucite
Animal	Wool/hair	Sheep, goat, rabbit, angora, yak and horse hair
	Silk	Tussar and mulbery
Plant	Bast	Flax, hemp, ramie, kenaf and jute
	Leaf	Pineapple, abaca, sisal, henequen, palm and agave
	Seed/fruit	Cotton, coir, milkweed and kapok
	Grass	Bamboo and bagasse

fuel efficiency and reduced emissions throughout its life cycle and (4) incineration of natural fibre composites at the end of their life cycle results in positive carbon credits. Consequently, this has resulted in a major global interest towards the development of sustainable and eco-friendly natural fibre reinforced polymer composites with economic advantages [4–6, 10].

Nevertheless, despite their potential advantages, there are some constraints in the application of natural fibre as reinforcements: low thermal stability, hydrophilic character, susceptibility to rotting, non-uniformity in their dimensions and low mechanical properties, particularly impact strength [11]. Variation in these properties largely depends on the age and geographic location of the plants. Besides this, the most important drawback of natural fibre reinforced polymer composites is the lack of proper fibre matrix adhesion at the interface that can lead to poor mechanical properties in the final product. Hence, in order to exploit full capabilities of natural fibre composites, it is essential to have good bonding at the fibre matrix interface. However, use of various physical and chemical treatment methods for surface modification of natural fibres can entail improvement in the properties of the ensuing composites.

Among the various types of vegetal fibres, hemp fibres are a promising candidate owing to their low cost of production, appealing mechanical properties and high cellulose content. Furthermore, the environmental conditions required for hemp plant growth, allows for their easy cultivation all over the world and offers additional benefit of CO_2 neutrality. As a result of these factors, the use of hemp fibres as a natural reinforcement for composite manufacture is becoming more popular and several studies have concentrated on the subject. In the literature, many articles and specialized reports are available on hemp fibres and their composites [12–14]. However, in view of the dynamically growing application-oriented research on hemp fibre reinforced composites, it is essential to have a comprehensive assessment in this area. Therefore, in order to allow a better understanding of the behaviour of hemp fibre reinforced composites, an up-to-date review in this area is essential. The overall objective of this paper is to provide a summary about the progress in the field of hemp fibre reinforced composites with an emphasis on the modification methods of hemp fibres, different techniques used for the manufacture of these composites and the effects of modification on the properties of corresponding composites.

Table 2: Chemical composition and mechanical properties of different fibres.

Fibre	Chemical composition						Physical properties			
	Cellulose (%)	Hemi cellulose (%)	Pectin (%)	Lignin (%)	Wax (%)	Micro-fibrillar angle (°)	Density (g/cm^3)	Tensile strength (MPa)	Elastic modulus (GPa)	Elongation (%)
Hemp	70–74	17.9–22.4	0.9	3.7–5.7	0.8	2–6.2	1.4–1.6	550–900	70	1.6–4.0
Jute	61–72	18–22	0.2	12–13	0.5	8	1.3–1.5	393–800	0.13–26	1.5–1.8
Kenaf	45–57	8–13	0.6	22	0.8	2–6.2	1.25–1.40	284–930	21–60	1.6
Flax	64–72	18–21	1.8–2.3	2–2.2	1.5	5–10	1.4–1.5	800–1500	60–80	1.2–1.6
Ramie	69–91	5–15	1.9	0.4–0.7	0.3	7.5	1.3–1.5	400–938	61.4–128	3–3.8
Sisal	78	10		8	2	10–22	1.3–1.4	390–450	12–41	2.3–2.5
Coir	36–43	41–45	3–4	0.15–0.25	–	30–49	1.15–1.25	220	4–6	15–40
Cotton	85–90	5.7	0–1	–			1.51	400	12	0.3–10
E-glass	–	–	–	–	–		2.5	2000–3500	70	0.5
Carbon	–	–	–	–	–		1.4	4000	230–240	1.4–1.8

Data from references [4–7].

2 Hemp fibres

Industrial hemp (*Cannabis sativa* L.) is one of the oldest plants to be spun into fibres for serviceable products, apart from flax and sisal. It belongs to bast natural fibre category and has been cultivated for more than 12,000 years specifically for the industrial production of ropes, textiles, papers, etc. Hemp plant is a native of Asia, but it quickly dispersed across Europe, Canada and Russia [15]. However, the cultivation of hemp plant has witnessed many highs and lows. During 16th to 18th century, hemp cultivation gained momentum owing to the phase of progressive civilization and industrialization. In the 19th century hemp production declined in Europe and America due to several legislative laws that prohibited the use and cultivation of hemp owing to its similarity with *Cannabis indica*, the source of marijuana. Contrarily, the psychoactive chemical Δ-9-tetrahydrocannabinol found in industrial hemp amounts to less than 0.3%, which distinguishes it from the marijuana plant. However, with the implementation of farm bill in 2014, United States has witnessed an increase in hemp production, with 25,500 acres planted in 2017 [16]. The leading producers of hemp include China, Canada, European Union, Russia and South Korea. The resurgence of interest in hemp production has been attributed to its superior fibre quality, sustainable agriculture practices and wide variety of renewable resources obtained from it. According to a report by the European Industrial Hemp Association, Europe has the world's single largest hemp production market and European cultivators produced hemp on over 80,000 acres in 2016 [16].

Fibre hemp varieties are cultivated in temperate and tropical countries but it grows best in well drained, loamy soils with high organic matter content that is rich in nitrogen [17, 18]. According to the agronomic conditions and genetic type, the plant differs widely in growth and development characteristics and can grow up to a height of 5–6 m. The hemp stem basically consists of two major parts: the outer bast or bark and the woody core. The outer bast contains three components: epidermis (20–100 μm), cortex (100–300 μm) and phloem and the inner core comprises cambium (10–50 μm) and xylem (1–5 mm) [19]. Harvested hemp is processed using various techniques in order to separate out the hemp fibre bundles.

Harvested hemp is initially subjected to retting process which is carried out either on field or in controlled laboratory conditions. During retting, the bacteria and fungi present in the water break down the chemical bonds holding the fibres together and release them from the stem. After retting, stems are dried and baled out. Hemp fibres mainly are of two types: bast or the outer long fibres associated with phloem, present along the length of the plant stem and hurds, the inner short fibres that arise from the cambium. Hemp fibres have a multi-celled structure in which elementary bast fibres are glued together with pectin or lignin and their length varies between 5 and 55 mm and thickness is about 20 μm. These fibres have a thicker cell wall and a small lumen and consist of 60–70% cellulose, 15–20% hemicelluloses, 2–4% lignin, 2–4% pectin and 1–2% wax [20]. Hemp fibres are particularly rich in crystalline cellulose micro-fibrils that are embedded in amorphous hemicellulose and lignin matrix. The elementary hemp fibre is made of primary cell wall, secondary cell wall and lumen. Secondary cell wall has three sublayers (S1, S2 and S3) and each layer has a distinct micro-fibrillar arrangement. However, the fibre properties, i.e., the fibre strength and stiffness are mainly dependent on the S2 layer as it is particularly rich in helically arranged crystalline cellulose micro-fibrils that act as reinforcement. A schematic representation of the hemp fibre structure is given in Figure 1. The orientation of micro-fibrils in hemp is close to the fibre axis, which results in a low micro-fibril angle. Hence, the Young's modulus and tensile strength of hemp fibre are comparatively high as compared to the rest of natural fibres owing to its low micro-fibrillar angle and high cellulose content as illustrated in Table 2.

Furthermore, as evidenced by the numerous studies on hemp fibre characteristics, the plant variety, stage of maturation and eco-physiological factors also contribute towards the variability of fibre structure and chemical compositions [21, 22]. The chemical composition, fibre structure and micro-fibrillar angle of the plant fibres are the most important variables that determine the overall properties of the fibres [23]. The chemical composition of natural fibres has significance as their susceptibility to degradation is dependent on individual chemical constituents. Hemicelluloses are hydrophilic in nature and responsible for biological and thermal degradation and high moisture absorption, whereas hydrophobic lignin is mainly responsible for ultraviolet (UV) and fire degradation. An investigation by Pickering et al. on the effects of growth time on the tensile properties of hemp fibres exhibited an increase in strength up to the flowering stage, with an optimum harvest time of 114 days, after which it declined [24]. This has been attributed to changes in the chemical composition of fibres during ageing of plant. This clearly signifies a strong correlation between the structure and chemical composition of hemp fibres that ultimately results in variable mechanical properties of the fibre and the corresponding composites.

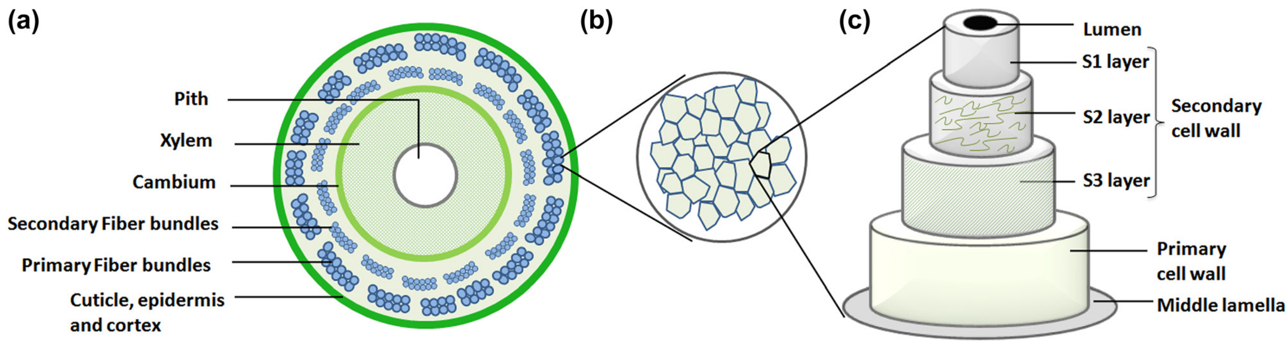

Figure 1: Hemp fibre structure:
(a) Cross section of hemp fibre stem, (b) hemp fibre bundle and (c) elementary hemp fibre.

3 Hemp fibre surface modification

The main drawback of hemp fibres in their applicability as reinforcement in hydrophobic polymer matrices is their hydrophilic character, which compromises their compatibility and ultimately limits their applicability [25]. The organization of cellulose micro-fibrils in an alternating arrangement of crystalline and amorphous zones within each parietal layer has a direct impact on the macroscopic mechanical properties of fibres [26]. The crystalline regions are more closely packed and ordered as compared to amorphous regions and hence are not easily accessible to chemical reagents. Contrarily, the hydroxyl (–OH) groups on the amorphous segments of cellulose, hemicellulose and lignin are comparatively free and readily react with other reagents [13, 27]. By virtue of this, the amorphous regions absorb atmospheric moisture that imparts a hydrophilic character to the hemp fibre surface. Besides this, under tensile loading, stress concentration points develop at the joints of moist cellulose microfibrils in the amorphous regions, initiating micro cracks that trigger ultimate fibre failure. Furthermore, hydrophilic character impedes the interfacial interaction between fibre surface and polymer matrix, resulting in fibre matrix de-bonding which reduces the mechanical properties of composites. This problem can be addressed by modification of fibre surface with suitable treatments. Surface treatment of fibres helps reduce –OH groups by substituting them with other chemical moieties and reconfigures the proportions of crystalline cellulose, hemicellulose and lignin within the fibres that directly affect the fibre strength [14, 28]. Besides this, surface modification not only prevents the reactivity of hydroxyl groups towards water, but it also improves the compatibility between hemp fibre and polymer matrix, yielding improved bulk composite properties.

In natural fibre reinforced polymer composites, the interphase and its properties are found to have a paramount influence on the interfacial bonding between fibre and matrix and the ensuing properties. The mechanism of interaction at the interphase is very complex to be elucidated as various factors need to be taken into consideration, like the thickness and properties of interface, surface tension, surface free energies, wetting phenomenon, etc. Strong interfacial interaction between hemp fibre and polymer matrix can be achieved by surface modification of fibres using two approaches. These are: (1) chemical methods and (2) physical methods. In this section, chemical and physical methods of hemp fibre surface modification will be presented in detail. A summary of the different chemical methods of hemp fibre treatment is schematically shown in Figure 2.

3.1 Chemical methods

The reagents that are generally employed for chemical modification of hemp fibre are those that contain functional groups that are capable of bonding with free hydroxyl groups on the fibre surface, thereby reducing their hydrophilic character and enhancing fibre polymer compatibility [29, 30]. Several types of chemical treatments have been reported in the literature amongst which alkaline treatment, silane treatment, maleic anhydride coupling, acetylation and graft polymerization are the most common and promising methods for fibre modification [12–14].

3.1.1 Alkaline treatment

Amongst the various chemical modification processes, alkaline treatment also known as mercerization is the most economic and widely used process in order to alter the complex cellulose structure [30–33]. In this approach,

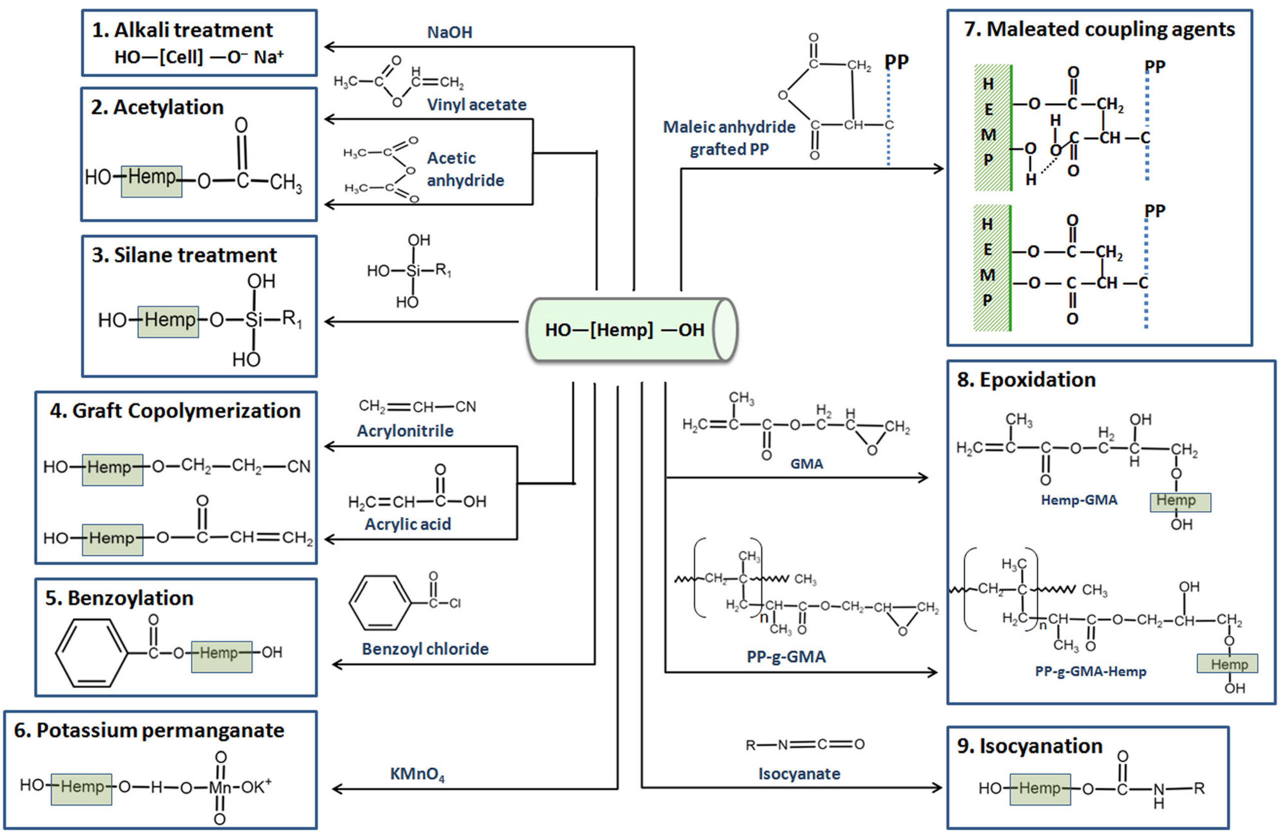

Figure 2: Summary of different chemical methods to modify hemp fibre surface.

hemp fibres are generally soaked in a 5% (w/w) sodium hydroxide solution over a period of time. NaOH reacts with the hydroxyl groups in the amorphous region and disturbs the hydrogen bonding, thereby depolymerizing the cellulose structure and exposing short length crystallites. This reduces the hydrophilic nature of fibres rendering them compatible with polymer matrices. This treatment also leads to the partial removal of non-cellulosic components: hemicelluloses, lignin and pectins along with impurities like wax and oil covering the fibre cell wall, thus providing fibres with increased surface roughness [34].

Moreover, alkaline treatment results in fibrillation of fibre bundles due to their axial splitting, thereby decreasing the fibre diameter and increasing the aspect ratio [35]. Consequently, the effective fibre surface area increases, and this coupled with the development of rough fibre texture provides additional sites for mechanical interlocking that enables better fibre matrix adhesion. In addition, partial removal of hemicelluloses due to alkaline treatment leads to a decrease in micro-fibrillar angle and an increase in molecular orientation, which facilitates better packing of cellulose chains with increased crystallinity [36].

Alkaline treatment of hemp fibres has been shown to enhance their mechanical properties. However, its optimization in terms of NaOH concentration, time and temperature is essential for their maximum efficiency. Variation of NaOH solution concentration for hemp fibre treatment in the range of 0.8%, 2%, 4%, 6% and 8%, respectively, exhibited a maximum tensile strength of 1064 MPa at 6% NaOH concentration and Young's Modulus reached a maximum value of 65 GPa at NaOH concentration of 4% compared to the tensile strength and Young's modulus of 591 MPa and 38 GPa, respectively, of untreated fibres [37]. Another study on the effects of various concentrations of alkali solutions has demonstrated that mechanical properties of hemp fibres are governed by the concentration of NaOH solution as it modifies the fibre structure and they are also dependent on the amount of cellulose and crystallinity index of individual hemp fibre bundles. Similarly, hemp fibre quality with respect to their fineness and flexibility can be improved by varying the temperature and duration of alkali treatment. Hemp fibre modification with 5% and 18% NaOH solution at room temperature and boiling point for different durations yielded finer fibres with higher flexibility and lower water retention values due to changes in their chemical composition [38]. The tensile properties increased in some cases but dropped drastically for 18% NaOH treated samples. This has been attributed to the removal of hemicelluloses along with swelling and shrinkage of individual cells that result in disorientation of fibrils. Hence, a high alkali concentration can cause excessive delignification, which may result in damaged fibres that are unable to support effective stress transfer between fibrillar networks.

3.1.2 Acetylation

Acetylation of natural fibres is basically carried out to reduce their hygroscopic nature, improve dimensional stability and prevent environmental degradation. It is the most commonly used esterification method wherein the hydroxyl groups on the fibre surface are substituted with the acetyl groups by reacting with acetic anhydride, maleic anhydride or vinyl acetate at an elevated temperatures with or without catalyst (e.g. potassium carbonate, acids, pyridine) [29, 39, 40]. Acetic acid or maleic acid is generally not preferred as they do not react sufficiently and give poor esterification yields.

Basically, the hydroxyl groups on hemicelluloses and lignin in the amorphous region give the esterification reaction, whereas the hydroxyl groups of cellulose, being closely packed owing to hydrogen binding prevents diffusion of reagents in the crystalline region and results in a low extent of esterification. However, the extent of acetylation has been found to be dependent on the ratio of hemicelluloses and lignin content in the fibres as reported by Tserki et al. [41]. In general, acetylation alters the surface morphology of the hemp fibres by the removal of hydrophilic hydroxyl groups from the non-cellulosic components and slightly affects the crystallinity index. This leads to better adhesion between fibre and matrix that enhances the interfacial bonding, resulting in improved mechanical properties of the composites (tensile strength, flexural strength and compressive strength) [42]. It has also been reported, that acetylation improves the thermal stability and water resistance of the fibres and corresponding composites.

3.1.3 Silane treatment

The incompatibility between hydrophilic natural fibres and hydrophobic polymer matrix can be considerably reduced by the use of coupling agents like silanes which are capable of forming covalent bonds between fibre and matrix. Basically, silanes are multifunctional molecules represented as: $R_{(4-n)}–Si–(R^1X)_n$, where $n = 1$ or 2, R represents an alkoxy group, X is an organofunctionality and R1 is an alkyl moiety [43]. Surface treatment of natural fibres with silane employs its dilute solution, which results in the formation of silanols after its hydrolysis. Silanol further undergoes condensation reaction, wherein one end reacts with the hydroxyl groups on fibre surface yielding cellulose–O–Si– bond, and other end reacts with the functional moieties on the matrix, forming a siloxane bridge between the two [44]. Silane coupling agents can penetrate the pores on the fibre surface to develop mechanically interlocked coatings that provide molecular continuity across the composite interface. This results in improved fibre matrix adhesion that significantly enhances the stress transfer between fibre and matrix and helps stabilize the mechanical properties of composites during loading.

The silane coupling agents which are most commonly used for hemp fibre treatment are trialkoxysilanes with organofunctionality like amino, mercapto, glycidoxy, vinyl, or methacryloxy groups, e.g., γ-methacryloxypropyltrimethoxysilane (MPS), γ-aminopropyltriethoxysilane (APS), γ-glycidoxypropyltrimethoxysilane (GPS) and 3-glycidyloxy propyltrimethoxysilane [43, 45–50]. However, the proper choice of organofunctionality in silanes for hemp fibre treatment can significantly contribute towards composite performance as evidenced in many studies. An investigation by Rachini et al. on the effectiveness of MPS and APS for hemp fibre modification demonstrated that the grafting quantity is dependent on the chemical structure of organosilane and it increases proportionally with the concentration of silane solution [51]. The high degree of grafting in the case of APS has been attributed to the formation of hydrogen bonds between the polar amino end group of APS and the unreacted hydroxyl moieties on hemp fibres.

A study by Panaitescu et al. on the surface and thermal properties of polypropylene (PP) hemp fibres composites employed APS, GPS, MPS, and potassium permanganate (KP) for surface modification of hemp fibres [52]. They demonstrated that all the treatments led to the splitting of fibre bundles and separation of elementary fibres with MPS treatment being the most effective among the various silanes with reference to the thermal stability and mechanical properties of the corresponding composites. In yet another study, Nishitani et al. investigated the effects of three different silane coupling agents, namely 3-(2-aminoethylamino)propyltrimethoxy silane, 3-glycidoxypropyltrimethoxy silane, and 3-ureidopropyltrimethoxy silane on the tribological properties of alkali treated hemp fibre reinforced biomass derived polyamide composites [53]. They observed that the tribological performance of the composites improved after the surface treatment of hemp fibres by silane coupling agents, with ureidosilane being the most effective. This improvement in the tribological performance has been attributed to enhanced interaction and interphase adhesion between the hemp fibres and polymer matrix subsequent to treatment, that modifies the friction and wear mechanism.

Similarly, a comparative study analyzed the influence of two different chemical treatments, namely alkali treatment and 3-glycidyloxypropyl trimethoxysilane treatment on the mechanical properties of woven hemp fibre reinforced epoxy composites [49]. It demonstrated that the tensile and flexural properties of silane treated fibre composites are considerably better than those of alkali treated fibre composites.

3.1.4 Graft copolymerization

Another technique frequently employed for effectively transforming the surface properties of natural fibres is graft copolymerization using vinyl monomers such as methyl methacrylate and acrylonitrile [54–56]. The aim of this treatment is to generate covalent bonding across the natural fibre and polymer matrix interface, thereby improving the compatibility between the two. Graft copolymerization also increases the surface energy of natural fibres resulting in improved wettability and enhanced interfacial interaction [57].

Grafting generally takes place on the hydroxyl groups at C2, C3 and C6 carbon atoms as well as the C–H bonds on the cellulose molecules [58]. Various chemical and physical methods of grafting have been used, like ionic grafting, free radical induced grafting, microwave grafting, plasma radiation induced grafting, enzymatic grafting, etc. However, in order to achieve an optimum grafting percentage proper selection of reaction parameters such as initiator, solvent, monomer concentration, reaction time and temperature is essential, as evidenced by Singha and Rana in their study on the kinetics of graft copolymerization of acrylic acid on hemp fibres [59]. Similarly, it has been observed that free radical induced grafting is preferred over ionic grafting as the later requires anhydrous and/or alkaline medium, which gives low molecular weight copolymers, whereas free radical induced grafting yields high molecular weight graft copolymers [60]. Likewise, in case of the redox initiation systems, $CuSO_4$–$NaIO_4$ (Cu^{2+}–IO_4^{-}) system is of particular importance as it does not require acid during graft copolymerization reaction, which is an added advantage given the degradation of fibres in acidic conditions [61].

On the other hand, it has been shown that microwave radiation technique for graft copolymerization is the best method as compared to conventional techniques in terms of cost effectiveness, time consumption and the extent of physicochemical stresses to which the fibres are exposed [62]. Sunn hemp fibres chemically modified with ethyl acrylate and binary monomers methyl methacrylate and acrylonitrile through microwave induced graft copolymerization were used to reinforce polyhydroxybutyrate resin [63]. In this study, it has been demonstrated that graft polymerization renders the surface of sunn hemp fibres rough and amorphous and increases their thermal stability. It also improves the interface and stress transfer between the fibre and the matrix, which significantly enhances the mechanical properties of composites.

3.1.5 Benzoylation

Hydrophilic character of hemp fibres can be significantly reduced by benzoylation treatment. The treatment employs benzoyl chloride for fibre treatment, which leads to the inclusion of aromatic moieties on the fibre surface, rendering them hydrophobic in nature and improving the fibre matrix interfacial interaction. This treatment also increases the moisture resistance thereby improving the thermal stability and overall performance of composites. Alkaline pretreatment is typically employed prior to benzoylation in order to remove the lignin, oils and waxes and expose the reactive hydroxyl groups on fibre surface [64, 65].

3.1.6 Peroxide treatment

Peroxide treatment of hemp fibres has attracted the attention of researchers due to its potential to improve the adhesion between fibre and polymer matrix and its easy processability. The process involves the treatment of alkali pretreated hemp fibres with dicumyl peroxide or benzoyl peroxide in acetone solution at high temperatures. Decomposition of peroxide yields free radicals ($RO^{\bullet}$), that can react with hydroxyl groups on fibre and hydrogen groups of polymer leading to the formation of an interphase with strong interfacial adhesion [66]. This treatment thus improves the mechanical properties of composites and also reduces the moisture absorption tendency of hemp fibres.

3.1.7 Maleated coupling agents

Surface modification of hemp fibres by reacting them with maleic anhydride has been proposed as an effective method to enhance the adhesion between fibre and matrix by means of chemical bonding. Maleic anhydride acts as a coupling/compatibilizing agent as it not only undergoes an esterification reaction with the hydroxyl groups in the amorphous region of the fibre but it can also react with the polymer matrix usually polyolefins, thus contributing to the formation of covalent bonds across the interface. At the same time, maleic anhydride can form hydrogen bonds with the hydroxyl groups on the fibre surface. The treatment also diminishes the hydrophilic character of the hemp fibres and decreases their water absorption tendency, along with an improvement in mechanical properties like Young's modulus, hardness, flexural modulus and impact strength of corresponding composites, as reported by Mishra et al. [67, 68].

A similar mechanism is applicable when maleated polyolefins like maleic anhydride grafted polyethylene or maleic anhydride grafted polypropylene (MAPP) are used as coupling agents for hemp fibre surface treatment. The mechanism involves the activation of maleated copolymer by heating at 170°C, so as to regenerate anhydride moiety which further gives esterification reaction with the hydroxyl groups of natural fibres [69]. This treatment increases the surface energy of natural fibres to a level comparable to that of polymer matrix resulting in better wettability of the fibres with enhanced interfacial interaction at the fibre matrix interface [28, 70]. However, the manufacturing process causes variation in the molecular weight and acid content of MAPP, which can significantly influence the chemical composition and interfacial properties. In general, long chain MAPP with many MA grafts is favorable for producing covalent bonds at the interface, while high molecular weight MAPP can get entangled with the polymer matrix, creating difficulty for anhydride groups to react with the surface hydroxyl groups of fibres [71].

In order to determine the extent of improvement MAPP involves, Mutje et al. used an interesting approach in which hemp fibres were chemically modified with octadecyltrimethoxysilane which reduced the number of –OH groups on the hemp fibre surface. They compared the PP composites based on untreated hemp fibre and 4 wt% MAPP with PP composites containing silane treated hemp fibres and 4 wt% MAPP and observed that the tensile strength and flexural strength for the formulations based on non-silanized hemp fibres are quite high as compared to formulations based on silanized hemp fibres. They attributed this decrease in mechanical properties of composites in case of silanized fibres to the low number of accessible –OH groups on the fibre surface due to their blocking by silane coupling agent that prevents the formation of ester bonds between hemp fibre and MAPP. This approach thus demonstrated that using hemp fibres without any prior chemical treatment in combination with MAPP improves the overall mechanical properties of composites due to the covalent interaction between the surface hydroxyl groups of hemp fibres and MAPP moieties improving the compatibility at the interface [72].

Pickering et al. in their study on optimization of industrial hemp fibres for composite fabrication reported that PP composites containing 40 wt% of optimally alkali treated hemp fibres and 3 wt% MAPP exhibited highest increase in tensile strength and Young's modulus by 107% and 356%, respectively, as compared to pure PP, owing to an improvement in fibre matrix interfacial bonding [24]. A similar approach was used by Sullins et al. in their study on PP based composites reinforced with chemically modified hemp fibres. They investigated the mechanical behaviour of composites by employing various combinations of surface treatments for hemp fibre that included NaOH treatment (0, 5, and 10 wt% NaOH) coupled with the addition of 5 wt% MAPP to the PP matrix. The use of MAPP increased the wettability of fibres, improving the interfacial adhesion between hemp fibres and PP matrix, with a direct effect on the tensile and flexural properties of composites. The composites with 5 wt% MAPP addition exhibited best mechanical properties, wherein the flexural strength and modulus and tensile strength and modulus increased by 37%, 37%, 68% and 213%, respectively, as compared to neat PP when the fibre loading was 15 wt% and in the case of 30 wt% fibre loading, the corresponding increase was 91%, 132%, 122% and 297% [73].

3.1.8 Sodium chlorite treatment

Sodium chlorite ($NaClO_2$) treatment is generally used for bleaching of natural fibres and it is carried out in an acidic medium which results in liberation of choleric acid ($HClO_2$), which further undergoes oxidation and forms chlorine dioxide (ClO_2). In case of hemp fibres, ClO_2 is mainly responsible for lignin removal as reported by Kostic et al. and it enhances the hydrophobic nature of the fibre [74]. Treatment of hemp fibres by $NaClO_2$ with or without the surface treatment by 3-ureidopropyltrimethoxy silane coupling agent has also been reported to improve the mechanical and tribological properties of plant-derived polyamide 1010 composites, which has been attributed to improved interfacial adhesion between fibres and matrix [75].

3.1.9 Heterofunctional monomers

3.1.9.1 Epoxides

Another way to enhance the interfacial compatibility in composites is through the use of heterofunctional monomers like epoxides, especially in case of polyolefin based composites. Epoxide such as glycidyl methacrylate (GMA), which is a bifunctional monomer, has been reported to form strong ether bonds with the hydroxyl groups of cellulose in hemp fibres that have been pretreated by alkali prior to chemical modification [29]. The chemical modification of hemp fibres using GMA, an unsaturated polar molecule was achieved by employing two approaches: the first involved the reaction between the hydroxyl groups of hemp and the epoxide ring of GMA, resulting in the formation of a stable ether bond at the fibre surface. Subsequently, the modified hemp fibres reacted with the PP matrix during melt mixing through the double bond of methacrylate in the presence of a radical initiator. In the second approach, radical grafting of GMA was done initially on the PP in the presence of a radical initiator followed by the reaction of the epoxide ring of GMA with the OH groups of fibres during melt mixing. The torque and weight percentage gain exhibited an increasing trend with reaction time in case of hemp fibres which were first subjected to etherification with GMA as compared to the other approach owing to the degradation phenomenon of PP during radical grafting with GMA [76]. The same study also assessed the effects of different compatibilizers such as styrene-(ethylene-co-butene) block copolymer (SEBS), PP-g-GMA and SEBS-g-GMA on the fibre matrix interfacial interaction and the ensuing thermal, rheological, morphological and mechanical properties of composites. In general, all the modified composites exhibited better fibre dispersion and higher interfacial adhesion as compared to unmodified composites, with the effect being more marked in the case of composites containing GMA modified hemp fibres. This improvement has been attributed to possible grafting reaction between fibre and PP in the presence of GMA. Chemical modification of fibres also improved the thermal stability and affected the crystallization behaviour of corresponding composites. Apart from that, an increase in tensile modulus, increased stiffness and lower elongation at break were displayed by all the composites. Pracella et al. obtained similar results in their study for different polymer matrices: PP, polystyrene (PS) and poly(ethylene-vinyl acetate), which employed different bifunctional monomers (GMA and MA) along with various compatibilizers (PP-g-GMA, SEBS-g-MA, PS-co-MA) in order to improve fibre matrix interfacial interaction [77].

3.1.9.2 Isocyanates

Isocyanates are another example of heterofunctional monomers (R–N=C=O) that can be used as coupling agents, where R represents a variety of organic groups. Their coupling ability is attributed to the isocyanate moiety (–N=C=O) which can react with the hydroxyl groups on the hemp fibre surface, forming a urethane bond that can enhance the interfacial compatibility and the corresponding strength of the composites. A study by Chen et al. investigated a novel isocyanate coupling agent, isocyanatoethyl methacrylate (IEM) for the treatment of hemp fibres in unsaturated polyester (UP) composites [78]. IEM is a heterofunctional monomer with two functions that can independently react with other vinyl monomers and active hydroxyl groups on hemp fibres. Hemp fibres were treated with different concentrations of IEM (1, 3, 5 and 7 wt%) and the resulting composites based on UP exhibited a significant enhancement in the tensile strength, flexural strength, flexural modulus and water resistance which has been attributed to the covalent bonding of the isocyanate group of IEM with hydroxyl groups on hemp fibres and covalent linkages between the vinyl group of IEM treated fibre and UP via styrene unit.

3.1.10 Permanganate treatment

Potassium permanganate ($KMnO_4$) treatment of hemp fibres offers a cheap and effective method for direct application at the industrial level. An interesting study by Panaitescu et al. employed various silanes and KP (0.05 wt %) solution in acetone for surface modification of hemp fibres [52]. They evaluated the influence of hemp fibres with modified surface on the properties of PP composites. It was noted that both the treatments are efficient in splitting of small fibre bundles and the separation of elementary fibres. The treatments also influenced the micro- and nano-mechanical properties of composites, with the KP treatment increasing the modulus of elasticity by 69% in case of 40 wt% hemp fibre reinforced composites. Similarly, KP treatment of hemp fibres increased the hardness of the corresponding composites as revealed by nanoindentation results, which has been attributed to the decomposition of unreacted KP on the fibre surface during melt processing, releasing manganese dioxide that is responsible for locally increasing the stiffness and hardness of the matrix. However, KP treatment slightly decreased the thermal stability of composites owing to the mild oxidation of hemp fibres. It has been reported that permanganate treatment decreases the thermal stability of hemp fibres due to a decrease in the crystallinity and disturbance of crystalline structure of hemp fibres [79].

In a similar study, Moonart and Utara accessed the effects of two different surface treatments, silane and permanganate on the curing characteristics, dynamic mechanical properties, morphological and thermal properties of alkali pretreated hemp fibre filled natural rubber composites [80]. However, they reported that silane treatment is more effective than permanganate treatment in improving the compatibility between hemp fibre and natural rubber and over all properties of the composites. This is probably due to the high moisture absorption properties of OH and Mn–O groups on the surface of permanganate treated hemp fibre, which causes poor compatibility between hemp fibre and the rubber matrix.

3.2 Physical treatments

Physical treatment of hemp fibres is another approach that can be employed with the aim to (1) separate the individual filaments from fibre bundles and (2) alter the surface properties of fibres thereby enhancing the compatibility with polymer matrix [81]. In general, the interface between fibres and polymers is influenced by physical modification by way of changing only the structure and surface properties of fibres thereby enhancing the mechanical bonding between fibre and matrix [82]. Physical treatments such as fibre beating, heat treatment, calendaring, steam explosion and microwave irradiation are known to separate individual fibres without extensively changing their chemical composition. On the other hand, physico-chemical treatment methods like corona discharge treatment, plasma treatment, dielectric barrier discharge (DBD) and UV treatment are employed for achieving surface modification of fibres.

Plasma treatment and corona discharge treatment are known to alter the surface energies of natural fibres thereby improving the compatibility between the fibre and matrix [81]. However, a basic difference between the two is that plasma treatment is potential free and the gas type, its flow and pressure can be controlled, whereas corona treatment uses a high voltage and atmospheric pressure to generate the plasma. Modification of hemp fibres can be achieved by selectively employing low temperature atmospheric pressure plasma treatment and the experimental results exhibited that hemp fibres processed in this way had increased wettability and dyeability [83, 84]. Study on the effect of atmospheric pressure plasma treatment of hemp fibres on the interfacial behaviour showed that the apparent interfacial shear strength increased for brief treatments of 1 min, which was attributed to the addition of functional groups, the removal of contaminating compounds that obstruct the adhesive process and increased surface roughness [85]. As a result, mechanical interlocking of the modified fibres with the polymeric matrix was enhanced. However, tensile strength, Young's modulus, and elongation at break of the hemp fibres decreased significantly for prolonged treatment. It was inferred that the heat produced during the plasma treatment dehydrates the fibres and the etching impact of the plasma damages their micro-fibril structure, which is responsible for deterioration of mechanical properties.

A study on PP composites that utilized corona modified hemp fibres exhibited that the treatment leads to significant increase in the mechanical properties (tensile strength and Young's modulus) with the composite sample's Young's modulus value increasing by 30% [86]. It was concluded that the etching effect generated by corona treatment, as confirmed by microscopy was responsible for the improved mechanical anchorage and interfacial adhesion between the matrix and fibres that enhanced the mechanical properties.

Another modification of the plasma technique is the DBD technique, which operates at atmospheric pressure and is characterized by the presence of a dielectric barrier in the form of glass between the two electrodes that restricts the direct flow of current. This treatment is known to change its surface chemistry and also increase its surface roughness as a consequence of intensive surface etching that increased the wettability of raw hemp fibres by five times, as reported by Pejic et al. [87]. Studies on the modification of hemp fibres by DBD technique have shown that the treatment alters the fibre surface by increasing the number of hydrophilic function groups on the fibre surface thereby improving wettability and adhesive properties without changing its physico-mechanical properties [88].

Another interesting study by Olaru et al. investigated the structural changes in hemp fibres subjected to hydrothermal treatment followed by exposure to UV radiation or gamma rays [89]. The study demonstrated that the combined effect of treatments leads to various structural changes ranging from cellulose organization by way of reorganization of hydrogen bonds and through formation of some macro-radicals in the cellulose chains at the supramolecular level, leading to a change in crystallinity due to degradation of cellulose structures in the hemp fibres depending on time and dose of radiation. This affects the overall physical and mechanical properties of the fibres.

Despite the progress in various physico-chemical treatments of hemp fibres, there are many issues that need to be addressed. Control over process parameters and optimization studies are required to better understand the behaviour of hemp fibres when subjected to such treatments and to improve their interfacial properties, thereby enhancing their compatibility with polymer matrices.

4 Processing techniques

The fabrication technique of natural fibre reinforced composites is one of the primary aspects that affect the overall performance of composites. The selection of fabrication method for natural fibre reinforced composites is based on different criteria, including the characteristics of raw materials, complexity of the design part, manufacturing cost as well as the properties of the final product. In general, the ideal manufacturing process should produce a composite with minimal defects.

The processing of hemp fibre reinforced composites is significantly influenced by some factors such as: fibre volume fraction, fibre size, moisture content of fibre and type of polymer matrix. Apart from this, the thermal degradation of hemp fibres above 200°C poses a challenge to their processing using traditional manufacturing techniques as they are primarily designed for synthetic fibre reinforced composites [47]. Therefore, pretreatment of hemp fibres is an integral part of processing hemp fibre reinforced composites. The processing methods commonly employed for hemp fibre reinforced composites include injection molding, extrusion, compression molding and resin transfer molding (RTM) technique depending upon type of polymer matrix.

4.1 Processing of thermoplastic composites

4.1.1 Injection molding

Injection molding is a commonly used approach for the industrial production of short natural fibre composites based on thermoplastics with complex geometries. In this process, polymer granules and short natural fibres are fed into a hopper, melted and then transported towards mold or die by twin screws. However, the process limits the amount of natural fibres that can be compounded with polymer due to an increase in the viscosity of mixture and the narrow gate and opening of the mold [90].

The dispersion of natural fibres in polymer matrix can be enhanced by increasing the screw speed (from 100 to 300 rpm) as demonstrated by Gunning et al. in their study on polyhydroxybutyrate composites based on hemp, flax and Lyocell fibres [91]. However, it was also established that increasing screw speed increases the barrel temperature due to shear heat that results in the thermal degradation of composites, especially in case of composites containing hemp fibres. Similar observations are reported by Benhadou et al. for hemp-PP composites prepared by industrial injection molding process [92]. The study evidenced that increasing the screw speed rotation above 65 rpm decreases the viscosity but causes a rise in temperature with degradation of composites above the screw speed of 125 rpm. Besides this, the high shear rates in this process result in severe fibre attrition and fibre length variation that affects the critical fibre length. Reduction of the fibre length below the critical fibre length can degrade the overall performance of the final product as the short fibres cannot bear the load for which the composites are designed. In an interesting comparative study, injection molded and extrusion–injection molded hemp-PLA composites were investigated by Siva et al. for their mechanical properties and water absorption properties [93]. The results of the study revealed that the extrusion–injection molded composite possesses higher mechanical strength and better bonding with matrix than the injection molded composite.

4.1.2 Extrusion

Extrusion is a hot-melt process extensively used in the polymer industry for continuous processing of chopped or short natural fibre composites. In this process, polymer granules and fibres are fed into a heated extruder and the molten mass is homogeneously mixed with the screw. The molten mass is used to produce composites of required shapes. Extrusion allows optimizing various process parameters like temperature, screw speed and residence time that directly influence the mechanical properties of the resulting composites. Generally, twin screw extruders are employed for homogeneous dispersion and wetting of the natural fibres. However, a major concern of this process is the breakage of fibres bundles and shortening of fibres that modifies the aspect ratio of fibres thus impacting the overall properties of the finished product [94].

Most of the hemp fibre reinforced PP and PE composites are processed by this method. Khoathane et al. studied PP composites produced by extrusion that were compatibilized with 1-pentene and reinforced with 0.8 mm hemp fibres, at different contents between 5 and 30 wt% [95]. They reported that an increase in fibre content increased the tensile strength, elastic modulus, and flexural strength but decreased the impact strength of composites.

Panaitescu et al. studied the effects of hemp fibres of different initial lengths (1, 2.5, and 4 mm) on the thermal and mechanical properties of PP/poly(styrene-b-(ethylene-co-butylene)-b-styrene) (SEBS) composites fabricated by extrusion and injection molding process [96]. The study demonstrated a reduction in the average length of hemp fibres from 1.10 to 0.57 mm, from 2.53 to 0.73 mm and from 4.19 to 1.03 mm which is about 2, 2.5, or 4 times as compared to the initial fibre length on extrusion and injection molding. This has been attributed to the presence of SEBS and extreme processing conditions that increased the melt viscosity and shear forces, resulting in defibrillation of hemp fibres.

4.1.3 Compression molding

Compression molding is one of the standard and versatile techniques for fabrication of natural fibre composites as it can be used for all forms of fibres including randomly oriented fibre mats and woven textiles. In this technique, the raw material is placed in the mold cavity and preheated, followed by compression with the core side of mold at certain pressure to allow the resin to impregnate into the fibres [97]. While maintaining the pressure, the mold is subsequently cooled until the composite solidifies and is later de-molded. In this process, the important process parameters that influence the performance of composites are temperature, pressure and heating and compression duration [98, 99]. According to a study on the effect of compression molding conditions on the tensile properties of woven hemp fibre reinforced PP composites, these composites had a desirable molding temperature of 180°C and a molding time of 20 min [100]. It was also demonstrated, that for optimized molding conditions the tensile strength was enhanced as compared to that of PP matrix.

In a novel study, Kobayashi and Takada investigated the effects of various compression molding conditions, including molding temperature, pressure and time on micro braided unidirectional hemp fibre reinforced PLA composites [101]. The study demonstrated an increase in tensile strength with an increase in molding temperature from 170°C to 190°C due to good impregnation of resin, whereas at high molding temperature of 210°C and above, decomposition of hemp fibres was reported.

4.2 Processing of thermoset composites

4.2.1 Hand layup

Hand layup is the oldest open molding technique for thermoset composites fabrication involving natural fibres in the form of woven mat, woven fabric or roving. In this process, an anti-adhesive is first applied to the mold surface, and woven fabrics are then positioned in the mold. Resin is poured and it is spread over the fibres with the help of brush or rollers. Layers are stacked in a similar way to achieve a desired thickness and entrapped air is removed manually with rollers to complete the laminate structure. Thermal and mechanical properties of epoxy composites manufactured by hand layup process based on three types of natural fibres viz. sisal, hemp and nettle fibres were studied by Lila et al. [102]. Tong and Xu studied and compared the mechanical properties of hemp fibre reinforced polyester composites fabricated by hand layup and compression molding technique [103].

4.2.2 RTM

RTM is a promising method for the fabrication of natural fibre reinforced thermoset composites as it is a cost effective method for the mass production of large and complex parts. It is a closed mold fabrication method and employs natural fibres in the form of mats or fabrics. Natural fibre preforms are placed in a closed mold and low viscosity thermoset resin mixed with catalyst is injected into the mold under low to moderate pressure to impregnate the preforms. The impregnation of fibres by resin can be improved by vacuum assisted injection and the process is then referred to as vacuum assisted RTM.

Rouison et al. adopted RTM process for fabrication of hemp fibre reinforced polyester composites and simulation was used to optimize this process [104]. The results showed that the simulations were in good agreement with experimental results, and the composites obtained by RTM were of high quality with a homogeneous structure. Also, the tensile, flexural and impact properties of these materials were reported to increase linearly with increasing fibre content.

5 Hemp fibre reinforced composites

Hemp fibres potential to replace synthetic fibres as reinforcement in polymer matrices for structural and engineering applications has resulted in a renewed interest in their composites owing to a need for the development of sustainable materials. In general, their renewable nature, relatively low cost, low fossil fuel energy requirements, good mechanical properties and applicability to available processing machines offer several advantages ecologically and economically over conventional fibres as reinforcement in polymer matrices. In natural fibre reinforced polymer composites, the polymer matrix plays an imperative role in determining the overall properties of composites as it not only protects the fibres from the environment but it also helps in load transfer to the fibres thereby governing the final mechanical properties of composites. Hemp fibre reinforced composites with both thermoplastic and thermoset matrices have been developed by several researchers. Table 3 lists the various polymer matrices employed for the fabrication of hemp fibre reinforced composites.

6 Hemp fibre-thermoplastic composites

Natural fibre reinforced thermoplastic composites predominantly find their applications in automobile sector in view of their low processing cost and design flexibility. Besides this, the modulus of natural fibres is higher than that of thermoplastics, which offer composites with higher modulus [12]. However, in consideration of the thermal degradation of hemp fibres above 200°C [47], which is mainly associated with the simultaneous thermal depolymerization of hemicellulose and pectins, only those thermoplastics whose processing temperature is below this can be utilized for fabrication of hemp fibre reinforced composites. Thermoplastics with high melting temperature bring about chemical changes in hemp fibres such as depolymerization, dehydration and oxidation which result in their thermal degradation. Hence, the most extensively used thermoplastics for this purpose are PP, PE, PS and polylactic acid (PLA). Hemp fibre reinforced composites based on thermoplastic matrices investigated in various studies are summarized in Table 4.

PP is the most widely studied thermoplastic given its low cost, low density, good mechanical properties, good impact strength, excellent processability and high temperature resistance. However, the main disadvantage associated with the use of hemp fibre in the PP matrix is a lack of good interfacial interaction between the two components. This is with regards to the presence of polar hydroxyl groups on the hemp fibre surface, which have difficulty forming a well bonded interface with the nonpolar polymer matrix. This ultimately results in poor stress transfer between the two components, which leads to overall inferior mechanical properties in the final product. Furthermore, the tendency of hemp fibres to form strong hydrogen bonds with each other often leads to their agglomeration upon incorporation in polymer matrix. The interfacial bonding between hemp fibres and PP matrix can be improved by various chemical modification methods as discussed in the previous section, but the most effective modification method from the standpoint of manufacturing economy is the use of coupling agents based on MAPP.

A study by Mutje et al. [72] investigated the effects of MAPP on the mechanical properties of hemp fibre reinforced PP composites. They observed that the wettability of hemp fibres in PP matrix was low due to their high surface energy as evidenced from the high polarity values of hemp fibres in contrast to PP matrix. However, using MAPP as a compatibility agent, not only enhanced the dispersion and adhesion at the interface between the two components but it also resulted in considerable increase in the mechanical properties of the resulting composites. The addition of 4% wt/wt of MAPP with respect to hemp fibres in composites with 40 wt% hemp fibres enhanced the ultimate tensile strength and flexural strength by 49% and 38%, respectively, compared with composites without coupling agent. Furthermore, another study by Mutje et al. compared the results obtained for MAPP compatibilized composites with glass fibre reinforced PP composites [116]. The results demonstrated that the mechanical properties of 40 wt% hemp fibres reinforced PP composites on the use of MAPP can amount to 80% of glass fibre reinforced PP composites.

Hemp fibre reinforced PP composites offer several advantages similar to those of glass fibre reinforced composites. However, they have low impact strength and modulus, which prevents their application in automotive parts. It has been reported by Espinach et al. that the Young's modulus of hemp fibre reinforced PP composites is not influenced by either alkali treatment or MAPP addition [117]. Similarly, a drastic decrease in the impact strength of PP composites has been reported on the incorporation of hemp fibres (10–40 wt%) regardless of their concentration, but the addition of MAPP assisted in restoring the impact strength [118]. This issue has been addressed in a novel study by Panaitescu et al. which analyzed the cumulative effects of adding differently treated hemp fibres along with a coupling agent MAPP and an impact modifier SEBS on the properties of PP composites [119]. The addition of hemp fibres to PP modified with MAPP and SEBS not only increased the tensile strength and modulus by 45% and 230%, respectively, but also enhanced the impact strength.

Table 3: General properties of different types of polymers.

Polymer	Density (g/cm3)	Tensile strength (MPa)	Failure strain (%)	Young's modulus (GPa)	Izod impact strength (J/m)	Melting temperature (°C)
Thermoplastics						
PLA	1.21–1.25	21–60	2.5–6	0.35–3.5	26	150
PP	0.89–0.92	26–41.4	15–700	0.95–1.77	21.4–267	160–176
HDPE	0.94–0.96	14.5–38	2.0–130	0.4–1.5	26.7–1068	120–140
PS	1.04–1.06	25–69	1–2.5	4–5	1.1	110–135
Thermosets						
Polyester	1.2–1.5	40–90	2	2–4.5	0.15–3.2	
Vinyl ester	1.2–1.4	69–83	4–7	3.1–3.8	2.5	
Epoxy	1.1–1.4	35–100	1–6	3–6	0.3	

PLA, poly(lactic acid); PP, polypropylene; HDPE, high density polyethylene; PS, polystyrene. Data from references [6, 8, 105, 106].

Table 4: Hemp fibre based thermoplastic composites.

Polymer	Surface treatment/coupling agent	Manufacturing method	Tensile strength	Flexural strength	Impact strength	Other properties	Observations	References
PP	Alkali treatment and MAPP as coupling agent	Extrusion and injection molding	✓	–	–	–	MAPP improved fibre–matrix interfacial bonding thereby improving composite strength and stiffness	[24]
PP	Alkali treatment and/or MAPP	Extrusion and compression molding	✓	✓	–	–	Alkaline fibre treatment and/or addition of a maleated coupling agent improve the interfacial adhesion with 30 wt% hemp fibre and 5 wt% MAPP addition exhibit best mechanical properties	[73]
PP/SEBS	MAPP	Extrusion and injection molding	✓	–	–	Thermal stability, storage modulus	A reduction of hemp fibre average length of about 2, 2.5, or 4 times, depending on the initial fibre length	[96]
PP	Maleic anhydride grafted polypropylene (PP–MAH), maleic anhydride grafted styrene-(ethylene-co-butylene)-styrene copolymer (SEBS–MAH) and maleic anhydride grafted poly(ethylene octane) (POE–MAH)	Extrusion	✓	✓	✓	–	Compatibilizer enhanced the interfacial adhesion between hemp fibre and matrix and combination of PP–MAH and SEBS–MAH or POE–MAH elastomer can be used to optimize the mechanical properties of composites	[107]
HDPE	Alkali treatment and triethoxyvinyl silane coupling agent	Extrusion and compression molding	–	–	–	Thermal properties and dynamic mechanical analysis	Thermal stability and stiffness increased with silane and alkali treatment with maximum stiffness at fibre volume fraction 40%	[108]
HDPE	Alkali treatment	Extrusion and compression molding	✓	✓	✓	–	Recycled PE/hemp fibre composites performed better than virgin PE/hemp fibre composites, with 40% fibre volume fraction being the optimized fibre loading	[109]
PE	Ethylene–MAH copolymer	Extrusion and batch mixer	–	✓	–	Moisture absorption	Coupling agent reduced the moisture absorption and composites prepared by batch mixing had better water resistance and mechanical properties	[110]
PLA	Alkali treatment	Hot press	✓	✓	–	–	Composite with 40% volume fraction of alkali treated fibre exhibited best mechanical properties	[111]
PLA	Alkali treatment and 3-aminopropyltriethoxy silane coupling agent	Extrusion and injection molding	✓	✓	✓	–	Tensile strength and flexural strength increased by 39% and 62% for composites with 40 wt% hemp	[112]

Table 4: (continued)

Polymer	Surface treatment/coupling agent	Manufacturing method	Tensile strength	Flexural strength	Impact strength	Other properties	Observations	References
							fibres and impact strength improved by 68% for 20 wt% hemp fibre composites	
PLA	–	Hot press	✓	–	✓	Thermal behaviour	Coefficient of thermal expansion decreased whereas mechanical properties increased with increasing the volume fraction of fibre	[113]
Polyamide 1010	Alkali treatment and ureido silane coupling agent	Extrusion and injection molding	✓	–	✓	Tribological behaviour	Mechanical properties and tribological behaviour improved with increase in fibre length	[114]
Poly(ε-caprolactone)	Nil	Extrusion	✓	✓	✓	Water absorption	Rate and quantity of water uptake increased with increase in aspect ratio of hemp fibres with a threshold value of 26 for optimal mechanical performance	[115]

PLA, poly(lactic acid); PE, polyethylene; PP, polypropylene; HDPE, high density polyethylene; MAPP, maleic anhydride grafted polypropylene.

The effect of MAPP and SEBS addition on mechanical and thermal properties of PP composites was most significant for alkali-silane treated hemp fibres, which doubled the tensile strength and tripled the Young's modulus due to strong interfacial interaction and defibrillation of hemp fibres.

An interesting study by Niu et al. analyzed the effects of three different compatibilizers, viz. maleic anhydride grafted polypropylene (PP–MAH), maleic anhydride grafted styrene-(ethylene-co-butylene)-styrene copolymer (SEBS–MAH) and maleic anhydride grafted poly(ethylene octane) (POE–MAH) on the mechanical and thermal properties of hemp/PP composites [107]. The results of the study demonstrated that use of all the compatibilizers significantly enhanced the interfacial adhesion between fibre and matrix, which resulted in improved stiffness, mechanical properties and thermal stability of the composites. Besides, combination of PP–MAH with SEBS–MAH or POE–MAH can be used for optimization of the mechanical properties of composites. Composites comprising 5 phr PP–MAH and 6 phr POE–MAH showed an increase in tensile strength, flexural strength, notched and unnotched impact strengths by 22%, 8%, 24% and 82%, respectively, compared to unmodified composites.

Polyethylene (PE) is another thermoplastic that is utilized for the fabrication of hemp fibre reinforced composites, considering its superior tensile strength and higher specific strength. Investigation on the hemp fibre/high density polyethylene (HDPE) composites analyzed the effects of alkali treatment and silane treatment on the thermal and thermo-mechanical properties of the composites [108]. Treated fibres showed higher thermal stability as compared to untreated fibres. On the contrary, thermal stability of the composites decreased with an increase in fibre loading. Besides, the stiffness of composites increased with an increase in fibre loading up to 40 wt%, with silane treatment exhibiting better thermomechanical properties than alkali treatment. In principle, this has been attributed to the stronger bond formation between fibre and matrix in case of silane treatment.

Another interesting approach was a comparative study of hemp fibre composites based on virgin and recycled HDPE matrix [109]. The results of the study showed that the mechanical properties of hemp/recycled HDPE composites were better than virgin HDPE composites, with an optimum fibre loading of 40% by volume, which exhibited highest tensile strength of 60.2 MPa and a flexural strength of 44.6 MPa. This improvement has been attributed to better compatibility of hemp fibre with recycled HDPE matrix, which has also been supported by impact testing results.

PLA a promising thermoplastic that has gained acceptance for commercial activities due to its biodegradable nature, is nowadays been widely exploited for the fabrication of natural fibre based composites as they result in green composites that are fully biodegradable. PLA has good mechanical properties, but its inherent brittle nature and high cost limit its commercial application for large scale production. Studies based on the use of hemp fibres as reinforcement in PLA have shown promising results in terms of improving the mechanical properties and lowering the total cost that can provide composites with potential applications for industries. As for the improvement of mechanical properties, Hu and Lim have shown that 40% volume fraction of alkali treated hemp fibres yielded PLA composites with tensile strength, elastic modulus and flexural strength of 54.6 MPa, 8.5 GPa and 112.7 MPa, respectively, which were much higher than that of PLA and they also exhibited lower densities [111].

7 Hemp fibre-thermoset composites

Thermoset polymers are the primary choice as matrix materials for fabrication of hemp and other natural fibre reinforced composites due to their superior mechanical properties and low processing temperature that prevents thermal degradation of natural fibres. They are predominantly used for structural applications like, for instance automobile interior linings, shipping, construction products, furniture and household products due to their good mechanical properties, high solvent resistance, dimensional stability and toughness. Polyesters, vinyl esters, epoxy and phenolics are some of the most commonly used thermoset polymers matrices for fabrication of hemp reinforced composites, which are summarized in Table 5.

Unsaturated polyesters are by far the most widely used thermoset matrices for hemp fibre composites owing to their low cost and flexibility for manufacturing large composite structures. A major obstacle in the use of hemp fibres is their high water absorption and their incompatibility with unsaturated polyesters. This has resulted in a lot of investigations on hemp fibre reinforced unsaturated polyester composites for further improvement of strength and water resistance. Mehta et al. studied the effects of alkali treatment, silane treatment, acrylonitrile treatment and unsaturated polyester-methyl ethyl ketone peroxide treatment of hemp fibres on the mechanical and thermal properties of the nonwoven hemp mat-polyester composites [45]. A comparison of mechanical properties between untreated and surface treated hemp fibre composites exhibited that acrylonitrile treatment was most effective in enhancing the tensile strength, tensile modulus, flexural strength, flexural modulus and impact strength by 80%, 25%, 35%, 7% and 180%, respectively, due to an improvement in the adhesion between hemp fibres and polyester matrix.

Similarly, the flexural and impact properties of surface treated hemp fibres reinforced polyester composites made by RTM process were studied by Sèbe et al. [125]. Surface treatment of hemp fibres with methacrylic anhydride introduced reactive vinyl groups at the surface via esterification reaction with hydroxyl groups on hemp. This enhanced the bonding between the fibres and the matrix due to which flexural strength and flexural modulus exhibited an increasing trend with fibre content, but impact strength decreased at low fibre content and then gradually increased with further fibre addition.

Epoxy is another thermoset matrix used for natural fibre composites as it has superior mechanical properties when compared to unsaturated polyesters. It offers excellent moisture barrier properties and possesses very good adhesion that makes it an ideal candidate for making natural fibre reinforced composites. Sepe et al. studied and compared the effects of alkali and silane treatments (3-glycidyloxypropyl trimethoxysilane) on the tensile and flexural properties of woven hemp reinforced epoxy composites [49]. It was reported that mechanical properties of silane treated hemp fibre composites are considerably better than those of alkali treated as the later leads to fibrillation of hemp fibres which results in fibre pull out and poor mechanical properties.

An interesting comparative analysis on the mechanical properties of epoxy and polyester composites reinforced with hemp fibres was done by Neves et al. [126]. It was found that epoxy composites with 30% volume of hemp fibres exhibited superior strength than those of polyester composites, with the flexural and tensile strength of 76.7 and 50.5 MPa, respectively, which was higher than corresponding polyester composites of 49.1 and 25.4 MPa, respectively.

The research done so far on hemp fibre thermoset composites has mostly been concentrated on the study of their tensile, flexural, impact properties and water resistance. All the studies showed that the use of hemp fibres in thermosets improved the mechanical properties to some extent, but their major drawback is their low impact strength compared to glass fibre composites. More research needs to be done to understand the impact behaviour of hemp fibre thermoset composites, enabling them to compete with glass fibre composites in the future.

8 Hemp fibres-hybrid composites

The concept of hybridization, by way of combining one or more polymers with more than one filler, offers the advantage of being able to overcome the shortcomings of conventional materials. Hybrid composite materials based on natural fibres tend to offer a balance between cost and performance because of their low cost, high strength-to-weight ratio and ease of manufacture that makes them viable for a wide range of uses in engineering applications and at the same time increases their eco-friendly nature [127–129]. Research based on natural fibre composites have consequently revealed that these composites are new age materials that provide an option to achieve high specific strength, modulus, impact and fatigue properties, which broadens their applicability for contemporary technologies specifically related to transportation, marine and aerospace sectors.

The possible variations of natural fibre based hybrid composites are obtained by the combination of natural–natural and natural–synthetic fibres. The hybridization of hemp fibres with both natural and synthetic fibres have been investigated in few studies, which are summarized in Table 6.

8.1 Hemp-synthetic fibre hybrid composites

Hybridization of hemp fibres with synthetic fibres is a promising way to enhance the performance and properties of resultant composites, especially for automotive sector. The effects of hybridization of hemp fibres with glass fibres in PP composites were investigated by Panthapulakkal and Sain with reference to their mechanical performance and water absorption behaviour [130]. The results of the study indicated that the hybridization with glass fibres enhanced the strength, stiffness, thermal stability and reduced the water absorption tendency of the corresponding composites. However, water ageing resulted in a significant reduction of strength and stiffness due to de-bonding at the fibre matrix interface as a result of water absorption [131].

Similarly, Asim Shahzad assessed the influence of short hemp fibre mat hybridization with chopped glass fibre mat on the impact and fatigue properties of unsaturated polyester composites [132]. The study reported that replacing 11% of hemp fibres with glass fibres considerably enhances the impact damage tolerance of the composites. However, impact damage tolerance was limited to impact energy of 15 J at which composites showed 90% loss in their intrinsic strength and stiffness. The hybrid composites also exhibited an improvement in fatigue properties.

Carbon fibres are another interesting class of synthetic fibres that find use in industrial applications in view of their characteristic mechanical properties. Using carbon fibres in combination with hemp fibres for production of epoxy hybrid laminates, it has been shown that it is possible to tune the flexural, inter-laminar shear and impact properties by changing the relative position of carbon and hemp layers in the stacking sequence of hybrid composites [133].

8.2 Hemp-natural fibre hybrid composites

Hybridization of natural fibres with natural fibres in fabrication of polymer composites has the advantage of overcoming the short comings of individual fibres, resulting in balanced performance as well as economic and environmental benefits by way of sustainability. Many studies are reported in the literature on the hybridization of hemp fibres with other natural fibres. An interesting comparative analysis was performed by Chaudhary et al. on the mechanical properties of natural fibre reinforced epoxy composites that employed different natural fibres and their combinations: jute/epoxy, hemp/epoxy, flax/epoxy and hybrid composites jute/hemp/epoxy, hemp/flax/epoxy and jute/hemp/flax/epoxy composites [135]. Among individual fibre composites, flax fibres improved the tensile strength and hardness, jute enhanced the impact strength, whereas hemp reinforced composites exhibited an improvement in flexural strength only. However, in jute/hemp/flax/epoxy hybrid composite, tensile strength, modulus and flexural strength were higher than in any other hybrid composite. Similarly, the impact performance of this hybrid composite was also improved in response to the enhanced load absorbing capacity and delayed crack initiation and propagation mechanism as compared to individual fibres.

In an extension of this study, the wear and dynamic mechanical behaviour of the above mentioned composites were also investigated by Chaudhary et al. [136]. It was reported that the wear behaviour of epoxy composites was significantly improved as compared to neat epoxy polymer on the incorporation of natural fibres, with jute/hemp hybrid composite exhibiting better wear performance.

Table 5: Hemp fibre based thermoset composites.

Polymer	Surface treatment/coupling agent	Manufacturing method	Tensile strength	Flexural strength	Impact strength	Other properties	Observations	References
Epoxy	Alkaline treatment and (3-glycidyloxypropyl) trimethoxysilane coupling agent	Vacuum infusion process	✓	✓	–	–	Silane treatment of hemp fibres improves, both tensile and flexural properties of the composites	[49]
Epoxy	Alkali treatment with 5 wt% NaOH and 2 wt% Na_2SO_3	Compression molding	✓	✓	✓	–	Alkali treatment separated fibres with cleaner surface topography, increased cellulose crystallinity and thermal stability thus increasing tensile and flexural strength but decreased impact energy	[120]
Unsaturated polyester	–	Resin transfer molding	✓	✓	✓	Flexural creep	Tensile, flexural and impact properties increased linearly with increasing fibre content but material not suitable for high load fatigue conditions	[104]
Unsaturated polyester	Alkali treatment	Hand layup and compression molding	✓	–	✓	Fatigue strength	1% and 5% alkali treatment improved tensile strength and fatigue strength, but no improvement in impact energy	[121]
Polybenzoxazine	Alkali treatment	Compression molding	✓	✓	✓	Water absorption	Tensile strength, flexural strength and impact strength increased as hemp fibre vol.% increased and fibre diameter decreased	[122]
Polyurethane	Alkali treatment and 3-mercaptopropyltrimethoxysilane or 3-aminopropyl trimethoxy silane agent	–	✓	–	–	Thermal conductivity	Modification of fibres improved the interfacial adhesion and increased tensile strength by 39%	[123]
Polyurethane	Alkali treatment	Brabender Plasti-Corder internal mixer and mini press	✓	✓	–	–	Maximum tensile and flexural strength observed for 15 mm optimum fibre length and 40% fibre volume	[124]

Table 6: Hemp fibre based hybrid composites.

Polymer matrix	Hybrid reinforcement	Processing method	References
PP	Glass fibre/hemp	Injection molding	[130]
PP	Glass fibre/hemp	Injection molding	[131]
Unsaturated polyester	Glass chopped strand mat/hemp short fibre mat	Hand layup followed by compression molding	[132]
Epoxy	Woven carbon fabric/woven hemp fabric	Hand layup and vacuum compression molding	[133]
HDPE	Basalt/hemp	Extrusion and injection molding	[134]
Epoxy	Jute/hemp/flax	Hand layup	[135]
Epoxy	Flax/hemp	Compression hand layup	[136]
Epoxy	Sisal/hemp	Cold pressing	[137]
PLA	Sisal/hemp	Extrusion and injection molding	[138]
PP	Sisal/hemp	Injection molding	[139]
Epoxy	Banana/hemp/glass fibres	Hand layup	[140]
Polyester	Nettle/hemp	Hand layup	[141]

PLA, poly(lactic acid); PP, polypropylene; HDPE, high density polyethylene.

Saha et al. studied the water absorption behaviour of flax–hemp reinforced epoxy composites fabricated by compression hand layup technique, with a focus on the thermo-mechanical properties. It was observed that the water absorption had a detrimental effect on the mechanical properties of the composites. However, hybridization of flax fibres with hemp fibres reduced the water absorption tendency of the composites, resulting in better performance with regard to hemp fibre reinforced composites.

Akash et al.'s study on 10% NaOH treated hemp/sisal fibre reinforced epoxy composites found an increase in flexural strength up to 40% of fibre loading [137]. Besides, the hardness of the composites increased with an increase in fibre content, but at the same time, water absorption increased for the composites.

The effects of hybridization have also been investigated in 100% green composites comprising PLA and hemp and sisal fibres. The study employed 15 wt% of hemp and sisal fibre each for hybridization and the results revealed an enhancement in tensile strength, tensile modulus, flexural strength, flexural modulus and impact strength of hybrid composites as compared to neat PLA [138].

9 Future scope

Hemp fibres are one of the most extensively utilized natural fibres in a wide range of structural applications because of their exceptional mechanical qualities. However, their potential as reinforcement in composites is limited owing to their hydrophilic nature, which makes them incompatible with most of the polymer matrices. Therefore, in order to enhance fibre–matrix adhesion, various physical and chemical methods of hemp fibre modification have been developed that allow for improved interfacial interactions at the fibre–matrix interface to yield composites with enhanced mechanical properties. Nevertheless, the necessity for greater in-depth research on hemp fibre modification is driven by their unique characteristics and the practical requirements for improving their properties to complement their intended applications. In light of these observations, recent efforts tried to combine multiple strategies in order to optimize the properties of hemp fibres and their effects in corresponding composites while lowering manufacturing costs and reducing the overall adverse side effects at the same time.

10 Conclusions

Hemp is one of the most widely used natural fibres that has proved to be a potential candidate for structural applications due to its specific mechanical properties. Owing to its wide range of applicability, it is essential to have a fundamental knowledge of hemp fibre processing and fabrication techniques that affect the properties of hemp fibre reinforced composites. Their hydrophilic character and incompatibility with polymer matrix are two key factors that are major limitations in the use of hemp fibres as reinforcement in composites. These factors result in poor fibre–matrix interfacial bonding, thereby reducing the mechanical properties of composites. An appropriate selection of chemical/physical modification methods can improve the interfacial interaction at the fibre–matrix interface and subsequently yield composites with enhanced mechanical properties. Similarly, the selection of a suitable manufacturing process depending on the type of polymer matrix would imply better quality and high performance of the composites. Thus, it can be concluded that hemp fibre reinforced composites are cost effective with the potential to be the future generation engineering materials for automotive, construction and consumer goods.

Author contributions: The conceptualization of this article, literature survey, data analysis, and drafting were solely performed by G. S. Deshmukh.
Research funding: None declared.
Conflict of interest statement: The author declares to have no conflicts of interests regarding this article.
Data availability: Data sharing not applicable to this article as no datasets were generated or analyzed during the current study.

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