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Interfacial Bonding Mechanisms of Natural Fibre-Matrix Composites: An Overview

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The development of natural fiber (NFr) composites for a variety of applications is on the rise. The optimization of the interfacial bonding (IFB) between the reinforcing NFr and polymer matrix is perhaps the single most critical aspect in the development of natural fibre polymer composites (NFPCs) with high mechanical performance. While the IFB is critical in determining the mechanical properties of the NFPCs, such as stress transfer, it is one of the least understood components. This article offers a summary of IFB mechanisms, different modification approaches targeted at lowering incompatibility and improving IFB, and evaluation of the impact of IFB. It has been found that 1) In general, interdiffusion, electrostatic adhesion, chemical reactions, and mechanical interlocking are accountable for the IFB; 2) the incompatibility of the fibre and matrix, which results in poor dispersion of the fiber, weak IFB, and ultimately worse composite quality, may be addressed through strategic modifications; and 3) Interfacial interactions between polymers and nanoparticles (NPs) are significantly improving their performance in areas like thermal, mechanical, robust IFB, and moisture absorption. As a result, this review study could be an important resource for scholars interested in coating and treating NFr to further enhance their surface characteristics.

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INTRODUCTION

The importance of composite materials in various NFPCs has grown as the demand for lighter density and high-performing materials has increased. In the aerospace and automotive industries, NFr reinforced composites have replaced cutting-edge metal alloys as structural components. The onus on current technology to deploy lightweight electric vehicles will undoubtedly enhance the importance of NFr composites among all reinforcing fibres composites in the next years. NFPCs have grown in prominence as a subset of composite materials during the previous decade. The advantages that natural fibres have over inorganic fillers and reinforcements, such as their availability, environmentally friendly nature, biodegradability, nontoxicity, cheap cost, manufacturing scalability, and high flexural and tensile modulus, are driving this rapid development (George *et al.* 2001; Mukhopadhyay *et al.* 2003). In the realm of NFPCs, readily available, renewable, and biodegradable NFr are replacing classic synthetic fibres such as glass and carbon.

NFPCs are the most exquisite and promising materials of this century. Their durability and integrity in diverse service settings can be affected by the response of their constituents, *i.e*., the fibre, polymer matrix, and the existing interface/interphase between the fibre and polymer matrix in that environment. The primary reasons for the usage of NFPCs in various applications have been reduced weight, reduced cost, increased performance, decreased fuel consumption, and decreased pollution (Balakrishnan *et al.* 2016). However, this study narrowly focuses on composites whose strength typically peaks at fibre contents of 40 to 55 wt% for matrix composites, with the reduction at higher fibre contents explained as a result of poor wetting leading to reduced stress transfer across the fibre–matrix interface and increasing porosity. Stiffness has been found to increase up to higher fibre contents of around 55 to 65 wt% with similar materials, possibly due to less dependency on interfacial strength than composite strength (Le and Pickering 2015; Madsen *et al*. 2009). In addition to being a problem for the short-term qualities of composites, high fibre volume fractions are also a concern due to the possibility of increased water absorption, resulting in the loss of longer-term characteristics. Significant progress has been achieved in this area, and there are a sufficient number of review papers available. Pickering *et al.* (2016), provided an overview of the performance characteristics of NFr reinforced polymer composites. Saba *et al.* (2016) compiled a list of dynamic mechanical characteristics of these composites. Vaisanen *et al.* (2017) conducted a critical review of the impacts of waste material. Faruk *et al.* (2012) reviewed all of the developments that occurred between 2000 and 2010. They discussed various NFr, their availability, treatment procedures, and the different polymer matrices employed. Sanjay *et al.* (2018), provide a complete analysis of the characterisation techniques and attributes of these composites. Sood and Dwivedi (2018) focused on the flexural characteristics of these NFPCs. Oqla *et al.* (2015), described NFPC-like conductive polymer composites. Thakur and Thakur (2014), described how natural cellulose fibre is processed and characterised in a thermoset matrix. Omrani *et al.* (2016) investigated the tribological characteristics and utilization of polymer matrix composites enhanced with these novel materials. Vaisanen *et al.* (2016), discussed the use of agricultural and forest industry waste and residues in these NFPCs. Regardless of these benchmarking properties, optimising the IFB between natural fibres and polymer matrix is one of the most important techniques for achieving the best NFPCs formulation (Thakur and Thakur 2014).

Surface treatment of fibres has been widely used to improve IFB by adding polar functional groups and improving acceptable surface roughness (Zheng *et al.* 2004; Li *et al.* 2017). Surface treatment is a meticulous and deliberate technique to change the chemical surface environment and topography of the fiber in order to trigger adhesion-promoting processes. The experiment and work done by Oushabi *et al.* (2018) indicated that the chemical treatments of date palm fibres, in particular the alkali treatment and the treatment with organosilanes, significantly enhanced the interfacial properties between the fibres and the organic matrices, such as polyurethane and epoxy, as determined by the pull-out tests. Similar research by Rizal *et al.* (2018), demonstrated that the alkali treatment of typha fiber enhanced the interfacial compatibility between the epoxy resin and typha fiber, resulting in enhanced mechanical capabilities and a hydrophobic composite. After 5% alkaline immersion, the tensile, flexural, and impact strengths of the typha fiber reinforced epoxy composite increased in comparison to the untreated typha fiber composite.

The IFB between NFr and matrix acts within an interaction or propagation region where two phases or elements interact mechanically, physically, and chemically, as shown in Fig. 1. The interfacial adhesion between the fibre and matrix is critical in determining the mechanical properties of the composite (Kabir *et al.* 2012). The IFB between the fibre and matrix is influenced by mechanical interlocking, chemical bonding, and attractive molecular forces. However, naturally hydrophilic NFr is inherently incompatible with hydrophobic polymers. In addition to the pectin and waxy components in NFr acting as an obstacle to interlocking with the nonpolar polymer matrix, the presence of a large number of hydroxyl groups inhibits its operative reaction with the matrix (Bledzki *et al.* 1998; Kazayawoko *et al.* 1999). Therefore, modifying the surface characteristics of NFr and hydrophobic polymer matrix is critical to develop a suitable composite with excellent IFB and efficient inherent stress convey across the interface region. Physical modifications (such as solvent extraction, heat treatment, corona, and plasma treatments), physic-chemical modification (such as laser, and UV bombardment) (John and Anandjiwala 2008), surface coating (Kabir *et al.* 2012), and chemical treatments including alkaline (Fiore *et al.* 2015), silane (Agrawal *et al.* 2000), acetylation (Tserki *et al.* 2005), benzoylation (Li *et al.* 2007), and coupling agents have all been tried to improve the compatibility and bonding between NFr and matrix (Saheb and Jog 1999; El-Abbassi *et al.* 2015). Chemical treatments and surface coatings, in particular, were shown to be useful methods for increasing the mechanical strength of composites by improving their wettability and interfacial characteristics. Kabir *et al.* (2012) highlighted the critical role of chemically treated NFr in enhancing the efficiency of their composites. Furthermore, Li *et al.* (2007), investigated various chemical treatment methods and their mechanism of action in improving the properties of composites. Mohanty *et al.* (2001), discussed how to modify the surface of NFr and how this affects their biocomposites. However, the majority of these surface treatments improved the surface of NFr, but the ultimate consequence was an improvement in the interfacial bonding (IFB) between the NFr and polymer matrix. Once the IFB is enhanced, all the mechanical characteristics of the NFPCs would be supported (Kabir *et al.* 2012).

These modifications significantly enhanced the IFB, wettability, roughness of NFPCs. However, such composites (with organic compounds) are not feasible in structural and high temperature applications. In general, the NFPCs are required to bear a high load in high structural applications (like tensile and compression). Improved compression strength, tensile strength, toughness, and stiffness are needed to withstand such a high load. Thus, to establish a durable interface region, an adequate scale of physicochemical interactions is required, which might be fostered by van der Waals, hydrogen, and covalent chemical bonding between NFr and matrix. Numerous surface treatments approaches have successfully improved IFB *via* chemical interactions and mechanical interlocking (George *et al.* 2001; Bogoeva-Gaceva *et al.* 2007; Li *et al.* 2007). However, a combination of modification approaches has also been used to achieve superior IFB (Karnani *et al.* 1997; Zhou *et al.* 2016). Recently, nano-reinforcement materials such as nano-$CaCO_3$, zinc oxide NPs, and titanium dioxide have been employed to alter the polymer matrix and fibre in order to achieve the maximum possible IFB through synergy (Wang *et al.* 2012; Soltani *et al.* 2013; Li *et al.* 2017).

This paper focuses primarily on the IFB mechanisms of NFr composites and is organized into three main sections: an overview of the IFB mechanisms, different surface modification approaches targeted at improving IFB between NFr and polymer matrix, and the evaluation the impact of IFB on the mechanical performance of NFPCs. This article will serve as a foundation for future research and industrialisation of NFPCs. In the current situation, where traditional modification techniques are widely utilized to modify NFr for potential industrial applications, researchers and engineers cannot agree on which modification techniques provide the most significant benefits while maintaining the integrity of NFr. As a result, it is vital to highlight the advantages and disadvantages of present NFr surface modification approaches and to envision concrete paths for future improvement. In response to the rising number of publications, this review will discuss the most recent developments in NFr surface modification and the current state of modified NFPCs. Numerous modification strategies have been discussed in detail with suitable examples to demonstrate the effect of surface treatments on the interfacial characteristics. Additionally, the strengths and weaknesses of each modification strategy have been evaluated based on the existing literature.

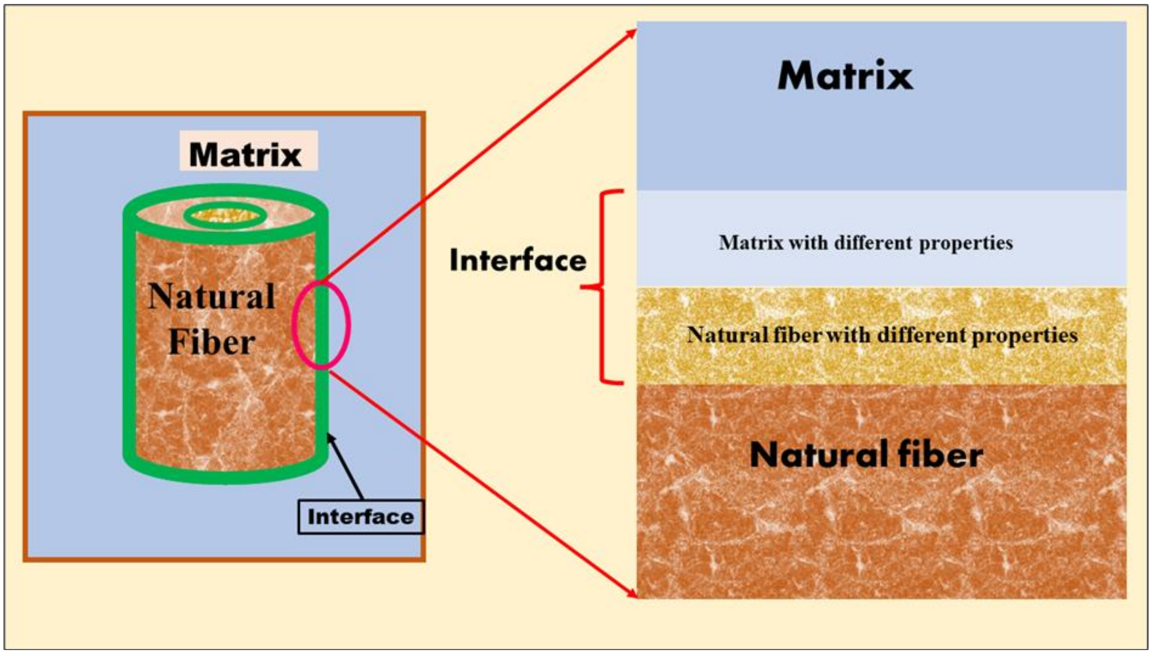

Fig. 1. Schematic illustration of the interphase in the composite

NATURAL FIBRE POLYMER COMPOSITES

NFPCs are multi-phase materials composed of natural reinforcing fibres originating from plants or animals. The combination of fibre and polymer matrix produces synergistic qualities that cannot be obtained from either component alone. NFr provide several advantages over synthetic fibres, including superior rigidity and strength, low cost, environmentally benign nature, biodegradability, and the fact that they are renewable materials. This review concentrates on composites made from natural-derived fibres. NFr can be categorised generically into the stem, fruit, and leaf fibres, as illustrated in Fig. 2.

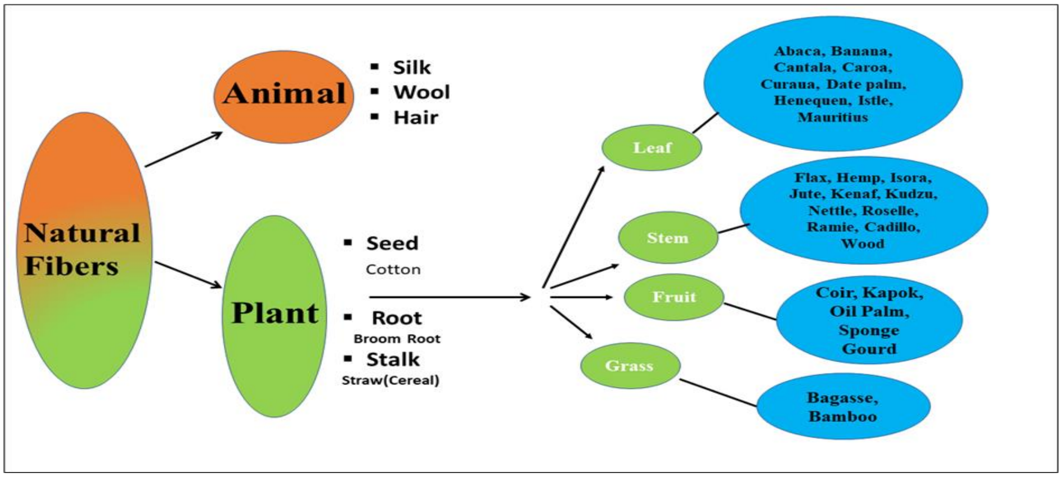

Fig. 2. Schematic representation natural fibres' origin

CHEMICAL COMPOSITION OF NATURAL FIBRE

Natural fibres primarily consist of cellulose, lignin, hemicellulose, pectin, and waxy surface, *etc*. (Pickering *et al*. 2016). Hemicellulose and lignin are major components of the secondary cell wall. The crystalline and amorphous portions of cellulose vary from plant to plant and determine the stiffness and mechanical strength of the fibre (Suryanto *et al.* 2014). Figure 3 is a simplified model of composition of natural fibres that depicts the structure of the cellulose molecule, which is made of D-glucopyranose, which is interconnected with b-1,4- glycosidic linkage having a degree of polymerization of 10,000 (Kbir *et al.* 2013; Celino *et al.* 2014). Intramolecular hydrogen bonding is accomplished *via* the –OH group in cellulose. The microfibril angle is the alignment of this cellulose molecule with regard to the fibre axis, which, along with the degree of crystallinity of the molecule, defines the fibre strength. The larger the crystallinity and smaller the microfibril angle, the greater will be the strength of the fibre (Eicorn 2001; Reddy and Yang 2005). The presence of bulky pendant groups in the structure of hemicellulose and lignin causes them to be amorphous. Hemicellulose comprises a mixture of 5 and 6 carbon ring polysaccharides with 50 to 300 degrees of polymerization, whereas lignin contains an aliphatic and aromatic component composed of 4-hydroxy-3-methoxyphenyl propane. They are quite stable and insoluble in most solvents; however, they dissolve in alkali (John and Thomas 2008). NFr can be used to make sustainable composite materials because of their abundance and accessibility of availability.

Natural fibres generally contain large amounts of the hydroxyl group, which can form hydrogen bonds with polar molecules such as water. Water adsorption is primarily influenced by the relative humidity of the surrounding atmosphere, crystallinity, and amorphous content of cellulose. Cellulose with a high degree of crystalline structure can only accommodate water on its surface. In contrast, if the proportion of the amorphous phase is higher, then water molecules can permeate the interior of cellulose material (Gauthier *et al.* 1998). As a result, the intake of water molecules has unfavourable consequences. During composite manufacturing, the –OH content of the NFr is accountable for its inability to adapt to a hydrophobic matrix. Water is absorbed and transferred to the bulk of the fibre from the surface by diffusion or capillary action (Munoz and Garcia-Manrique 2015).

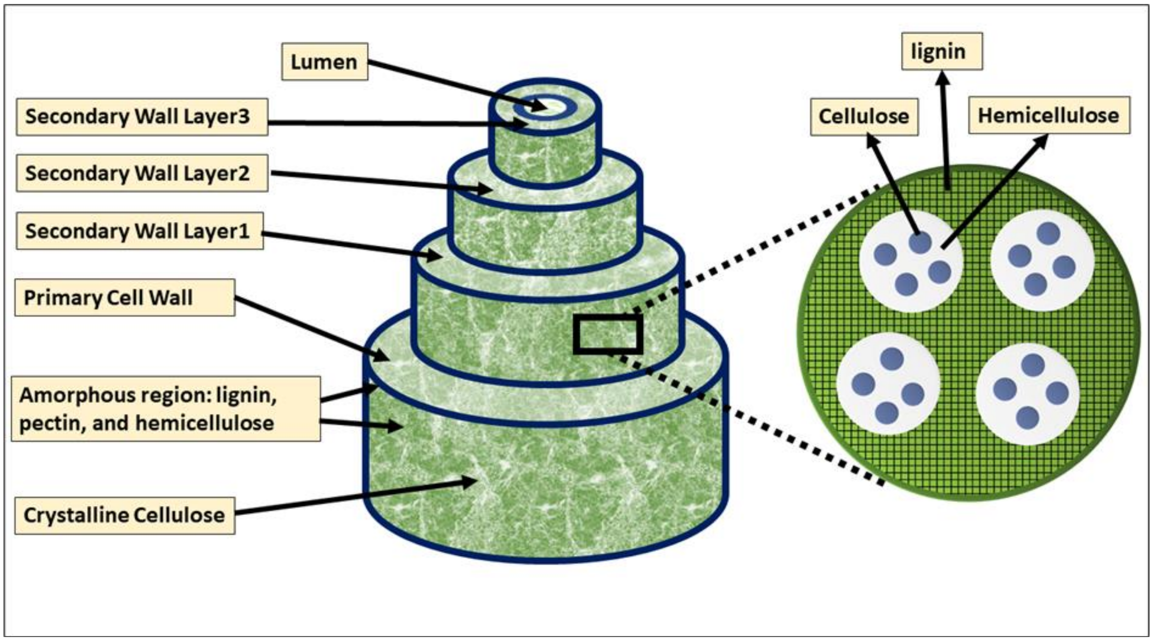

Fig. 3. Simplified model of the composition of natural fibres

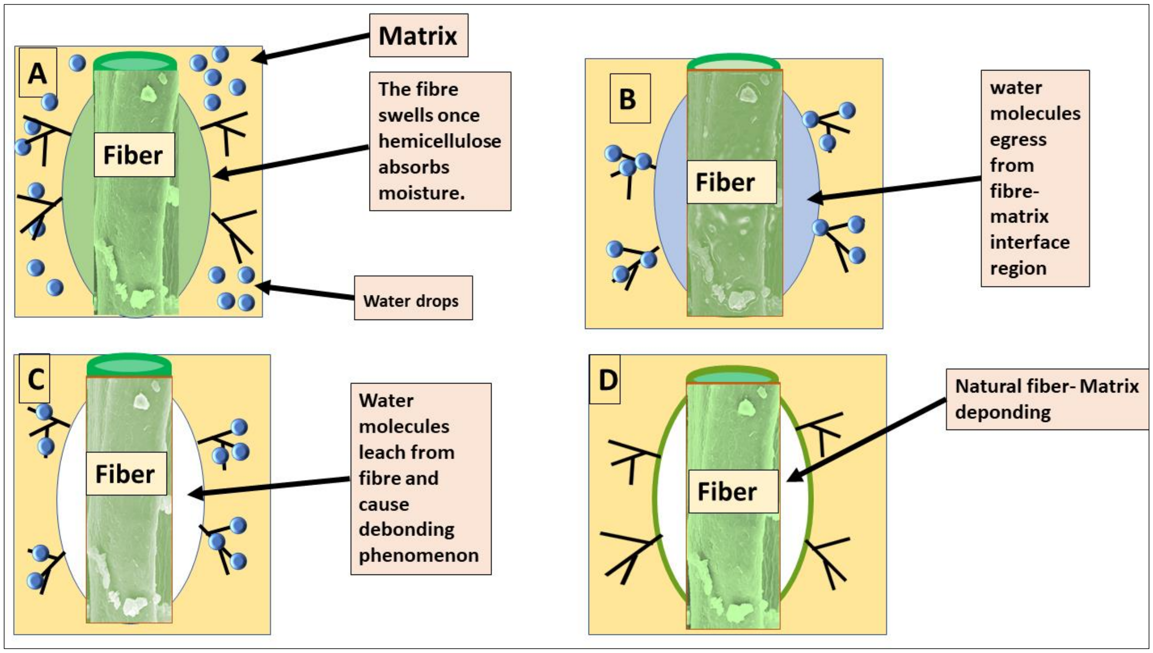

Fig. 4. Schematic representation mechanism of water absorption

As shown in Fig. 4, water flows into the fibre-matrix interface region, which results in the leaching of water-soluble compounds from surface of the NFr and the debonding between NFr and matrix (Azwa *et al.* 2013). The strength of the composites produced is determined by the efficiency of the IFB between NFr and matrix. If the IFB between the NFr and the matrix is too robust, then the applied load will result in the formation of a fracture that does not distribute the stress from the matrix to the NFr, producing a brittle material that breaks down (Hidalgo-Salazar *et al.* 2013). Simultaneously, if the IFB is inadequate, the necessary stress transfer from matrix to NFr will not occur. As a result, the mechanical strength of the composites decreases with time. According to research conducted by many researchers, NFr have a higher water absorption capacity, which rises with increasing fibre loading and time spent in humid circumstances, resulting in poor transfer of stress from matrix to NFr, culminating in composite failure (Vijayakumar *et al.* 2014; Bujjibabu *et al.* 2018). Because of inherent flaws in NFr, surface treatment of the fibres can be accomplished through chemical treatments or additives during composite production.

SIGNIFICANCE OF FIBRE/MATRIX INTERFACE

When fibres are introduced to a matrix, the mechanical characteristics (*e.g.*, tensile strength and bending stiffness) of the material can be increased. Fibres serve two functions: 1) carrying most of the compressive or tensile stress imparted to NFPCs, and 2) bridging matrix cracks and reducing crack propagation by dissipating energy around the fracture tip. The stress is transmitted to the NFr *via* the surrounding matrix when an NFPC is exposed to axial tensile stress, as shown in Fig. 5(a).

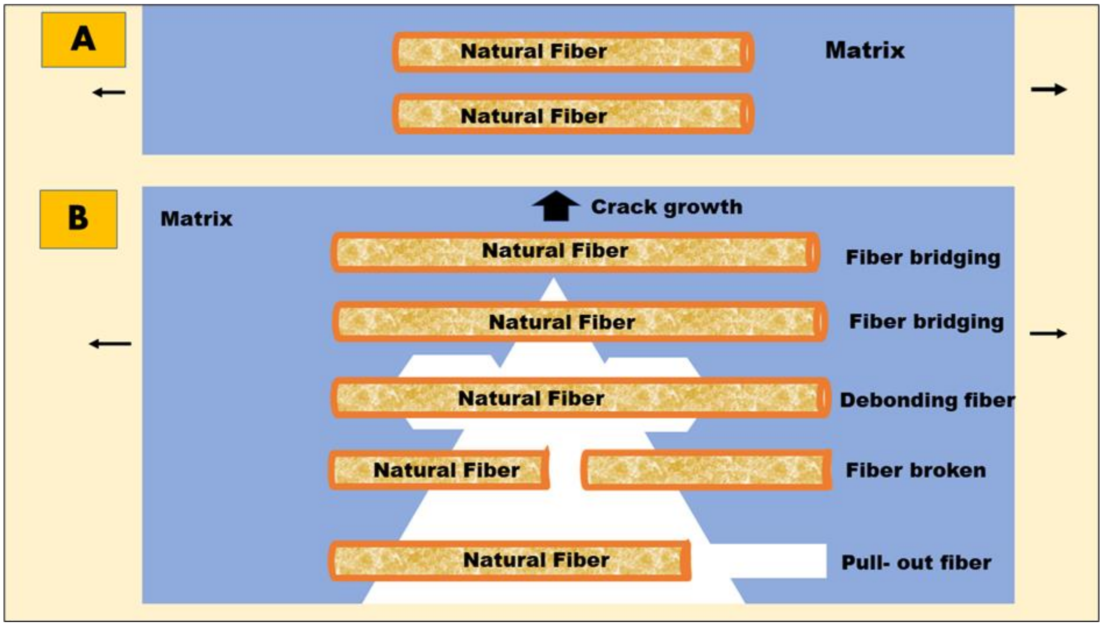

Fig. 5. (a) Simplified model of the load transferring at fibre/matrix interface and (b) fibre bridging

The process begins with matrix deformation due to the imposed load. The matrix deformation then causes shear stress at the IFB region between the NFr and matrix (Young 2015). To balance the shear stress, the tensile stress of the fibre is triggered. The fibre carries most of the tensile stress imparted to the NFPCs during this procedure. Figure 5(b) shows the mechanism of fibre bridging. Microcracks begin in the matrix because of its lower fracture strain than fibres. When the strain energy near the crack tip is more than the energy required to build a new surface, the crack will propagate. The stress carried by the matrix is transferred to the fibre *via* the interface when fibres bridge a crack in the matrix. Based on the interfacial shear strength of the fibre-matrix interface and the tensile strength of the fibre, interface failure (*e.g.*, fibre pull-out and interface debonding) or fibre fracture will occur as the stress increases. The strain energy is dissipated around the crack tip by interface failure and fibre fracture, preventing crack propagation.

In NFPCs, the interface is the shared boundary between the fibre and the polymer matrix, through which load can be transferred from the matrix to the fibres based on strain compatibility. If the interface fails prematurely, then the deformation of the matrix (or strain) is incompatible with that of the fibre, preventing the load from being transferred from the weak component (matrix) to the robust component (fibre). In other words, it is impossible to accomplish the reinforcing impact of fibres on the polymer matrix. Consequently, strain compatibility at the interface is vital for the structural integrity of NFPC's materials. Due to the different mechanical (*e.g.*, elastic modulus) and physical (*e.g.*, swelling ratio) characteristics of fibres and the matrix, it is difficult to establish strain compatibility under stressed and hostile environmental circumstances. Stress concentration is more likely to occur at the interfacial region than in the fibres or matrix, making it easier to develop microcracks at the IFB between fibre and matrix. The load-carrying capability of the NFPCs material declines as the microcracks propagate. It is critical to investigate the fibre-matrix interfacial characteristics to make the best use of NFPCs material.

Furthermore, the IFB between fibre and matrix is crucial for the long-term mechanical characteristics of NFPCs in hostile environments. For instance, when a NFPC composite is subjected to a high humidity environment (*e.g.*, 95% RH), the mechanical characteristics of the complete NFPCs decline due to the degradation of the fibres, matrix, and IFB between fibre and matrix. The hydrolysis occurs in the matrix and NFr, causing the mechanical characteristics degradation of natural fibres and matrix. The transported moisture degrades the interfacial bonding by weakening the chemical bonds and mechanical interlocking at the interface of the fibre and matrix. In addition, the difference in swelling between the fibre and matrix causes microcracks at their interface (Bradley and Grant 1995; Heshmati 2017). When exposed to high humidity for an extended period of time, the microcracks generated by the swelling mismatch in the early stages of exposure create additional pathways for moisture diffusion, accelerating the propagation of the damage in the natural fibres, matrix, and the IFB between fibre and matrix. Compared to natural fibres derived from plants, synthetic fibres undergo comparatively little hydrolysis.

An investigation was carried out by Sanjeevi *et al*. (2021), to understand the effects of water absorption on the mechanical properties of hybrid phenol formaldehyde (PF) composite fabricated with Areca Fine Fibres (AFFs) and Calotropis Gigantea Fibre (CGF). The mechanical characteristics of the hybrid composites in a wet condition were inferior than those in a dry condition. The tensile strength of 45 wt% composite at wet condition was 40 MPa lower than the 35 wt% composite in a wet condition due to the penetration of water molecules into the fibre–matrix region. The penetration led to the change of dimensions of the composite specimens and caused weak IFB, thereby decreasing the tensile characteristics. In addition, Mohammed *et al*. (2017) studied the weather effects on mechanical properties of pure kenaf and the kenaf/glass fiber hybrid composites. The unexposed pure kenaf composites and the kenaf/GF hybrid composites showed an approximate tensile strength of 60.8 and 70.9 MPa, respectively. There was a decreasing trend in the tensile strengths of the pure kenaf composites and the kenaf/glass fibre hybrid composites. Based on the results, it can be observed that the pure kenaf composites showed a higher decrement as compared to the kenaf/GF hybrid composite. This was due to the inclusion of GF to the top and bottom layers of the pure kenaf composites, which worked as a barrier and impeded moisture transfer to the kenaf fibres. The ideal interface ought to fulfil the following objectives.

Bonding Layer

The appropriate interface should have the optimum bonding strength for the interface. When the matrix crack stretches to the IFB phase, the acceptable IFB between fibre and matrix can mitigate the stress concentration at the front end of the matrix crack, prevent fibre debonding, prevent crack deflection, prevent the crack from expanding into the fibre, and consume fracture energy (Hestiawan *et al.* 2018). To accurately capture the damage initiation and propagation at the constituent level, three-dimensional Representative Volume Element (RVE) models have been developed by modelling the fibre, the matrix, and the coating material separately. In addition, numerous geometrical parameters, including the random distribution of the fibres, the fiber-coating, and the matrix-coating interface decohesions, were considered (Daggumati *et al.* 2020). In addition, the extended finite element method (XFEM) technique was utilised to capture the coating and matrix material fracture behaviour (Daggumati *et al.* 2020).

Load Transfer

The IFB phase must be sufficiently strong to operate as a bridge between the matrix and the NFr, allowing stress to convey between them to occur (Merotte *et al.* 2018). As a result of the poor interface bonding, the interface tends to debond under low tension, making it difficult to convey the load properly. As a result, the strengthening effect of NFr is not fully realized, resulting in inferior mechanical characteristics in composites. If the strength of IFB between fibre and matrix is excessively high, then the interface will be unable to modify the stress distribution, resulting in brittle fracture when the composite is loaded. Only when the interface layer has adequate material, and bonding strength that enables the interface simultaneously convey load and modify stress, allowing composites to achieve superior mechanical qualities (Arbelaiz *et al.* 2005; Sain *et al.* 2005).

In a mechanically loaded composite, it is usually assumed that the fibres and the matrix are bonded together and that no different movement exists between them. The strain in each component of the composite can then be deemed same and equal to the composite's overall strain (Yin *et al.* 2017). For an elastic response, the stress on a unidirectionally reinforced composite loaded parallel to the fibre axis σ_c, can be calculated by multiplying the elastic modulus in that direction E_c, by the measured strain ε_c

$$\sigma_C = E_C \varepsilon_C \tag{1}$$

A weak interfacial bonding tends to dissipate more energy than that of a robust interfacial bonding. As it pertains to fiber-reinforced composites, debonding can improve the stick–slip friction under imposed cyclic loads, leading to enhanced damping qualities. Geethamma *et al*. (2005) examined the influence of chemical treatment of coir fiber on damping; the results indicated that the poorer the interfacial bonding, the better the damping capacity is. It has been found that the weak bonding between fibers and matrix could be attributed to the relative motion of fibers. To create composites with high damping, both energy-dissipating material and strong shear strain are required. Thus, improving energy dissipation behaviour through fibre surface treatment to strengthen interfacial damping effects is effective. It was presented that the interfacial damping mechanisms in fiber reinforced composite are closely related to interfacial debonding and friction. Recently, dampening behaviour has evolved to be a hot topic. Fiber reinforced composite with robust damping characteristic is cost-effective for increasing energy attenuation in engineering system applications (Tang and Yan 2020). The intrinsic viscoelastic damping of the matrix is the principal contributor to damping in fibre-reinforced composites. Under dynamic loading, it has been demonstrated that the matrix carries both extensional and shearing stresses. Particularly, damping behaviour related with stress distribution perpendicular to the reinforced fibre axis, commonly known as in-plane shear stress, is essentially entirely dictated by matrix damping (Treviso *et al.* 2015). It has been claimed that raising the matrix volume percentage at the expense of stiffness and strength can increase damping. The reason can be attributed to the higher stresses in the matrix due to closer spacing which resulted in higher energy dissipation capacity during the dynamic loading (Haddad and Feng 2003; Melo and Villena 2012).

Various types of fibres are currently employed for engineering applications, with carbon fibre dominating lightweight structural composites because of its high modulus and low density. Among the most popular natural fibrous materials, the damping behavior of short coir fiber reinforced natural rubber composites (Geethamma *et al*. 2005), banana fiber reinforced polyester (Pothan *et al*. 2003), flax- and hemp-fiber reinforced polypropylene (Wielage *et al*. 2003), ramie/glass hybrid fiber reinforced polyester composites (Romanzini *et al*. 2012) has been extensively studied in various applications. The results indicated that the storage modulus of the composites augmented with the increase of hemp fiber loading. The maximum damping ratio was achieved when the composites were reinforced with 30wt% noil hemp fiber (Etaati *et al*. 2014). In addition, Senthilvelan and Gnanamoorthy (2006) revealed that increasing the amount of short sisal fibre and short banana fibre in polyester composites might enhance their damping qualities. The importance of fibre in the damping behaviour of composites is growing. Moreover, damping composites comprised of high-performance fibre are utilized for a variety of purposes, such as sound absorption, energy storage, vibration control, *etc*.

Physical and Chemical Compatibility

Prestress will be formed at the interface if the thermal expansion coefficients of the NFr and the matrix are mismatched, particularly during the cooling process, affecting the performance of NFPCs (Lu and Oza 2013). The thermal expansion coefficients of the NFr and the matrix must be properly matched to avoid a thermal expansion mismatch between them (Krishnasamy *et al.* 2019). A coating can also be applied between the NFr and the matrix to act as a buffer zone, reducing the stress concentration induced by the thermal mismatch between the NFr and the matrix (Nurazzi *et al.* 2021). Chemical compatibility must be considered between the interface, NFr, and matrix; that is, the chemical composition of the NFr, interface, and matrix must be thermodynamically stable (Chawla 2008). TA Instruments Q400Ethermomechanical analysis equipment (TA Instruments, New Castle, DE, USA) was utilized to assess the matrix glass transition temperature, thermal expansion, and coefficient of natural fibre composites (Thomason *et al.* 2017).

Prevention or Inhibition of Oxidation

NFPCs are frequently subjected to oxidising conditions at elevated temperatures during their manufacturing or application. At high temperatures, interdiffusion between the matrix and the NFr and erosion in an oxidising environment significantly decrease the characteristics of the NFr (Matuana *et al.* 2011). Therefore, the IFB region should protect the NFr from damage caused by high temperatures. On the one hand, atomic diffusion and chemical reactivity between the NFr and matrix must be controlled, whereas erosion in an oxidising environment must be avoided (Fabiyi *et al.* 2008). In short, the resistance of NFr and even composites to high-temperature oxidation must be enhanced (Wang *et al.* 2005;

Matuana *et al.* 2011). The low thermal stability increases the likelihood of cellulose degradation and the likelihood of volatile material emissions, both of which could negatively impact the composite's characteristics. Therefore, processing temperatures are restricted to approximately 200 °C, but it is feasible to employ greater temperatures for brief durations. 60% of the thermal breakdown of most natural fibres occurred between 215 and 310 ˚C with an apparent activation energy of 160 to 170 kJ/mol (Yao *et al.* 2008). By exposing samples to a specified radiant flux, cone calorimeters are commonly used to evaluate the flammability of polymer compounds. A cone calorimeter measures heat release rate (HRR), total heat released (THR), mass loss rate (MLR), time to ignition (TTI), smoke emission (SEA - specific extinction area), and average carbon monoxide (CO) and carbon dioxide (CO_2). Lower HRR indicates smaller contribution to a fire (Stark *et al*. 2010; Zhang *et al*. 2012). Furthermore, thermogravimetric analysis (TGA) is utilized to study the thermal stability of NFr. A typical TGA curve for composite thermal degradability shows that a sample subjected to heat will slowly suffer weight drop, then the weight will drop sharply over a narrow range and finally turns back to zero slope as the reactant is exhausted. The shape of the TGA curve is determined by the kinetic parameters of the pyrolysis such as reaction order, frequency factor and activation energy while the values obtained depend upon atmosphere, sample mass, sample shape, flow rate, heating rate and the mathematical treatment used to evaluate the data (Azwa *et al.* 2013).

Appropriate Thickness of the Interface Layer

The IFB will be excessively strong if the interface layer is too thin, resulting in brittle fracture. If the interface layer is overly thick, then the IFB will be insufficiently robust, resulting in reducing the strength of NFPCs. It is possible to achieve a moderate bonding strength while preserving the strength and toughness of the composite by adjusting the thickness of the interface layer (El Jaouhari *et al.* 2018; Banerjee *et al.* 2019). Yu *et al.* (2011), fabricated SiC/SiC composites by coating SiC fibers with PyC layers with different thicknesses. When the thickness of the PyC interface layer is 0.1 μm, the flexural strength of the SiC/SiC composite reaches its maximum value, and the optimal thickness corresponding to the highest fracture toughness is 0.53 μm.

In general, the scanning electron microscope (SEM) is the mostly utilized technique for investigating fibre-matrix interactions at fracture surfaces and polymer distributions in plant fibre composites. It permits the monitoring of monomer-impregnated samples directly and after composites have cured, yielding information on the polymer-fibre interaction (George *et al*. 2001). Utilizing SEM, Migneault *et al*. (2015) examined the variation in wetting at the fibre-matrix interface of composites among the different fibres employed, including aspen wood, spruce bark, and spruce wood fibres. At the fibre-matrix interface, the SEM micrographs also revealed variations in interfacial adhesion and mechanical interlocking. The interpretation of the fibre-matrix adhesion is of special significance for the successful design and proper utilisation of NFPCs. There are several micromechanical testing methods for measuring the fibre-matrix interfacial adhesion. Examples include the single fibre fragmentation test (SFFT), the single fibre pull-out test (SFPT), and the microbond test (MT). The detailed summarisation of these tests can be found in the book of Kim and Mai (1998).

MECHANISMS OF INTERFACIAL BONDING

In NFPCs, the major components are the reinforcement NFr and the polymer matrix phases. Three key parameters influence the qualities and performance of composites: matrix, reinforcement, and interface. The interfacial region between the NFr and the matrix plays a significant role in determining the overall material behavior. In composites, the interface, which is frequently referred to as an intermediate area generated by the binding of the fibre and matrix, is a zone of compositional, structural, and characteristic gradients whose width typically ranges from a single atomic layer to micrometers (Zhou *et al.* 2016). At the interface, there is a strong link between the processes that take place at the atomic, microscopic, and macroscopic levels. Indeed, understanding the sequences of events that occur at these many levels is critical for comprehending the characteristics of IFB phenomena. The IFB between fibre and matrix is responsible for stress covey between the NFr and matrix and is primarily determined by the interfacial adhesion level. A sufficient interfacial strength guarantees that the maximum stress level is maintained without disturbance throughout the interface and from fibre to matrix. The molecular interaction at the interface, as well as the thickness and strength of the IFB region, impact the efficacy of load transmission (Drzal and Madhukar 1993; Awang 2013).

According to the interface definition, a fibre in NFPCs can be divided into two parts. One part does not come into contact with the matrix and retains the characteristics of the original fibres. The other part of the fibre is impacted by the matrix, resulting in characteristics that differ from the original fibre. This difference in the characteristics has two origins: first is the treatment of adsorbed materials on the fibre or matrix prior to composite formation. Functional groups can be added or removed from the initial surface of fibres using surface treatments. As a result, the surface of the fibre differs chemically and structurally from the original fibre. When the fibres or matrix are subjected to air prior being assembled, chemical species in the air may adsorb at the surface, altering/eliminating specific surface reactivity, resulting in a decrease in IFB (Drzal *et al.* 1983). The second reason is diffusion or chemical reactions between the fibres and matrix.

Physical Attraction Mechanism

The formation of an interface in composite materials includes physical attraction between electrically neutral bodies, molecular entanglement, inter-diffusion of elements, electrostatic adhesion attraction, chemical bonding reaction between groups on fibre and matrix surfaces, and mechanical interlocking (Drzal *et al.* 1983). In addition, there are forces with low energy, such as van der Waals forces and hydrogen bonding. Physical attractions (including electrostatic attraction and physical attraction between electrically neutral bodies) may occur before the NFr and matrix contact each other during the formation of IFB. Other mechanisms, such as molecular entanglement and interdiffusion, begin to operate after the NFr and matrix make contact. The final bond between the NFr and matrix is generated by all the mechanisms operating independently or, more likely, in concert (Kim and Mai 1998). Figure 6 depicts the individual bonding mechanisms.

Physical attraction between electrically neutral bodies leads in interfacial bonding on an atomic scale *via* electron interactions. This interaction is governed by van der Waal forces, which are the attraction between neutral molecules or atoms. It may also be determined by the acid-base interaction, *i.e.*, the polar attraction of Lewis acids and bases, such as electron-deficient and electron-rich elements (Ebnesaiiad and Ebnesajjad 2014). When the distance between the atoms of the fibre and matrix is within a few atomic diameters, the interaction occurs (Kim and Mai 1998). This physical attraction is commonly described in terms of wettability (the ability of liquids to spread over a solid surface) (Kim and Mai 1998).

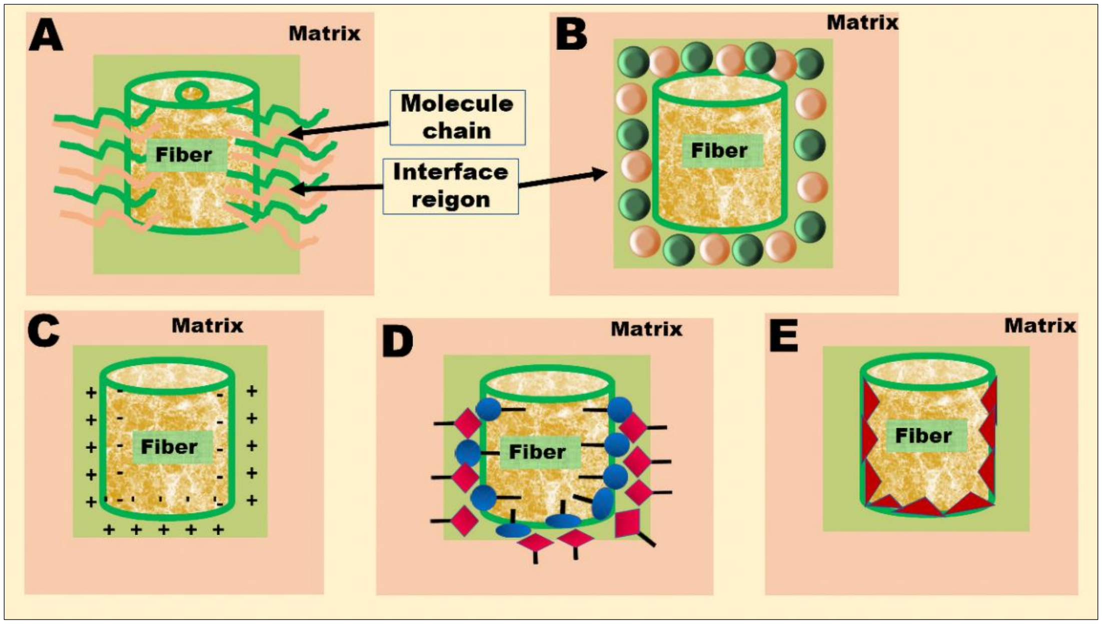

Fig. 6. Interfacial bonding formed by (a)molecular entanglement; (b) inter-diffusion of elements; (c) electrostatic attraction; (d) chemical reaction between groups on reinforcement and matrix surfaces; (e) mechanical interlocking

Interdiffusion Mechanism

Interdiffusion occurs as a result of intimate intermolecular interactions generated by Van der Waals forces or hydrogen bonding between the molecules of the NFr substrate and the matrix (Liu *et al.* 2012). Indeed, this adhesion mechanism consists of two stages: adsorption and diffusion. The first stage requires intimate interaction between two constituents, the NFr and matrix, guided by two actions: spreading and penetration. After adequate wetting, permanent adhesion is formed *via* molecular attractions such as covalent, electrostatic, and Van der Waals. On the other hand, enough wetness between the substrates results in the interdiffusion of NFr and matrix molecules. The extent and degree of diffusion are mostly determined by the chemical compatibility of the two ingredients and the penetrability of the substrate (Kim and Pal 2010). Despite the benefits, inter-diffusion can damage the NFr and lower the toughness of composites when exposed to a high-temperature oxidation environment. Under these conditions, various energy loss mechanisms such as fibre debonding, fibre bridging, fracture deflection, and fibre pull-out may impact the interface between the NFr and the matrix (Xu *et al.* 2021). The molecules that have diffused may become entangled with other molecules. The ambient temperature affects molecular entanglement (Kim and Mai 1998). As the temperature rises, the molecules in polymer chains gain sufficient energy to overcome local obstacles (such as molecule chains interpenetration) that obstruct molecular motion. When the temperature approaches the glass transition temperature, molecular motion becomes so active that even molecular entanglement is ineffective at preventing molecule chains from slipping (Sethi *et al.* 2015). As a result, the interfacial connection becomes weaker as the molecular entanglement is gradually unraveled.

Electrostatic Adhesion Mechanism

Electrostatic adhesion is a result of the formation of opposite charges (anionic and cationic) on the interacting surfaces of the NFr and matrix as a result of an electron or ion imbalance, as illustrated in Fig. 6(c); therefore, an interface region composed of two layers with opposite charges is produced, which accounts for the adhesion between the two components of the composite. The interfacial strength is reliant on the charge density (Kim and Mai 1998). When materials are separated, electrostatic attraction is efficient, and the active distance is in the centimeter range, which is greater than the range of other adhesion processes (Hays 1991). As previously stated, interactions based on van der Waal forces can only occur within a few atomic diameters. However, when substances are in close proximity, the electrostatic impact is relatively weaker compared to van der Waals forces (Petrie 2013).

Interestingly, electrostatic discharge treatments were applied to the surfaces of polymer and electrostatic fibres to produce electrostatic adhesion at the interface region, hence enhancing the performance of the composites (Rajak *et al.* 2019). In addition to this, the adsorption of particles on the surface of fibre through electrostatic interaction may also affect the surface adhesion between the fibres and matrix (Yamamoto *et al.* 2017; Yamamoto *et al.* 2016). Due to the aforementioned benefits, electrostatic adhesion was introduced into the fibre-reinforced composite structure (Heath *et al.* 2015, 2016) to increase the interfacial strength. As previously stated, the poor IFB between the fibre and matrix may limit the practical application of NFr. Thus, it is critical to optimise the matrix-fibre interaction mechanism to broaden the applications of fibre reinforced composites.

Chemical Bonding Mechanism

Chemical bonding occurs at the interface when compatible chemical groups are connected between the NFr and matrix. One common way of enhancing chemical bonding is by utilizing coupling agents, which are bifunctional molecules with one end of the molecule capable of reacting with the compatible chemical group on the fibres and the other end capable of reacting with the compatible chemical group in the matrix. Silane agents and maleated coupling agents are examples of traditional agents. For silane agents, one end of silanes can interact with hydroxyl groups (hydrophilic groups) of the NFr; while the other end interacts with the hydrophobic groups in the matrix (Li *et al.* 2007; Huda *et al.* 2008). The function of coupling agents is influenced by the environment when NFPCs with coupling agents are used in practical environments such as hygrothermal and UV temperatures. As a result, an appropriate coupling agent must be chosen based on the operating conditions of NFPCs (Yeh *et al.* 2021).

Mechanical Interlocking Mechanism

Mechanical interlocking happens between the matrix and rough surface of the natural fibre. The roughness of fibre surface may be defined by the regularity or irregularity valleys, and crevices of fibre surface (Liu *et al.* 2012). When the surface is rougher, the binding area between matrix and NFr augments, and thus the strength of bonding in interface region can be increased. Apart from the roughness of the NFr, the residual clamping stress induced by the differential in thermal expansion or shrinkage between the NFr and the matrix is also advantageous for the mechanical interlocking of the NFr and the matrix (Kim and Mai 1998). It occurs at the millimeter and micron length scales, while diffusion entanglement occurs at the nanoscale within the cell wall pores of natural fibre. By grafting NPs (*e.g.*, nano clay and nanocarbon tube) to fibres, the surface roughness of the NFr can be increased, hence improving mechanical interlocking. Alkali treatment is a frequently used technique for increasing the mechanical interlocking of NFPCs. Natural fibres contain fat and wax substances that smooth the fibre surface and have a detrimental impact on interlocking. Alkali solution can be used to eliminate fat and wax, thereby roughening the surface of the fibre. As a result, mechanical interlocking can be enhanced. Following alkali treatment, grafting NPs to the NFr surface can augment the roughness and wettability of the fibre surface, hence improving IFB. Zinc oxide NPs have been increasingly common in NFPC research because they possess NPs characteristics and exhibit high chemical reactivity due to their intrinsic functional groups, water repellence, and resistance to ultraviolet radiation (Mohammed *et al.* 2019). Inorganic NPs were integrated into the cell wall and likely occupied vacuum spaces (micropores) that would otherwise be exposed to water molecules, lowering the hygroscopic qualities of the modified natural fibres (Donath *et al.* 2004; Hill 2007).

On the other hand, during hydrolysis and polycondensation, the hydroxyl groups of cell wall constituents that are primarily responsible for moisture absorption were probably blocked by forming hydrogen bonds with inorganic NPs, which results in a decrease in hygroscopicity, significantly improves IFB, and further increases other mechanical characteristics, such as tensile strength. Mohammed *et al*. (2019) reported that zinc oxide nanoparticle-treated kenaf fibre-reinforced unsaturated polyester composites exhibited increased tensile strength compared to untreated composites. The modulus, Tensile strength, break at elongation, flexural modulus, flexural strength, and impact strength of the composites with higher content kenaf/ZnO nanoparticle are 560 MPa, 58MPa, 1.8%, 1300 PMa, 68 MPa, and 31 MPa, respectively. As a result of the amalgamation of zinc oxide nanoparticles, the mechanical characteristics and IFB were improved, and the contact angle results revealed an improvement in wetting of the fibres with the addition of ZnO nanoparticles. Notably, the presence of water molecules at the interface showed a significant effect on the mechanical interlocking. Water in the interface, acting as a lubricant, would exacerbate the slip between the fibre and matrix. On the other hand, the shrinkage and swelling ratios of the fibre and matrix are different. By increasing the moisture content, the residual clamping stress on the interface induced by the shrinkage differential between the fibre and matrix can be reduced (Liu *et al.* 2015).

Permeability is another fibre-related aspect that affects matrix penetration. Permeability varies according to the surface properties and direction, for example, tangential, radial, and longitudinal. This mechanical interlocking process is commonly employed in polymer composites by etching the polymer surface to augment surface roughness, hence augmenting the surface contact with the fibre and mechanical interlocking (Kim and Pal 2010). In contrast, an augment in mechanical interlocking results in the improvement of other bonding systems.

In general, the fibre-matrix interfacial bonding mechanisms include inter-diffusion, electrostatic adhesion, chemical reactions, and mechanical interlocking. These mechanisms work together to cause adhesion, and one of them is usually dominant. These particular phenomena occur predominantly in all almost natural fibr composites, but they can be minimised or reduced to minimal with a very good process (Zhou *et al.* 2016).

Besides the aforementioned risk, an additional risk of utilizing NFr is the poor compatibility between fibres and polymer matrix, which results in a non-uniform dispersion formation of fibres within the matrix, and thus generates poor interfacial bonding characteristics. Most polymers, especially thermosetting, are non-polar (hydrophobic, water repelling) substances, which are not compatible with polar (hydrophilic, water attracted) wood fibres, and therefore cause poor adhesion between the fibre and matrix interaction surfaces. That will be discuss in the next section (Mohd Nurazzi *et al*. 2017).

COMPATIBILITY BETWEEN NATURAL FIBRE AND SYNTHETIC POLYMER

The cellulose structure of the fibres is differentiated by crystalline and amorphous areas. The crystallite area forms a large number density of intramolecular hydrogen bonds, which are strong in comparison to van der Waals interactions. This results in the formation of a cellulose block, making it harder for other chemicals to penetrate. However, the amorphous region readily absorbs dyes and resins. The hydrophilic hydroxyl groups in this area combine with water molecules from the surrounding environment. Water molecules are generally held by hemicellulose, lignin, pectin, and waxy compounds. This results in the NFr being hydrophilic and polar, which reduces its compatibility with non-polar/hydrophobic matrixes (Mwaikambo and Ansell 2002; Abdelmouleh *et al.* 2007). Polymeric matrices are the most commonly utilised in NFPCs today since they are light weight and can be produced at low temperatures. Both thermoplastic and thermoset polymers have been employed for matrices with NFr (Holbery and Houston 2006). Matrix selection is constrained by the temperature at which natural fibres degrade. Most of the NFr utilized for reinforcement in NFPCs are thermally unstable above 200 °C, although under some circumstances it is possible for them to be processed at higher temperature for a short period of time (Summerscales *et al*. 2010). Due to this constraint, only thermoplastics that soften below this temperature such as polyethylene (PE), polypropylene (PP), polyolefin, polyvinyl chloride, and polystyrene and thermosets (which can be cured below this temperature) are useable as a matrix material (dos Santos *et al*. 2008). Indeed, PP and PE are the two most commonly adopted thermoplastic matrices for NFPCs. The main thermosets used are unsaturated polyester (UP), epoxy resin, phenol formaldehyde, and vinyl ester (VE) resins. This hydrophilic property impedes the effectively react with the matrix.

Additionally, pectin and waxy materials coat the reactive functional groups of the fibre, acting as a hindrance to matrix interlock. To dilate the crystalline region, remove hydrophilic hydroxyl groups, remove surface impurities (waxy substances), and improve the effectiveness of interfacial bonding, the surface of the fibre must be treated using various chemical treatments, reactive additives, and coupling agents. Chemical and physical modification of natural fibres is required (Dash *et al.* 2000).

MODIFICATIONS OF NATURAL FIBRE COMPOSITE TO IMPROVE INTERFACIAL ADHESION

High Energy Treatments

Without utilising chemical agents, physical treatments of natural fibres change their structure and surface characteristics, consequently affecting their mechanical interaction with the polymer matrix. Radiation and discharge treatments such as gamma radiation, corona, and plasma treatments are the most frequently utilized physical treatments in NFr composites for improving the functional properties and changing in the surface structure and surface energy of the NFr (Gassan and Gutowski 2000; Khan *et al.* 2009a).

Gamma radiation treatment

Gamma radiation treatment of composite materials is not a novel concept. The majority of research examined the effects of gamma radiation on the various characteristics of composite materials. Gamma radiation might be an excellent alternative due to several advantages, including reduced time requirements, uninterrupted operation, environmental friendliness, and design flexibility (Shubhra and Alam 2001; Khan *et al.* 2009b). Gamma radiation is powerful ionising radiation that can alter the internal structure of a material and decrease its hydrophilicity, resulting in improved crosslinking between the NFr and matrix (Khan *et al.* 2009c). Numerous scientists have investigated the effect of gamma radiation on composite materials. Khan *et al.* (2009b) studied the mechanical properties of jute fabric-reinforced polypropylene composites following gamma radiation treatment and reported that irradiated composites outperformed all other non-irradiated composites (Khan *et al.* 2009c). Hoque *et al.* (2017) investigated the influence of gamma radiation on the mechanical qualities of raw and polyethylene glycol-modified bleached jute-reinforced polyester composites and discovered that gamma radiation increased mechanical properties up to a point, then began to degrade them. Gamma and electron beam radiation was also used on a PAN carbon fibre-based composite by Jafari *et al.* (2016). They revealed that, by increasing the gamma and electron doses, the thermal behavior of the composites indicated a higher decomposition degree as a function of the temperature. Besides these, Martínez-Barrera *et al.* (2020) studied the effects of gamma radiation on the physicochemical properties of polyester resin. According to their findings, commercially available catalysts are insufficient for the complete polymerization of polyester resin. In contrast, gamma radiation can complete such activity, resulting in a high degree of cross-linking and morphological change on the surface when used as a matrix on a composite material.

Corona discharge treatment

Corona discharge treatment is an effective method for surface modification of cellulosic fibres used to fabricate composite materials. It is a cost-effective and environmentally friendly treatment that improves the NFMIF in composites. Corona discharge is an alternative technology for modifying the surface property of fibres. It is used to activate the surfaces of synthetic polymers (for example, polyethylene before printing) and woods (*e.g.*, graft polymerization of the vinyl monomer or ethylene imine to wood) (Uehara and Sakata 1990; Dong *et al.* 1993). Corona discharge is frequently utilized to enhance the wettability, adhesion, and hydrophilic properties of materials (Belgacem *et al.* 1994; Ooi *et al.* 2004) or to graft molecules on polymeric surfaces (Seto *et al.* 1999; Benhadi *et al.* 2011).

Compared with chemical activation, corona discharge activation of polymers (which includes lignocellulosic materials) has numerous advantages: exceptional efficiency and commercial feasibility (Mihailovic *et al.* 2011); environmental acceptability as a solvent-free, continuous process (Benhadi *et al.* 2011); capacity to modify surface properties without impacting bulk properties (Lei *et al.* 2000), can be conducted in a variety of atmospheres (Benhadi *et al.* 2011), and allows for the grafting of a wide variety of species (Bataille *et al.* 1994; Bataille *et al.* 1991). Treatment variables such as time and current determine the degree of improvement. Sakata *et al.* (1993), demonstrated that increasing the degree of corona treatment reduces the contact angle of a droplet of urea-formaldehyde resin on a wood surface. Improved wettability was said to contribute to increased binding strength in that situation.

The treatment induces oxidation on the surface of the specimen in the presence of oxidizing agents such as atomic oxides, oxygen-free radicals, and ozone. Due to oxidation, the surface of the specimen is cleansed while also increasing its mechanical characteristics (Tuominen *et al.* 2010). Corona treatment improves the bonding and compatibility of fibres and fillers, as Gholshan Tafti *et al.* (2018) concluded in their study. Tuominen *et al.* (2010) found that corona treatment improved the oxidation level and surface energy of the specimen. After the corona treatment, Gassan and Gutowski (2000) found fibre and matrix compatibility and specimen surface oxidation increased.

Plasma treatment

Plasma treatment is another effective physical approach for enhancing the surface characteristics of NFr and polymeric materials by harnessing high-energy photons, electrons, ions, radicals, and excited species. Plasma treatment introduces polar or excited groups onto the fibre surface or grafts a new polymer layer onto the fibre surface, allowing for strong covalent bonds between the NFr and the polymer matrix. Additionally, this process roughens the fibre surface, improving mechanical interaction between the fibres, polymer matrix, and IFB (Yuan *et al.* 2002). The alteration of NFr by treatment in cold oxygen plasma produced from a corona discharge under ideal operating circumstances transforms the fibre into a semi-active filler for the composite (Vladkova *et al.* 2004, 2006). As a result, the adhesion at the IFB region is augmented with the plasma treatment. The consequence is an enhancement in the mechanical characteristics of the composites (such as flexural strength and modulus, tensile strength, and modulus). Nevertheless, the fibre may degrade over a prolonged exposure time because of the continual effect of particles on the surface, which eventually reduces the IFB between NFr and the matrix (Morales *et al.* 2006; Oporto *et al.* 2009).

When subjected to electric field energy, free electrons in the plasma atmosphere begin colliding with neutral gas molecules and transfer energy. This results in the production of reactive species, which are predominantly made up of ions. By connecting these excited ions to the solid surface, the chemical and physical modification on the surface of the material is established. When treated with plasma, the NFr does not change its bulk properties; it only modifies its surface characteristics (Sun 2016).

Macedo *et al.* (2019) used cold plasma to treat kapok fibres and found changes in the interfacial adhesive characteristics of the polyethylene reinforced kapok fibre composite. After treatment, it was noticed that the mechanical strength of the specimen was augmented, while its thermal stability decreased. Gibeop *et al.* (2013), investigated the effects of plasma treatment on the mechanical parameters of a jute fibre composite reinforced with polylactic acid (PLA). It was revealed that following treatment, the strength of the specimen rose due to the plasma treatment's "heating and etching action," which also increased adhesion by roughening the surface of the fibre. Yuan *et al.* (2004), examined the change in mechanical characteristics of polypropylene composite reinforced by wood fibre. Increased interfacial adhesion and viscoelastic dissipation were seen due to the roughening effect on the fibre surface. Fazeli *et al.* (2019), studied the effect of atmospheric pressure plasma treatment on the interfacial adhesion of starch matrix and cellulosic fibre. It was observed that after plasma treatment at low pressures, the tensile strength of the specimen rose dramatically due to improved interfacial adhesion between matrix and fibre.

The primary impacts of plasma treatment on NFr are as follows: Cold plasma treatment can improve the mechanical properties of natural fibres. A considerable augment in tensile strength can be noticed when treated with low pressure air plasma, owing to the increased interfacial adhesion between the NFr and matrix. Surface modification by oxygen plasma treatment results in a significant increase in qualities such as wettability, toughness, compatibility, and IFB. Physical approaches do not alter the chemical composition of fibres, but they do change their surface and structural qualities. Table 1 summarises the significant impacts of different physical treatments on NFPCs.

Table 1. Impact of Physical Treatment on Natural Fiber Composites

Treatment	Fiber/ polymer	Result	Reference
Gamma irradiation	Basalt fibre/epoxy Polymer	During the irradiation process, polymer chain scission and oxidation were induced on the surface and within the resin matrix, resulting in an increase in interlaminar shear strength while maintaining the tensile property of the irradiated composite.	(Li *et al*. 2015)
UV radiation	Jute fiber/PE/PP	The tensile and bending strength of the composite increased with UV radiation strengthening up to 50 radiation doses; compared to the untreated composite, the treated composite rose by 18% tensile strength and 20% bending strength, respectively.	(Zaman *et al*. 2009)
Corona treatment	*Miscanthus* fibre/PP and *Miscanthus* fibre/PLA composites	The corona treatment of fibres caused surface oxidation and etching, which led to better compatibility between the fibres and the matrices. As a result, the mechanical and thermal properties of the treated composites (Young's modulus, stress at yield, glass transition temperature, and decomposition temperature) were greatly improved.	(Ragoubi *et al*. 2012)
Plasma treatment	Flax fibre/polyester composite	The tensile strength, flexural strength, flexural modulus, and interlaminar shear strength of flax fiber-reinforced polyester composites increased by 34%, 31%, 66%, and 39%, respectively, after air plasma treatment at a plasma power of 300 W, owing to improved adhesion between the treated fibre and polyester matrix.	(Sarikanat *et al.* 2016)

Chemical Treatments

Natural fibres are hydrophilic due to their lignocellulosic composition, which contains highly polar hydroxyl groups. As a result, these fibres are fundamentally incompatible with hydrophobic thermoplastics such as polyolefins. The major limitations of employing these fibres as reinforcements in such matrices include poor interfacial adhesion between polar-hydrophilic fibres and non-polar hydrophobic matrix, and difficulties in mixing due to poor wetting of the fibres with the matrix. Therefore, it is imperative that NFr should be subjected to chemical modification to increase the compatibility and adhesion between NFr and matrix (John and Anandjiwala 2008). It would also be desirable that the chemical used for the modification of NFr preserves the biodegradable nature of NFr. Ideally, the chemicals utilised for modification should be derived from sustainable sources as well (Kabir *et al*. 2013).

Natural fibers have also been treated with various chemicals such as alkali, silane, water repelling agents, peroxides, permanganates, *etc*. It has been observed that some of these chemical treatments (for example, alkali treatment) can significantly enhance the mechanical characteristics of NFr by modifying their crystalline structure, as well as by removing weak components such as hemicelluloses and lignin from the fiber structure (Li *et al*. 2007). Also, moisture absorption and subsequent swelling of NFr can be reduced through selective chemical treatments (*e.g*., water repelling agents). Furthermore, chemical treatments (for example, using silane coupling agents) can increase the fibre-matrix interfacial interactions by forming strong chemical bonds, resulting in a significant improvement in composite mechanical performance (Xie *et al*. 2010).

Natural fibers are amenable to chemical surface treatment due to the presence of hydroxyl groups. The hydroxyl groups may participate in hydrogen bonding inside cellulose molecules, activating these groups, or they may introduce new moieties that form effective interlocks within the system. Chemical modification can increase surface properties such as wetting, adhesion, surface tension, and fibre porosity. The irregularities of the fiber surface play an important role in the mechanical interlocking at the interface. The interfacial qualities can be improved by modifying the components, which results in changes in physical and chemical interactions at the interface (John *et al*. 2008; John and Anandjiwala 2008).

Cao *et al*. (2006) examined the mechanical characteristics of bagasse fiber-reinforced polyester composites before and after alkali treatment. NaOH solutions of 1, 3, and 5% concentrations were employed. Superior properties were obtained for composites made from 1% NaOH-treated bagasse fibers. The authors observed a 13% improvement in tensile strength, 14% in flexural strength, and 30% in impact resistance, respectively, due to chemical modification. Researchers have reported the utilized of benzoic acid for chemical modification of bagasse fibers in polyvinyl chloride composites (Zheng *et al*. 2007). It was found that the tensile strength of modified composites rose by around 35 %, indicating improved reinforcing. The tensile strength of composites containing untreated fibres was 38 MPa, while that of composites with chemically modified fibres was 52 MPa.

Xue *et al*. (2007) investigated the influence of the maleic anhydride grafted polypropylene (MAPP) coupling agent on the mechanical characteristics of aspen fiber-reinforced polypropylene (PP) composites. The coupling agent enhanced the adhesion between fiber and matrix and augmented the tensile characteristics of the composites. The tensile strength of the composites containing compatibilizer increased by 15% and flexural strength registered an increase of 40%. The greater interfacial adhesion was also evident. In an interesting study, Teramoto *et al*. (2004) studied the biodegradation of aliphatic polyester composites reinforced by abaca fiber. Abaca fibers, chemically modified with acetic anhydride, showed greater resistance to biodegradation due to the existence of a protective covering on the fibers.

Alkaline treatment

When natural fibres reinforce thermoplastics and thermosets, alkaline treatment or mercerization is one of the most often utilized chemical treatments. The most significant modification caused by alkaline treatment is the disruption of hydrogen bonding in the network structure, hence increasing the roughness of the surface (Alnaid *et al.* 2018; Dahham *et al.* 2018). This treatment eliminates a certain amount of lignin, wax, and oils that cover the external surface of the fibre cell wall, depolymerizes cellulose and exposes the short length crystallites (Mohanty *et al.* 2001). The sensitive hydroxyl groups (OH) are degraded throughout the treatment and hence react with water (H-OH), leaving the ionized reactive molecules to produce alkoxide with NaOH (Eq. 2) (Li *et al*. 2007; Huda *et al*. 2008).

$$\text{Fibre-cell-OH} + \text{NaOH} = \text{Fibre-cell-O-Na} + H_2O + \text{Impurities} \qquad (2)$$

As a result, the surface of the fibre becomes clean. As long as the matrix material distributes well over the cellulosic material, this is likely to expose more microvoids in the cellulosic material, which can be conducive to bonding. In other words, the removal of hydrophobic substances from the surface can result in a rougher surface, hence enhancing the stress transmission capacity between the ultimate cells. Furthermore, it reduces the diameter of the fibre and hence increases the aspect ratio (length/diameter). This increases the effective surface area of the fibre, which is necessary for optimal adherence to the matrix (Joseph *et al.* 2003; Alnaid *et al.* 2018; Dahham *et al.* 2018). The ionization of the hydroxyl group to the alkoxide is promoted by adding aqueous sodium hydroxide (NaOH) to NFr (Agrawal *et al.* 2000). Thus, alkaline processing directly affects the cellulose fibrils, the degree of polymerization, and the extraction of lignin and hemicellulosic compounds (Jahn *et al.* 2002). Alkaline treatment has two impacts on the fibre: (1) it enhances surface roughness, resulting in improved mechanical interlocking, and (2) it increases the amount of cellulose exposed on the fibre surface, hence increasing the number of potential reaction sites (Valadez-Gonzalez *et al.* 1999). This treatment dramatically improves the mechanical and thermal properties of the NFPCs. Excess delignification of the fibre can occur if the alkali concentration is higher than the recommended level, resulting in weakening or damage to the fibres (Li *et al.* 2007; Wang *et al.* 2007; Dahham *et al.* 2018). A schematic representation of the cellulose fibre structure before and after an alkali treatment is shown in Fig. 7. Treated fibres have lower lignin content, partial removal of wax and oil cover materials and distension of crystalline cellulose structure.

Cai *et al*. (2016) demonstrated that a 5% alkaline treatment enhanced the mechanical characteristics of abaca fibre reinforced epoxy composites. The 5 wt% alkali treatment increased the tensile strength and young’s modulus of composites by 8% and 35% respectively as compared to untreated abaca fibers. Also, Vilay *et al*. (2008), carried out a study on the effect of chemical treatment of sodium hydroxide (NaOH) and fiber loading on mechanical properties of bagasse fiber reinforced polyester composites and and concluded that the higher tensile and flexural values were attained for treated fiber composites compared to those of untreated fiber based composites. The influence of alkali treatment on mechanical characteristics of bagasse fiber reinforced polyester composites was investigated by Cao *et al*. (2006). From this study, the authors found that the alkali treatment improved the tensile strength by 13%, flexural strength by 14%, and impact energy by 30%, respectively.

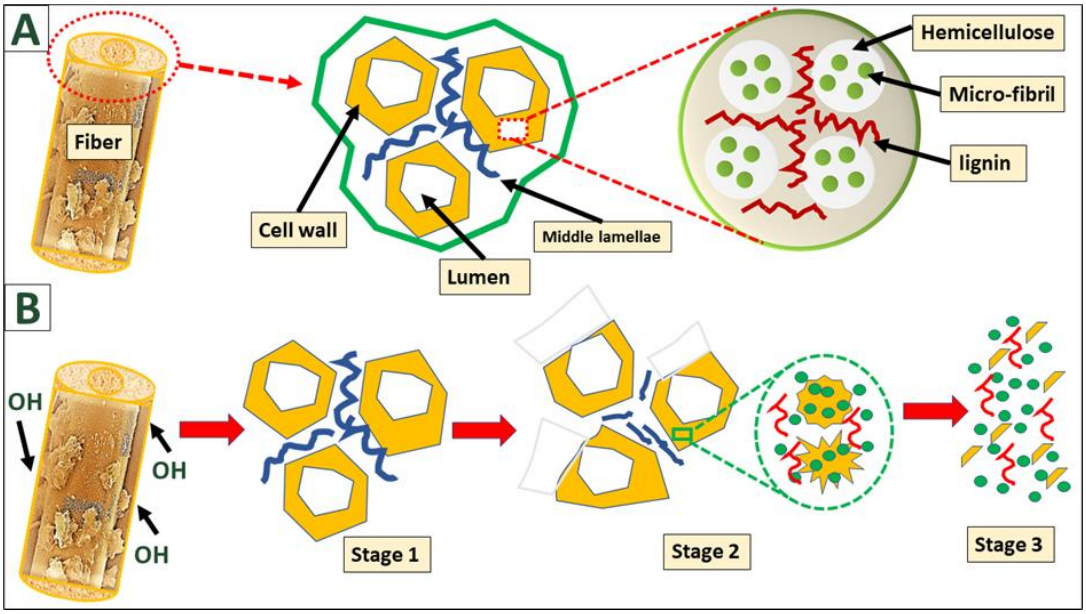

Fig. 7. The schematic drawing of the hierarchical structure of natural fibres (a); the alkaline degradation process of natural fibres (b)

Silane treatment

The word silane, which literally has the chemical formula SiH_4, also can be used to represent a family of reagents that can be used to modify cellulosic surfaces. Thus, silanes have been utilized as coupling agents to treat the surface of NFr. A typical silane coupling agent contains two reactive groups: one end with alkoxysilane groups reacts with hydroxyl-rich surfaces, such as wood or other NFr, while the other end interacts with the polymer matrix, allowing the NFr to adhere to the polymer matrix and thus stabilize the composite material (Zhou *et al.* 2015). Silane coupling agents may lower the number of cellulose hydroxyl groups in the IFB between NFr and matrix.

Trialkoxysilanes have received much attention as effective and practical agents for modification of lignocellulosic materials (Szlek *et al*. 2022). In the presence of moisture, the hydrolysable alkoxy group leads the production of silanols. The silanol then interacts with the hydroxyl group of the fibre, generating stable covalent bonds to the cell wall that are chemisorbed onto the surface of NFr. The fourth substituent attached to the silicon atom in a trialkoxysilane is often selected to be an alkyl chain (Szlek *et al*. 2022). As a result of the diffusion of hydrocarbon chains into the matrix, the hydrocarbon chains generated by the application of silane restrict the swelling of the fibres by establishing an entangled network (Dhakal *et al.* 2007). It was hypothesised that the hydrocarbon chains generated by the application of silane affected the wettability of the NFr, hence enhancing the chemical affinity of the polymer matrix.

Silane treatment boosts the tensile strength of the NFPCs, decreases the impact of moisture on the characteristics of the NFPCs, and augments adhesion, and thereby the strength of NFPCs (George *et al.* 2001; Bogoeva-Gaceva *et al.* 2007; Li *et al.* 2007). The surface of NFr has micro-pores, and silane coupling agents are surface coatings. This penetrates into the pores and develops mechanically interlocked coatings on fibre surfaces. Silane-treated fibre composites have been found to have better tensile strength characteristics than alkali-treated fibre composites (Valadez-Gonzalez *et al.* 1999). The most commonly used silanes are amino, methacryl, glycidoxy, vinyl, azide, and alkylsilanes. During the silane treatment of natural fiber as shown in the Fig. 8, the hydrolysis of alkoxy groups on silane takes place to form silanol (Si–OH) groups, which can then react with hydroxyl groups on the fiber surface (Pickering *et al*. 2016).

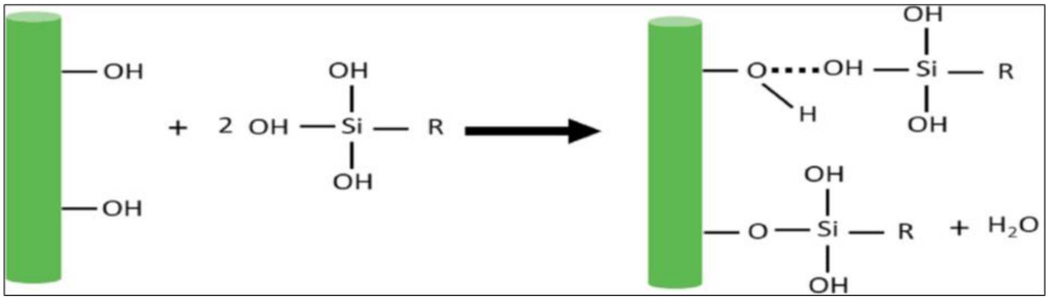

Fig. 8. Reaction of silane with NFr (R representing organic group, … representing hydrogen bonding) (Islam *et al.* 2021; Creative Commons Attribution (CC BY 4.0))

In a study by Seki (2009), the impact of oligomeric siloxane treatment of jute fabrics on mechanical characteristics of jute/epoxy and jute/polyester composites was studied. At first, the jute fabrics were treated with 5% (w/w) NaOH solution for 2 h, and then the alkali-treated jute fabrics were treated with 1% siloxane. Hand lay-up was used to make the jute/epoxy and jute/polyester composites. The mechanical characteristics such as tensile, flexure, and interlaminar shear strengths of the siloxane treated composites were increased by ~32%, ~22%, and ~109% for jute/epoxy composite and ~31%, ~37%, and ~103% for jute/polyester composite compared to untreated jute fiber composites. In the work of Wang *et al*. (2020), the surface of the bamboo fibers (BF) was treated with three kinds of silane coupling agents terminated with amino functional groups (KH550), epoxy functional groups (KH560), and methyl functional groups (KH570) to enhance IFB. The effects of silane treatment on the mechanical characteristics and thermal behavior of BF/polypropylene (PP) composites were examined. Mechanical test results showed that the order of modification effectiveness was KH570 > KH550 > KH560. KH570-treated fiber composite exhibited the best mechanical characteristics. The tensile strength and flexural strength of 5 wt% KH570 treatment reached to 36.1 and 54.7 MPa, which were 15.4% and 23.6% higher than those of UBF/PP composites.

Chemical coupling agents are often molecules with dual functionalities. The first function is to react with hydroxyl groups of cellulose and the second function is to react with functional groups of the matrix. Bledzki and Gassan (1999), outlined several mechanisms of coupling in materials, namely: (a) elimination of weak boundary layers; (b) creation of a strong and malleable layer; (c) The formation of a strongly crosslinked interphase area with a modulus intermediate to that of the substrate and that of the polymer; (d) improvement of the wetting between polymer and substrate; (e) formation of covalent bonds with both materials; and (f) alteration of acidity of substrate surface. Figure 9 shows the coupling agent (*i.e.*, silane) mechanism between the hydrophilic fiber and hydrophobic polymer matrix. In this case, the compatibilizing agent is chemically bonded to the fiber and blended by wetting in polypropylene (PP) polymer chain (Arjmandi *et al*. 2017). As compatibilizer polyethylene-octene grafted maleic anhydride (POE-g-MAH) demonstrated faster mixing and better dispersion when blended with PP compared with conventional polyolefin elastomers (Lim *et al*. 2008; Bai *et al*. 2004). It is interesting to note that POE-g- MAH acts as compatibilizer by forming hydrogen bond between maleic anhydride group of POE and hydroxyl group of fibers.

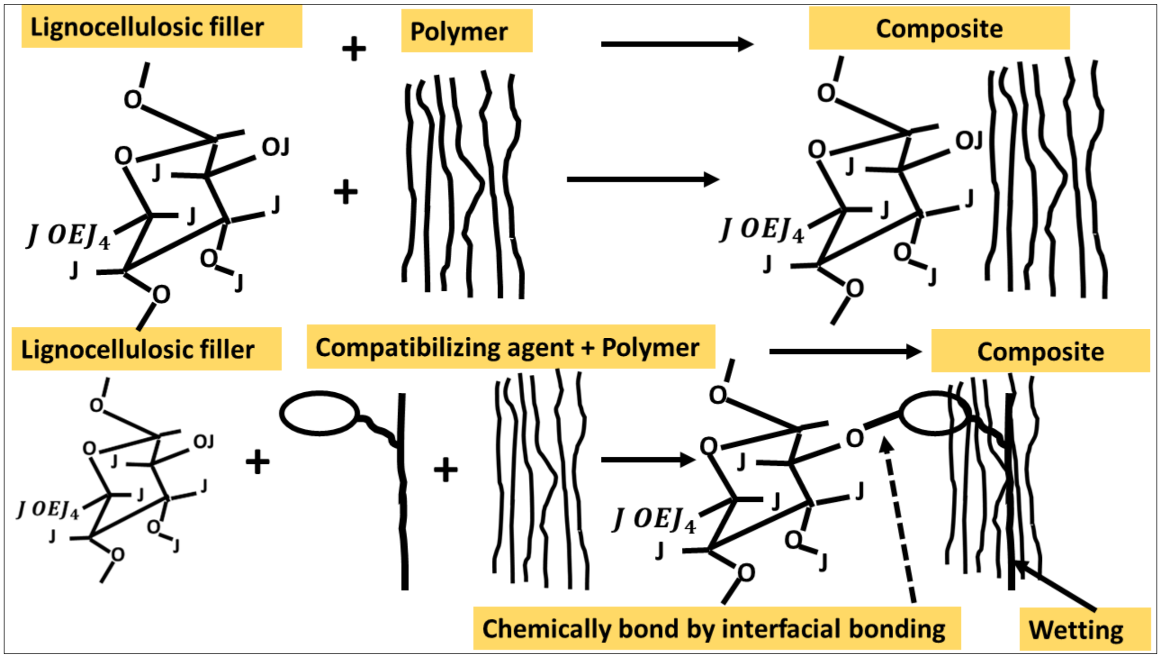

Fig. 9. Mechanism of compatibilizing agent between hydrophilic filler and hydrophobic matrix polymer (Figure redrawn and credit to Arjmandi *et al.* 2017)

Acetylation treatment

The acetylation of NFr is a well-known esterification method of adding plasticization to cellulose fibres. Acetylation has been widely used to stabilize NFr, enhancing its dimensional stability and resistance to environmental degradation. Acetylation is a process that occurs when the hydroxyl groups in the cell walls of lignocellulosic materials react with acetic at elevated temperatures (usually without a catalyst) (Dhakal *et al.* 2007). Acetic acid (product from the reaction) must be eliminated before the fibres can be utilized. The hydroxyl groups of lignin and hemicelluloses (amorphous material) react with the reagent, but the hydroxyl groups of cellulose (crystalline material) are firmly packed with hydrogen bonds, inhibiting diffusion of the reagent and resulting in comparatively modest reaction rates. Esterification has been found to enhance the dispersion of NFr in a polymer matrix as well as the dimensional stability and IFB between NFr and matrix of the final composites (Bogoeva-Gaceva *et al.* 2007; Li *et al.* 2007).

Acid catalysts such as sulphuric acid and acetic acid are frequently employed throughout the treatment to speed the reaction and optimize the degree of acetylation (Bledzki *et al.* 2008; Kabir *et al.* 2012). The esterification reaction not only stabilizes the cell walls, particularly in terms of moisture absorption and subsequent dimensional change of NFr, but it also reduces the void content of the fibre rough surface topography, thereby improving adherence of NFr to the matrix (Sreekala *et al.* 2002; Haseena *et al.* 2007). According to some reports, acetylation of NFr can increase the resistance of NFr to moisture absorption by up to 65%, depending on the degree of acetylation (Bledzki and Gassan 1999; Bledzki *et al.* 2008). Most significantly, compared to untreated NFr reinforcing composites, this esterification of NFr improves the stress convey efficiency at the interfacial region and the mechanical characteristics of its composites (Joseph *et al.* 2005). Additionally, the greater hydrophobicity of the treated fibres enabled the composite to have a larger volume resistivity than untreated composites by lowering the dielectric constant of the NFPCs (Haseena *et al.* 2007).

Rowell *et al*. (1996), investigated acetic anhydride treatment on different types of NFr to analyze the impacts of equilibrium moisture content and reported improved moisture resistance characteristics. This was due to the removal of hemicellulose and lignin constituents from the treated fibre. In a fascinating study, Mishra *et al*. (2003), employed acetic anhydride treatment (with glacial acetic acid and sulphuric acid) on alkali pre-treated (5% and 10% NaOH solution for 1 h at 300 °C) dewaxed sisal fibre and revealed, improved IFB characteristics of the composites. In addition, Bledzki *et al*. (2008), studied different concentration of acetylation treatment on flax fibre and reported 50% higher thermal characteristics. Moreover, 18% acetylated flax fibre polypropylene composites showed 25% higher tensile and flexural properties compared to the untreated fibre composites.

Benzoylation treatment

Benzoylation modification employs benzoyl chloride to reduce the hydrophilicity of the NFr and increase interfacial adhesion between NFr and matrix, enhancing the strength of the NFPCs. It also improves the thermal stability of the NFr (Nair *et al.* 2001; Li *et al.* 2007). During benzoylation treatment, alkali pre-treatment is utilized. Extractable components such as lignin, waxes, and oil-covering compounds are eliminated at this stage, exposing more reactive hydroxyl (OH) groups on the fibre surface. Following that, benzoyl chloride is applied to the fibres. The OH groups of the NFr are then replaced with benzoyl groups ($C_6H_5C{=}O$), which are connected to the cellulose backbone. This increases the hydrophobicity of the NFr and enhances adherence to the matrix. Joseph *et al*. (1996), applied benzoyl chloride treatment on alkali pre-treated sisal fibre and reported higher thermal stability compared to the untreated fibre composites. Similar treatment was carried out on flax fibre reinforced in low density polyethylene composites by Wang *et al*. (2007). The claimed outcome was a 6% augmentation in tensile strength and a 33 % increase in moisture resistance.

Peroxide treatment

Interface characteristics of NFr and matrix can be enhanced by peroxide treatment. The peroxide-induced grafting of polyethylene adheres to the NFr surface. Furthermore, peroxide-induced free radicals interact with the hydroxyl group of NFr as well as the matrix. Therefore, fibre matrix adhesion along the interface is enhanced. Additionally, this treatment decreases the moisture uptake tendency of the NFr and increases its thermal stability (Kalaprasad *et al.* 2004; Wang *et al.* 2007). Sapieha *et al.* (1990) described the pre-treatment of fibres with benzoyl or dicumyl peroxide, resulting in improved mechanical characteristics of the NFPCs. The peroxide treatment process begins with an alkali pre-treatment of the NFr, followed by 30 min of coating with benzoyl peroxide or dicumyl peroxide (about 6% concentration) in acetone solution. Peroxide can be decomposed by heating the solution to a higher temperature (Kalaprasad *et al.* 2004; Li *et al.* 2007). Joseph *et al.* (1996), evaluated the optimal concentrations of benzoyl peroxide (6%), and dicumyl peroxide (4%) treatments on short sisal fibre reinforced polyethylene composites and observed an improvement in tensile strength characteristics. Sood *et al.* (2015) investigated the impact of chemical treatments on tensile and flexural characteristics of polymer composites fabricated by reinforcing treated sisal fibers into 50-50 mixture of fresh and recycled HDPE matrix. The sisal fibers are treated with NaOH + Maleic Anhydride (MA) and NaOH + Benzoyl Peroxide (BP) + MA. The effect of chemical surface treatments on mechanical characteristics of composites is compared at two different contents (7.5 wt% and 30 wt%) of treated sisal fibers. It was observed that the composites containing 30 wt% of NaOH + BP + MA treated sisal fiber showed higher tensile strength than that noticed for NaOH + MA treated sisal fiber composites. However, at lower fiber content of 7.5 wt%, both the treated fibers composites exhibit almost comparable tensile strength. When compared to the equivalent composites with the same amount of NaOH + MA treated sisal fibres, the flexural strength of NaOH + BP + MA treated sisal fibres was significantly higher at 30 wt% sisal fibres but relatively lower at 7.5 wt% sisal fibres.

Maleated coupling agents

Maleated coupling agents is a highly effective method for enhancing the interfacial interactions between NFr and the matrix. Maleic anhydride groups interact with the hydroxyl groups in NFr, removing them from the fibre cells and thereby decreasing their hydrophilicity. Furthermore, the maleated coupler generates a C-C bond with the polymer chain (George *et al.* 2001). The strongest adhesion is produced when covalent bonds are established at the interface between NFr and the coupling agent, and molecular entanglement between the coupling agent and the matrix (Huda *et al.* 2006). The reaction mechanism of the coupling agent with NFr and matrix can be described by activating the copolymer before fibre treatment and subsequently esterifying the NFr.

Table 2. Comparison between Chemical Treatments

Chemical Treatment	Its Impact on the Natural Fibers
Alkaline Treatment	(1) It improves mechanical interlocking by increasing surface roughness; (2) It increases the amount of cellulose exposed on the fiber surface, hence increasing the number of possible reaction sites; (3) It eliminates impurities, non-cellulose natural fiber such as lignin, hemicellulose, pectin, wax, *etc*. Consequently, alkaline treatment has a lasting impact on the mechanical behaviour of natural fibers, especially on their strength and stiffness.
Silane Treatment	(1) It prevents fibre swelling; (2) It improves interfacial adhesion between fibres and the polymer matrix; and (3) It boosts water absorption resistance.
Acetylation Treatment	(1) It enhances the dispersion of fibres in a polymer matrix as well as the dimensional stability and interface of the final composites; (2) It enhances the stress transfer between natural fibre and matrix.
Benzoylation Treatment	(1) It improves thermal stability; (2) It improves fiber-matrix adhesion; (3) It raises the strength; and (4) It decreases water absorption.
Peroxide Treatment	(1) It reduces the ability of natural fibres to absorb moisture; (2) It increases the thermal stability of the fibres.
Maleated Coupling Agents	(1) It increases the mechanical characteristics; (2) It improves of interfacial bonding between fibers and polymer matrix

This treatment lowers the surface energy of NFr to a level considerably closer to that of the matrix, resulting in increased wettability and improved IFB between the NFr and matrix (Li *et al.* 2007). During the reaction mechanism of maleic anhydride, polypropylene (MAPP) and the cellulose fibre. MAPP copolymer is activated at a temperature of 170 °C and then the esterification of the cellulose fibres is carrying out. This treatment improves the wettability of the fibre and increases interfacial adhesion (Keener *et al*. 2004). Mohanty *et al*. (2004) utilized jute fibre (30% fibre loading, 6 mm on fibre length) treated with 0.5% MAPP concentration in toluene for 5 min and reinforced in PP matrix. The composite showed around 72% higher flexural strength characteristics compared to the untreated fibre composites. The water absorption tendency of the treated fibre composite was also reduced. Similar studies were performed by Mohanty *et al*. (2004), who reported that 1%MAPP treated sisal fibre–PP composite exhibited around 50%, 30%, and 58% higher tensile, flexural, and impact strength characteristics respectively. Significant effects of these chemical treatments on NFPCs are summarized in Table 2.

Coating on Natural Fibres

A coating can be defined as a substance (often a liquid) applied to a surface and then dried as a continuous or discontinuous coating. However, the application process and the resulting dry film are also considered coating (Jones *et al.* 2017). Evaporation or curing (cross-linking) by oxidative, thermal, or ultraviolet light, as well as other available methods, are used to dry the liquid coating. Coatings occur in both organic and inorganic forms. Organic coatings are mainly employed for ornamental and functional objectives, whilst inorganic coatings are primarily used for protective purposes (Jones *et al.* 2017). Typically, coatings are applied in multi-layered systems comprised of a primer and a topcoat. Each coating layer is applied to accomplish a distinct task, while its activities are influenced by the activities of the other layers in the system. Interactions between different layers and interfacial phenomena all contribute significantly to the overall performance of multi-coat systems (Hegedus 2004). Various properties of coatings are typically associated with specific parts of a coating system, as shown in the Fig. 10 (Verkholantsev 2003).

Coatings are typically applied to surfaces for decorative, protective, or functional purposes, though they are frequently used in combination. The phrase "functional coatings" refers to systems that provide additional functionality in addition to the traditional features of a coating (*i.e.*, ornamentation and protection) (Wulf *et al.* 2002). This added functionality can take on a variety of forms and is dependent on the application of the coated substrate. Typical examples of functional coatings are self-cleaning (Nun *et al.* 2002; Parkin and Palgrave 2005), easy-to-clean (anti-graffiti) (Kuhr *et al.* 2003), antifouling (Perez *et al.* 2003), soft feel (Zhou and Koltisko 2005), and antibacterial (Tiller *et al.* 2001; Ferraris *et al.* 2010). Although numerous techniques and applications are involved, there is a common element that is advantageous and meets the needs of some users. Apart from their unique features, functional coatings frequently have to meet other standards; for example, nonstick cookware coatings must be scratch, abrasion, and temperature resistant. Typical expectations of functional coatings include:

- Durability
- Reproducibility
- Easy application and cost effectiveness
- Tailored surface morphology
- Environmental friendliness

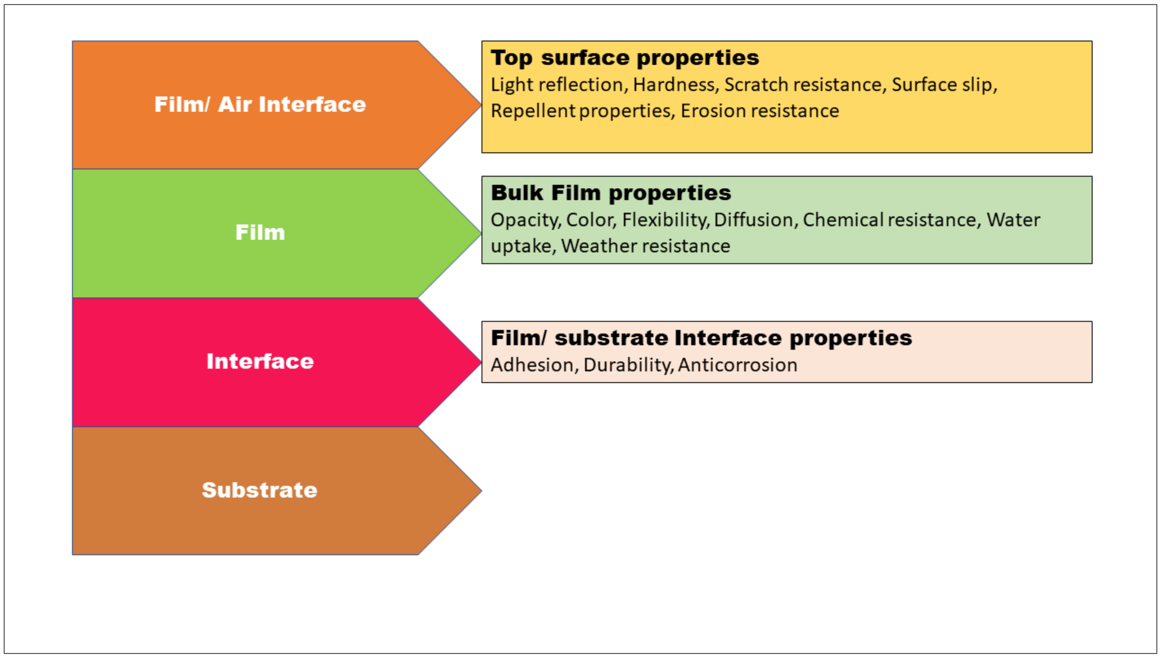

Fig. 10. Topographical classification of coating properties

NFr composites have applications in a wide variety of study disciplines. Consequently, much development has been accomplished in the past on NFr and their composites. However, one of the most important disadvantages and obstacles associated with NFr is their hydrophilic nature (Ali *et al.* 2018). Additionally, the high moisture absorption rate of NFr precludes their use in open environments due to the significant swelling of the NFr. As described in the following section, multiple surface coatings and chemical modifications have been performed to the surface of NFr to alleviate these disadvantages and expand their application possibilities.

Polymer Coatings on Natural Fibres

Some researchers have also attempted the polymer coating on NFr to boost their hydrophobicity as the outcome of good IFB between NFr and matrix, hence increasing the mechanical properties of the bio-composites. Rodriguez and Francucci (2016), demonstrated that vinyl ester-based alkali-treated jute composites combined with PHB-coated jute composites exhibited the best mechanical and water resistance properties. Gupta and Singh (2019) addressed the constraints of jute composites by treating them with alkali and covering them with polylactic acid (PLA). It was suggested that composites composed of treated and coated fibres displayed the best mechanical, thermal, and water resistance property enhancements. Another study examined the mechanical, thermal, and water absorption properties of PLA-coated sisal/polyester composites and concluded that coating fibres improves the properties of their composites (Gupta 2020). Mokhothu and John (2017) coated NFr with a bio-based coating composed of polyfurfuryl alcohol resin and polyurethane to improve the water resistance and mechanical performance of their phenolic resin-based biocomposites. The effect of PLA coating on the static and dynamic mechanical characteristics of polyester reinforced with PLA coated sisal fibres was investigated (Sahu and Gupta 2018).

Flax fibre is a significant NFr formed from the bark of the stems of subtropical plants. Natural flax fibre is a cellulosic polymer. However, its structure is more crystalline, which increases the hardness and tensile strength of natural composites. The modified flax fibre was utilized as a sensor (El Jaouhari *et al.* 2018), and the authors described the spectroscopic response of polypyrrole PPy coated flax fibres to trimethylamine vapour. The outcomes indicated that the flax-PPy sensor effectively senses harmful vapours.

Cotton fibres are the most often utilized NFr in the textile industry to manufacture cloth. Contrary to popular belief, cotton is also employed in the manufacture of medical masks. Thus, antimicrobial resistance is a critical property to consider when considering the use of cotton fibres in medical applications. The dip-coating procedure used natural rubber latex on the surface of cotton fibres to enhance their antibacterial characteristics (Banerjee *et al.* 2019). The results indicated that incorporating silver NPs into cured natural rubber latex coatings considerably increased the elongation of cotton fibres (by 25%). Additionally, the enhanced wettability of the natural rubber latex coating resulted in a higher level of antimicrobial resistance against Gram-positive and Gram-negative bacteria.

Due to the versatility and economics of dip coating, the researchers focused on depositing diverse polymer coatings on the surface of NFr. Recently, a biocomposite of modified clay (Mt-Tbz) coating utilizing dip coating on the surface of date palm fibre (Dpf) was produced and recommended for use in antibacterial food packaging (Hassani *et al.* 2020). Additionally, the Mt-Tbz coating enhanced Young's modulus and tensile strength of date palm fibre. The addition of Mt-Tbz coating enhanced plasticity significantly because of their excellent interactions with chitosan, cellulose, and water molecules, consequently increasing tensile strength.

Nanoparticle Treatment Natural Fibre Polymer Composites

Environmental consciousness has led to substantial research and industry investment in another class of NFPCs with matrices consisting of starch, cellulose, chitin and chitosan, collagen, lignin, natural rubber, polyhydroxyalkanoate, polylactic acid (PLA), and soy-based resins. In comparison to synthetic polymer-based composites, the biodegradable composites benefit from the same polarities of their reinforcements and matrix, which enhances their compatibility and IFB (Satyanarayana *et al.* 2009). However, certain surface treatments of the fibres are required for the purpose of reducing moisture susceptibility, which often results in dimensional instability due to NFr swelling and consequent loss of interfacial integrity.

Nanoparticles are transforming material science and polymer composites in unprecedented ways. This advanced method involves adding a small amount of NPs to a variety of polymers and other materials, and thereby significantly improving their performance and quality in areas like thermal, mechanical, moisture adsorption, and flame retardancy (Mohammed *et al.* 2017a, b). Nanoparticles can be regarded as solid additions that can improve the multiple characteristics of original materials without augmenting their density. Although researchers are increasingly using NFr as a substitute for synthetic fibres in polymer composites due to their low cost, biodegradability, light weight, excellent mechanical characteristics, and sustainability, the compatibility issue between hydrophobic polymer matrix and hydrophilic fibre remains a concern, resulting in the degradation performance of these composites. Various surface treatment procedures such as alkylation, acetylation, and maleated coupling have been used to promote the compatibility of the matrix and NFr. NPs or nanofillers offer enormous potential for usage as filler materials in polymer composites to improve their qualities (Mohammed *et al.* 2018). Academics and researchers have improved the characteristics of NFr reinforced polymer composites by including nano clay, nano-silica SiO_2, carbon nanotubes, and a variety of other NPs (Mohammed *et al.* 2017a, b). Nanoclay has demonstrated its ability to reduce moisture adsorption in sisal fibre composites, which is an extremely desirable property.

Similarly, nano-silica SiO_2 is quite beneficial in improving the mechanical characteristics of these composites. Additionally, carbon nanotubes have been shown to be beneficial in enhancing the mechanical and moisture adsorption capabilities of bamboo and ramie fibre composites (Devnani and Sinha 2019). Nanocomposite has numerous uses in various industries due to its larger surface area and greater aspect ratio, as well as its interesting features. Due to their environmental friendliness, nanocomposites applications provide new technology and business potential for a variety of industries, including aerospace, automotive, electronics, and biotechnology (Mohammed *et al.* 2017a,b). Hybrid bio-based composites that use the synergy between NFr and a nanoparticle reinforced bio-based polymer can result in better characteristics while preserving an appealing environmental profile (Hosseini 2017).

The purpose of modifying NFr is to maximize the improvement process at the interface by regulating intermolecular forces. Customization of the molecular attraction is possible by fibre modification, polymer matrix modification by integrating nanoparticle-reinforcement, or a combination of the two approaches (Zhong *et al.* 2009; Karger-Kocsis *et al.* 2015; Zhang *et al.* 2018). To improve adhesion and decrease matrix dominating qualities, the idea of synergy or synergistic impact has emerged. The fibre surface is treated with an appropriate functional group, and the matrix is also modified simultaneously by adding virgin or functionalized of NPs to accomplish the synergistic effects. NPs, such as Nano-$CaCO_3$, Zinc oxide NPs, and titanium dioxide, have been frequently employed to modify the polymer matrix because of their outstanding mechanical and physical characteristics (Wang *et al*. 2012; Soltani *et al.* 2013; Li *et al.* 2017). However, the full potential of the NPs can only be achieved if it is homogeneously dispersed within the matrix. NPs are prone to agglomeration due to their high surface energy, which has been demonstrated to be detrimental to the mechanical characteristics of composites in particular (Zabihi *et al*. 2016).

Further, higher loading of NPs results in agglomeration, which has a negative effect on the final characteristics of the composites (Zabihi *et al*. 2016; Mohammed *et al.* 2019). Therefore, an optimal quantity and carefully modified surface are desirable for maximizing the unique characteristics of NPs. The NPs may operate as a bridge between the NFr and the polymer matrix, enhancing interfacial adhesion (Mohammed *et al.* 2019). The interfacial characteristics can be improved by modifying the fibre, matrix, and NPs in hybrid composites (Pracella *et al.* 2010). NPs is considered to increase the surface roughness of the fibre, hence increasing interfacial adhesion by mechanical interlocking (Mohammed *et al.* 2018).

Various nanoparticles utilized in natural fibre reinforced polymer composites

Since the last several years, nearly everything has become "nano," including materials that have been around for over a century, such as carbon black, which has been widely utilized as reinforcement or filler in rubbers. Nanofillers are found in both organic and inorganic environments. Inorganic NPs materials include silica (SiO_2), titanium dioxide (TiO_2), calcium carbonate ($CaCO_3$), and polyhedral oligomeric silsesquioxane (POSS). However, fillers, such as coir nanofiller, carbon black and cellulosic nanofiller, and many others, are derived organically and naturally represent organic NPs. According to Mohammed *et al.* (2019), the general principle of nanocomposites is to create a comprehensive interface between the nanoscale building blocks and the polymer matrix. Frequently, the homogenous distribution of NPs presents a dilemma.

NPs are defined as solid materials that can be added to polymer matrixes and lignocellulosic fibres. They are structurally and chemically distinct from polymer matrices and lignocellulosic fibres. They can be one-dimensional, such as plates and laminas, two-dimensional, such as nanotubes and nanofibres, or three-dimensional, such as isodimensional NPs. They are mainly composed of inorganic compounds, but occasionally organic ones as well. Various NPs such as nano clays, primarily montmorillonite, (Ashori and Nourbakhsh 2011; Mohan and Kanny 2011), nano SiO_2 (Hosseini *et al.* 2014), nano TiO_2 (Vilakati *et al.* 2010), carbon nanotubes (Kushwaha *et al.* 2014), and nanographene are employed in NFr reinforced polymer composites (Chaharmahali *et al.* 2014; Javanshour *et al*. 2021). Besides these conventional fillers, additional nanofillers such as oil palm nanofiller have been evaluated (Saba *et al.* 2016). Using a trace amount of these fillers significantly improves the mechanical, thermal, and moisture adsorption qualities. Nanomaterials are categorized into three types according to their shape, as illustrated in Fig. 11.

Among the inorganic NPs utilized for fabric functionalization, ZnO is particularly appealing because of its unique mix of desirable qualities such as low cost, high abundance, environmental friendliness, and excellent mechanical, thermal, and chemical stability. Additionally, ZnO nanostructures with customized morphologies (rods, prisms, flowers, wires, and tubes, for example) can be easily synthesized *via* wet and dry methods (Preda *et al.* 2013; Tchoe *et al.* 2020) and engineered to yield advanced functional materials for use in superhydrophobic surfaces (Preda *et al.* 2013; Florica *et al.* 2016), photodetectors (Costas *et al.* 2019; Tchoe *et al.* 2020), and other applications.

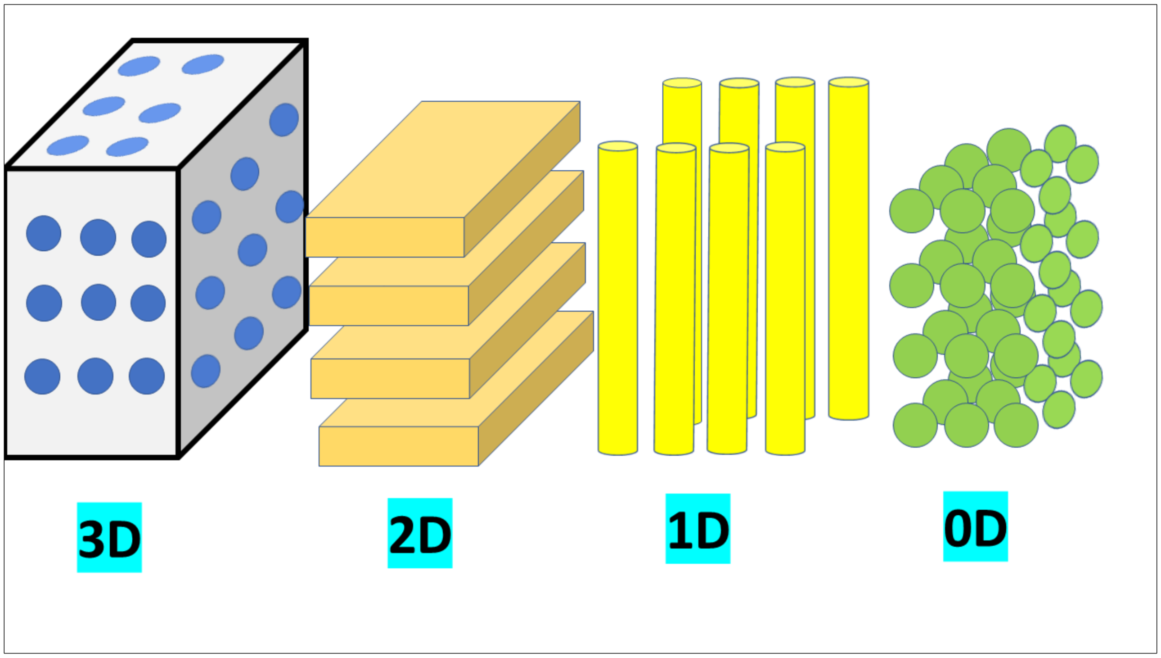

Fig. 11. Various types of nanoscale materials (Figure redrawn and credit to Kumar *et al.* 2009)

In recent years, the development of ZnO nanostructured interphases for enhancing IFB between fibre and matrix in polymer composites has garnered substantial interest, as the mechanical properties of the composites are ultimately dependent on the performance of the interphase. Thus, for a particular fibre and resin system, the production of composite materials with optimum properties for specific applications requires the design of appropriate interphase capable of controlling the interfacial shear strength. As a result, ZnO electroless deposition can be considered an excellent method for increasing the mechanical performance of NFr composites, hence broadening their potential application as structural materials.

In the period of development of nanotechnology solutions for diverse applications, the exploitation of nanomaterial to improve interfacial adhesion in carbon fibre composites could potentially lead to enhanced mechanical interlocking as well as chemical interaction (Naebe *et al.* 2010; Bafekrpour *et al.* 2012). Nanomaterials can be introduced onto the fibre surface in two ways: by dip coating the fibre in water-based solutions containing NPs or by directly grafting them onto the fibre surface (*e.g.*, chemical vapor deposition, chemical vapor infiltration, and injection chemical vapor deposition). Nonetheless, there have been limitations such as optimization and reduction of fibre strength. Additionally, these processes are easily scaled up to large production volumes, although the cost of carbonous nanomaterials (*e.g.*, graphene, carbon nanotube, and carbon nanofibre) is relatively high (Sharma *et al.* 2014; Yao *et al.* 2018).

A significant justification for employing NPs is their inherent large surface area-to-volume ratio, which increases the degree of interactions between NPs and matrix, thereby enhancing the overall impact on the material behavior. A nanoparticles system with a homogeneous distribution lead to better nanocomposite behavior. Agglomeration of NPs degrades material performance by forming voids initiation, which serve as sites of failure and crack propagation (Yu *et al.* 2000). As a result, the quality of NPs distribution in polymer matrix is critical. NPs within the nanometer range of less than 100 nm are prone to agglomeration or clustering due to the dominating intermolecular van der Waals interactions that exist between NPs (Qian *et al.* 2000). The apparent differences in the characteristics of polymer nanoparticles composites can be deduced from their extremely broad interfacial nanoparticles area. The interfacial region of NPs is orders of magnitude larger than that of traditional macro or micron dimensioned particles (Bai *et al.* 2018). Consequently, composites of polymer nanoparticles exhibit superior gas barrier, solvent resistance, and flame retardant characteristics compared to pure matrices (Hegde *et al.* 2020).

Increased incorporation of NPs above a certain threshold level can result in the formation of a network that further restricts the mobility of the polymeric chains. Furthermore, enhancement in the characteristics of polymer nanoparticle composites is dependent on the interfacial region, the proximity of the NPs, and the interfacial interactions (Yang *et al.* 2011). As a result, in addition to the behavior of the composites ingredients, additional aspects of the composites, such as the interfacial area, play an important role in the final performance of the composite. Enhancing the interface between NFr and matrix is also critical for green biocomposites, particularly NFr polymer biocomposites and nano-biocomposites because the bonding between hydrophilic NFr and some hydrophobic polymers is relatively weak (Mazurkiewicz-Pawlicka *et al.* 2020). Furthermore, robust bonding at the polymer/NFr and polymer/NFr/NPs interfaces is critical for efficient stress convey from damaged to undamaged fibres, hence improving the mechanical properties of the NFPCs. Additionally, poor bonding enables the formation of energy-absorbing sites across interfacial faults, enhancing material toughness. Therefore, efficiently controlling the polymer/NFr and polymer/NFr/NPs interfaces makes it possible to derive composites with variable properties from the same polymer/NFr and polymer/NFr/NPs interfaces. Table 3 displays the effect of nano fillers on the properties of NFr reinforced composites.

Interaction Nanoparticles with Natural Fibers and Polymer Composites

Nanoparticles could penetrate the cell wall before hydrolysis and poly condensation. However, nanoparticles become mostly deposited in cell lumens and partially in cell walls. Due to the presence of nanoparticles in both the cell walls and lumens, the physical and mechanical properties of the ensuing natural fibres would be affected to varying degrees (Wang *et al*. 2012). Inorganic nanoparticles were integrated into the cell wall and were likely occupying the vacant space (micropores) that would otherwise be accessible to water molecules, hence decreasing the hygroscopicity of the modified natural fibres (Donath *et al*. 2004; Hill. 2007). On the other hand, the hydroxyl groups of cell wall components mainly responsible for moisture absorption possibly were blocked by formation of hydrogen bonds with inorganic nanoparticles during hydrolysis and polycondensation, which may have contributed to reduced hygroscopicity and increased IFB.

Amalgamation of the inorganic nanoparticles into cell walls reduce the ability of the cell wall to swell owing to bulking, hence enhancing the dimensional stability of impregnated natural fibres. In reality, only the inorganic nanoparticles integrated into the cell wall are anticipated to affect hygroscopicity, dimensional stability of natural fibres, and IFB, whereas those deposited in the cell lumen are anticipated to play a small impact (Tshabalala *et al*. 2011). The inorganic nanoparticles loaded into cell wall and on the surface may provide strong static electric attractive forces to nonpolar polymer surface, and hence the compatibility between the fibers and the polymer matrix caused enhanced mechanical characteristics and decreased water absorption. In the other words, the impregnation of the inorganic nanoparticles into the micropore structure of the fiber cell walls can reduce the micro void volumes in the fiber, as well as reduce air bubble formation during the composite fabrication process (Mai and Militz 2004).

The behavior of composite is determined by the nature of the interface between the NPs and the matrix (Douce *et al.* 2004). However, interactions between NPs and matrix are classified as covalent or non-covalent based on hydrogen bonding (Fu *et al.* 2008), van der Waals (VDW) (Liao and Li 2001), and electrostatic forces (Choi and Ryu 2010). As a result of the interactions of the composite constituents, numerous enhancements in the mechanical behaviour of the composites occurred.

The chemical structure of the polymer and the surface charge of the NPs govern the interfacial interaction between polymer reactive groups and NPs (Lordi and Yao 2000). These interaction activities are characterized according to their ability to form covalent bonds: carboxyl and sulfonation groups, amide or amine groups capable of forming covalent bonds with other groups on the NPs, such as hydroxyl groups (Liu *et al.* 2017). Ionic-bonds: These are chemical bonds formed by the action of coulombic forces between positively induced and negatively disposed charges. In general, the accessible polymeric chain network and NPs have opposing charges, which helps the formation of nanocomposites. Chiral-bonds: Chiral activity is used to prepare nanocomposites using in-situ polymerization. Here, organic matrices express a pair of electrons that engage in chiral activities with open electron orbitals, resulting in a chemical reaction. Affinity effect: In this case, the attraction of NPs to surface groups on polymer chains facilitates covalent bonding. This characteristic enables the NPs to be distributed uniformly within the matrix. Nevertheless, interactions between NPs and matrix are categorised as follows:

Non-covalent interaction

The non-covalent interaction between NPs and polymer matrix can be improved by incorporating bridges, polymeric wrapping, interfacial zone, and enlargement (Mu and Winey 2007). Bridging happens when a polymer chain simultaneously interacts with two or more reinforcing NPs. On the other hand, as NPs content is increased and a greater molecular weight polymer is used, the opportunity for bridging occurrence will be increased (Gao *et al.* 2011). The particular interactivity zone between NPs and polymer matrix is a significant factor that affects composite behaviour. It is defined as the polymer-NPs-per-unit-volume interfacial region. It is related to the density ratio of polymer matrix in relation to their concentration, NPs content, and NPs diameter (Nish *et al.* 2007). Thus, studies have indicated that polymer matrix is linearly correlated to the total interfacial zone of the NPs, meaning that the low NPs incorporation is more likely to exert a bigger impact on the behaviour of the end product. Wrapping the NPs with polymeric chains and increasing the interaction is essential for enhanced NPs dispersion in the matrix. Additionally, interfacial crystallisation of a semicrystalline polymeric host can be used to improve interfacial interactions between NPs and polymer matrix. Here, NPs will act as nuclei, while the semicrystalline substance will crystallise at the interface region between NPs and polymer matrix (Hersam 2010).

Covalent interactions

Covalent interaction activity takes place when polymer chains are chemically bonded to the reinforced NPs (Hirsch 2002). For this purpose, it is critical to apply appropriate chemical modifications to the NPs surface in order to attach functional groups able to interact with the matrix. Another possibility is to modify the polymer matrix before chemically binding it to the NPS. Although covalently bonding the NPs and the matrix is more effective at increasing interfacial strength due to increased adhesion, the pre-treatment technique requires careful attention.

Influence of Interface Between Nanoparticles and Natural Fiber Polymer Composites on Mechanical Behaviour

Nano-reinforcements have been recognised as the superior option for enhancing the mechanical properties of NFPCs. Due to the fact that nanoscale fills are typically defect-free, the applications in the field of polymeric composite region setting have the potential to circumvent the limitations of the conventional micrometre scale. Consistent and ubiquitous inclusion of nanoparticles creates a wide interfacial contact surface, which affects relaxation behaviour as well as physical, molecular mobility, and thermal performance (Pasupathi *et al*. 2020). Jiang *et al*. (2013) studied the effect of nano-silica on the properties of polymer matrix composites containing urea formaldehyde. In addition, the study found that nano-silica improved the adhesion of composites, while nano-impregnated composites increased the adhesion and bending young's characteristics of wood by around 15% and 21 %, respectively. In addition, Kushwaha *et al*. (2014) anticipated the effect of carbon nanotubes (CNT) on epoxy-based polymer composites with bamboo in various fractions and evaluated the physical properties of such composite material. The addition of CNT to an organic fibre epoxy composite material enhanced its mechanical characteristics. Furthermore, the study indicated that the inclusion of carbon nanotubes affected the water-retention capacity of composite materials. The effect of titanium dioxide nanoparticles on the thermal and physical characteristics of a bagasse-based composite was studied by (Vilakati *et al*. 2010). While the matrix stress measured 12 MPa, the addition of 2 % nano-titania filler increased the strength by 11 %.

Sharma *et al.* (2022) examined the effect of nanoclay reinforcement in luffa/epoxy (LE) polymer composite material at various loading fractions (0%, 1.0%, 2.0%, and 3.0%). In terms of mechanical property enhancement, the addition of nanoclay up to 2.0 % in LE composites were found to be increasingly advantageous. However, the addition of nanoclay after 2.0% had ended-up in adverse impacts due to the agglomeration of the nanoparticles. The highest percentage improvement in tensile, compression and flexural properties was observed as 20.0%, 18.69%, and 21.44%, respectively with the addition of 2.0% of nanoclay in LE composites. The percentage of enhancement in tensile, compression and flexural properties was suppressed by 6.44% 8.5%, and 8.32%, respectively with the addition of 3.0% nanoclay comparing to the previous fraction.

Polymer matrix interfacial interactions with NPs have a crucial influence on the mechanical behavior and performance of manufactured polymer nano composites. According to research, nanofillers can be used to modify the characteristics of polymer matrix (Ren *et al.* 2018). Inorganic microparticles such as glass, talc, and calcium carbonate have been widely used to improve the mechanical properties of traditional composites (Karthikeyan *et al.* 2017; Yan *et al.* 2019). These qualities can be further enhanced by raising their aspect ratio and shrinking the dimensions of the NPs to the nanoscale scale. However, Hubbe and Grigsby (2020) emphasized in their review that depending on many findings of different studies, there does not appear to be any consistent and dependable advantage of using nano-sized cellulosic reinforcements when attempting to achieve high values of composite strength or modulus. The characteristics of NPs and polymer matrix, NPs dispersion, interfacial adhesion, and processing procedures are parameters to consider (Nuriel *et al.* 2005) since the behavior of polymer nanocomposites is strongly dependent on their microarchitecture. The mechanical properties of polymer nano composites are highly dependent on the NPs dimension, applied load, and NPs/polymer matrix interfacial bonding (Tran *et al.* 2008). The interfacial bonding is crucial for evaluating mechanical load transfer from polymer matrix to NPs (Vijayaraghavan and Zhang 2019). Notably, the interfacial adhesion is the primary determinant of the toughness and strength of the composites. Consequently, the distribution of NPs within the matrix, interfacial adhesion, geometrical dimensions, etc., play crucial roles in enhancing the mechanical properties of the composites (Vijayaraghavan *et al.* 2015). In addition, mechanical characteristics are evaluated using standard methods such as tensile, and compression analysis (Zolali and Favis 2017).

The tensile strength of nanocomposites is determined by the effectiveness of stress transfer between the elements of the nanocomposites and the interfacial adhesion of the composite elements. Strength is increased when the load is efficiently transferred to the NPs (Lau and Hui 2002; Zhong *et al.* 2018). At specified NPs volume fractions, the smaller NPs have a larger interfacial area, resulting in a large area of stress transfer regions (Lau *et al.* 2004). Strength also influences the efficiency of load transfer (Paiva *et al.* 2004). In contrast to composites with strong interfacial interaction (Thostenson and Chou 2002), composites with poorly adherent NPs would have insufficient strength due to non-continuity produced by interfacial de-bonding, which inhibits the NPs from effectively transferring the applied load. Multiple studies on surface treatment of NPs for composites materials have been conducted (Reddy *et al.* 2018; Ahmad *et al.* 2019), resulting in enhanced dispersion and interfacial interaction, as a result, increased tensile strength of the composites. These findings suggest that chemical bonding at nanofiller- polymer matrix interfaces can significantly increase the strength of composites.

Even though NPs improve the mechanical properties of polymer composites the NPs tend to agglomerate throughout the manufacturing process (Singh *et al.* 2002; Jux *et al.* 2017). Agglomeration produces NPs with poor dispersal in the polymer matrix and deteriorated material characteristics. The measured mechanical characteristics rely on the size distribution of the agglomeration and the NPs weight fraction (Uddin and Sun 2010; Fankhanel 2020). West and Malhotra (2006) manipulated the dispersion quality of NPs by subjecting the probe to a variety of ultrasonic treatments and observed that the dispersion quality had a substantial effect on the mechanical properties. Zare and Rhee (2017) demonstrated that reducing the size of NPs while increasing the diameter of NPs aggregates results in low reinforcing efficiency. Ma *et al.* (2017) used the Paul and Maxwell models to find the agglomeration moduli. They discovered that the agglomeration of NPs reduces Young's modulus substantially compared to the fine dispersion of NPs.

However, with more significant loadings, such as 5% and 10% of NPs, the interfacial bonding strength increased by 24 and 17.19%, respectively. The lesser rise in interfacial bonding with increasing NPs content may be attributable to the inhibiting impact of NPs on the surface for further penetration of the polymer matrix into the NFr surface, resulting in incomplete wetting. To elaborate more on this aspect, it can be deduced that the interfacial adhesion relies on the thickness of the interface region in which the NPs exist. Higher NPs concentrations can result in the formation of congested nanomaterial on the NFr surface, limiting the percolation of polymer matrix and its penetration into the NFr surface; hence, some portions of the fibre surface remain intact. Consequently, as the NFr is exposed to the load, it is hypothesized that the thick interphase is capable of sliding easily (Karger-Kocsis *et al.* 2015; Zabihi *et al.* 2017). This putative mechanism is depicted schematically in Fig. 12.

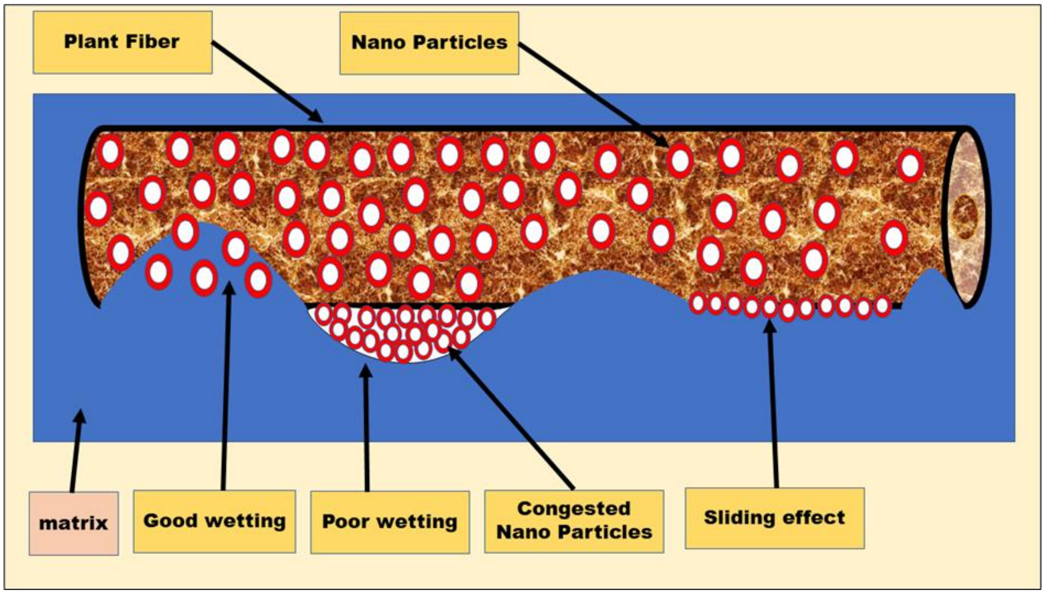

Fig. 12. Schematic representation effective mechanisms in interfacial performance of modified natural fibre with high level of nanoparticles loading

Table 3. Summary of Effect of Nano Fillers on the Properties of NFr Reinforced Composites

Polymer/ Natural Fibre-Nanocomposites	Achievement	Reference
Epoxy/Jute/Nano-titania (nano-TiO_2)	The addition of 1% nano-titania nanoparticles to an epoxy/jute composite decreased water diffusion by 18.18% and increased impact strength by 39.36%.	Gift *et al.* (2022)
Epoxy/kenaf fibre/nano-SiO_2	The tensile strength, compressive strength, and impact strength increased by 20.61%, 23.71%, and 22.88%, respectively, with 2% fraction of nanofiller in composite.	Jotiram *et al.* (2022)
Epoxy/jute fibre/carbon nano tube	With a 6% fraction of nanofiller in composite, the tensile strength, flexural strength, and impact strength improved, correspondingly.	Saiteja *et al.* (2020)
Vinyl ester resin / basalt fibre / Silicon carbide nano (SiC)	With a 4% nanofiller percentage in composite, the mechanical properties improved enhanced fibre mat-matrix interfacial interaction. Moreover, the composites have superior wear resistance and had a low capacity for water absorption with 2% wt.	Thooyavan *et al.* (2022)
High-density polyethylene (HDPE) / wheat straw fibre (WSF)/ nano-silicon carbide (nano-SiC)	With nano-SiC content of 0.4 wt%, the tensile strength, flexural strength, and flexural modulus increased by 15.5%, 11.5%, and 12.4%, respectively. In addition, the water absorption was enhanced, although the thermal stability dropped little. Dimensional stability and interfacial characteristics have been enhanced	Zhang *et al.* (2022)
Epoxy/ flax fibre/ nano TiO_2	There was an improvement in the tensile, flexural, and interlaminar shear strength values of 22%, 24% and 16%, correspondingly. Water diffusion coefficient of fibre modified composites were lowered by 42% for 0.6 wt% TiO_2 coated samples.	Prasad *et al.* (2021)
High-density polyethylene (HDPE)/wood fibre/ nano-montmorillonite (MMT)	10% nano-OMMT achieves the best values for impact, tensile, and flexural strength (38.1 MPa, 21.4 KJ.m^2, and 60.8 MPa, respectively). With increasing nano OMMT, the thermal degradation temperature and flame retardant performance of the composite increased. The moisture absorption composite decreased as nano-OMMT content increased.	Fan *et al.* (2021)
High-density polyethylene (HDPE)/straw fibre/nano-xylan	Through nano-xylan treatment, the antifungal performance of straw fibres can be enhanced. Nano-xylan with smaller particle size displays superior performance in minimising mass and strength losses.	Gao *et al.* (2021)
Polyester/vakka fibre/ nano clay	The tensile strength and flexural strength of nano clay-filled vakka fibre polyester composites are 21.2% and 55% more than those of composites without the addition of nano clay.	Gudapati *et al.* (2019)
Epoxy/pineapple fibre/ nano-calcium carbonate (N-$CaCO_3$)	The addition of N-$CaCO_3$ up to 3 wt% improved the tensile, flexural, fracture toughness, and thermal stability of PF/Ep composites, whereas adding 1 wt% of nano-calcium carbonate (N-$CaCO_3$) increased the impact strength.	Mahadevaswamy and Suresha (2020)
Epoxy/Flax fibre/Titanium dioxide (TiO_2) nanoparticle	When the TiO_2 concentration in composites was increased from 4% to 6%, the tensile strength value increased to 12.31%, and the Young's modulus of the composite increased to 23.17%.	Prasad *et al.* (2018)

CONCLUSION AND FUTURE PROSPECTS

1. Natural fiber polymer composites (NFPCs), in which a variety of fillers and fibres are utilised as agents of reinforcement, have piqued the interest of experts in various engineering and technology application. Fibres made of natural materials are gaining popularity as an alternative to materials made of synthetics in reinforcing composites due to their bio-renewable properties and environmentally friendly behaviour, and can therefore be used effectively in a variety of applications despite the fact that artificial fibres are commonly used to manufacture composites.
2. This paper presents a comprehensive review of the interfacial properties of NFPCs. The interface mechanism and the interfacial properties of NFPCs are studied. The interfacial characteristics are also introduced. The mechanism of interphase in composite materials includes physical attraction between electrically neutral bodies, molecular entanglement, interdiffusion, chemical bonding, reaction bonding, and mechanical bonding. The final bonding is most likely formed by the combination of some of these mechanisms.
3. The natural fiber (NFr)-matrix interface and the relationship between interface and bulk composites were detailed in depth to provide a better understanding, which lead to the development of a unique approach for better bonding between NFr and matrix, and compatibility with a wider range of materials/components. Understanding the relationship between the processing methods and mechanical properties of the composites and the recycling characteristics/options of the NFRCs will result in the development of more beneficial technologies and applications for NFr and its composites.
4. This review summarizes recent scientific research on the surface treatment of NFr. Physical, chemical, and nanoparticle treatment approaches have been discussed, with a focus on how each treatment affects the surface characteristics of the fibre and the IFB characteristics with the polymer matrix. Most of the treatment approaches presented were shown to be efficient in enhancing the interfacial adhesion of NFr with polymer matrix but at the price of the intrinsic strength of the fibre.
5. Although plasma and corona treatment are versatile fibre treatment technologies, their high operating costs and technical problems have limited their usefulness for large-scale utilization. Combining two or more modification methods reveals intriguing multi-functional properties. However, given the various methodologies and high cost, this strategy does not appear scaleable. The development and selection of efficient and cost-effective surface modifiers for both fibre and matrix are the first critical area to be considered for future development. The tendency is toward treatment that not only gives excellent customization but also has a low economic impact.
6. Polymer matrix and NFr have been infused with nano-reinforcements such as carbon nanotubes, nano-$CaCO_3$, and titanium dioxide to increase interfacial adhesion and acquire multifunctional characteristics. The purpose of inserting these NPs was to achieve synergy in the strengthening mechanism by forming physical and chemical anchor points that were excellent for interfacial bonding (IFB). According to the current data, the incorporation of nano-reinforcements with NFr would undoubtedly open the way for the production of high-performance composites with multifunctional properties that are suited for advanced applications.

7. The interface and adhesion of biobased nanocomposites are tremendously advantageous to their development and would help bridge the gap between scientific difficulties and practical production due to the nanoscale interfacial of the composites determining stress transmission, interfacial penetration, interface, and adhesion at the nanoscale.
8. In the examination of the fibre-matrix interface, the approach to improve the IFB based on the interface mechanisms continues to be a crucial factor. Utilizing nanoparticles can improve IFB through a variety of interface mechanisms, such as enhancing mechanical interlocking, increasing the wettability of fibre surfaces, and chemical bonding. Researchers are currently focusing on a "growth" technique that makes nanoparticle grafting more efficient than soaking fibres in a nanoparticle-containing solution. This “growth” technique currently is employed in the fiber polymer composites containing one type of fibre. It will take more research to adapt the technology to the intra-layer hybrid fiber polymer composites which have different fibres in one fiber polymer composites.
9. To achieve the practical application of NFPCs-nanoparticles in composite materials, it is necessary to investigate both experimentally and theoretically the influence of nanoparticles on the interfacial bonding under various conditions (such as weathering, ultraviolet, and hygrothermal environments) for long-term exposure. Additional research can concentrate on assessing the chemical or physical changes of the interface with nanoparticles throughout exposure and developing life forecasts that account for the deterioration of the fibre matrix interface with nanoparticles.

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