Review

Recycling of natural fiber composites: Challenges and opportunities

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ABSTRACT

Natural fibers have been widely used for reinforcing polymers attributed to their sustainable nature, excellent stiffness to weight ratio, biodegradability, and low cost compared with synthetic fibers like carbon or glass fibers. Thermoplastic composites offer an advantage of recyclability after their service life, but challenges and opportunities remain in the recycling of natural fiber reinforced polymer composites (NFRPCs). This article summarized the effects of reprocessing/recycling on the material properties of NFRPCs. The material properties considered include mechanical performance, thermal properties, hygroscopic behavior, viscoelasticity, degradation, and durability. NFRPCs can generally be recycled approximately 4–6 times until their thermomechanical properties change. After recycling 7 times, the tensile strength of NFRPCs can decrease by 17%, and the tensile modulus can decrease by 28%. The mitigation approaches to overcome degradation of material properties of NFRPCs such as adding functional additives and virgin plastics are also discussed. The main challenges in these approaches such as degradation and incompatibility are discussed, and an effort is made to provide a rationale for reprocessing/recyclability assessment. Future applications of NFRPCs such as additive manufacturing and automotive part use are discussed.

Abbreviations and nomenclature

CTE	Coefficient of thermal expansion
HDPE	High density polyethylene
HDT	Heat deflection temperature
LDPE	Low density polyethylene
MAPE	Maleated polyethylene
MAPP	Maleic anhydride-grafted polypropylene
MDFB	Medium density fiber board
NFRPC	Natural fiber reinforced polymer composite
PBS	Poly (butylene succinate)
PE	Polyethylene

PHBV	Poly (3-hydroxybutyrate-co-2-hydroxyvalerate)
PLA	Polylactic acid
PLLA	Poly (L-Lactic acid)
PP	Polypropylene
PVC	Polyvinyl chloride
T_c	Crystallization temperature
T_d	Degradation temperature
T_g	Glass transition temperature
T_m	Melting temperature
WPC	Wood-fiber thermoplastic composite

1. Introduction

Natural fiber reinforced polymer composites (NFRPCs) are a subset of fiber reinforced polymer composites. This subset includes various polymer matrices, both thermosetting and thermoplastic, reinforced by a variety of natural fiber sources arising from wood, plant stalks (bast and core fibers), grasses, seed hairs, and so on (Bongarde and Shinde, 2014). NFRPCs are typically processed using common polymer processing techniques, such as compression and injection molding, as well as extrusion and pultrusion. Common benefits of utilizing natural fibers for reinforcing polymers include their "green" and sustainable nature, biodegradability, excellent stiffness to weight ratio, low abrasion, and low cost compared with synthetic fibers such as glass or carbon fibers (Liu et al., 2020; Zhao et al., 2020b).

NFRPCs have been used for at least 100 years, with most commercial activity occurring during the past 30 to 40 years, especially in thermoplastic composite technology (Gardner et al., 2015). Common applications of NFRPCs include automotive parts, such as interior panels, and building materials, such as architectural moldings, decking, and railing components (Holbery and Houston, 2006; Klyosov, 2007). Recently, NFRPCs have been applied in additive manufacturing (Zhao et al., 2019). The most common thermoplastics utilized for NFRPCs are polyolefins (e.g., polypropylene [PP] and polyethylene [PE]), polyvinyl chloride (PVC), polyamides (e.g., nylon), and, recently, biobased polylactic acid (PLA). Thermoplastic polymer matrices are promising in NFRPCs because of their low cost, wide availability, and low processing temperatures (typically $< 225\ °C$), which are suitable for processing natural fibers without experiencing thermal degradation (Wang et al., 2020; Yao et al., 2008; Zhao et al., 2020b).

Natural fiber feedstocks for fiber reinforced composites can often be obtained from residues from primary processing of wood or agricultural materials. These feedstocks are obtained as both powders (e.g., wood flours) and chopped bast fibers. Wood flour is typically manufactured from sawdust, planer shavings, and molding residues and obtained from secondary processing of wood lumber. However, bast fibers are obtained from the processing of plant stalks such as jute, kenaf, and hemp. Once collected, natural fibers are appropriately comminuted to a particular size, dried to remove moisture, and then compounded with plastics and/or other additives (e.g., processing aids, colorants, UV protectants) that will comprise the final composite. Feedstock suppliers and composite producers utilize virgin polymers and recycled polymers (post-consumer or post-industrial), depending on the availability and end use application of the material.

NFRPCs are often regarded as green, sustainable, and recyclable materials. Mechanical and thermal (e.g., gasification) (Parparita et al., 2015) recycling methods have been used to recycle NFRPCs. Unfortunately, commercial recycling and reuse activity for these materials is limited in North America, although recycling activity is becoming more common in the European Union, per legislative policies. Recycling is becoming more important for materials relative to the circular economy (Moazzem et al., 2021) in which future manufacturing processes will be designed to minimize and eventually remove waste out of the system (Zhao et al., 2022). Fig. 1 shows a flow diagram of recycling of natural fiber composites. In the future, composite materials will be expected to be reused and/or repurposed rather than relegated to landfills. There are challenges as well as opportunities in the recycling of NFRPCs. For example, obtaining NFRPCs at the end of life is currently neither feasible nor cost-effective. In the automobile industry, cars at the end of their useful lifetime are often sent through a shredder. During this process, polymeric materials have to be separated from metal and glass. This is certainly possible, but the resulting polymeric mixtures often cannot be recycled without additional separation steps. In the building materials area, there are currently no commercial recycling programs for decking, railings, and architectural moldings at the end of life, so these materials end up in the landfill or are combusted for energy.

When manufacturers utilizing NFRPCs in the production of materials are in control of the feedstock supply chain, there are some excellent examples of recycling and reprocessing of materials for NFRPCs commercially. For example, when Trex began manufacturing decking in the 1990s, their primary feedstocks were recycled low density PE (LDPE) plastic grocery bags and wood flour (Trex, 2020). Anderson Windows took advantage of their residue streams for wood and vinyl window production, and they created PVC-wood composites (Fibrex®) for new window applications (Andersen, 2020). Producers of wood plastic composite decking reprocess their off-spec material in home construction to use as feedstocks for new decking, usually in the core of co-extruded profiles. These are just a few examples of recycling and reprocessing practices currently applied in the NFRPC industry.

The terms "recycling" and "reprocessing" are often used interchangeably to refer to reuse of processed material. The terms "external recycling" and "internal recycling" are also sometimes used to indicate if the material is being used after its service life (external) or reprocessed
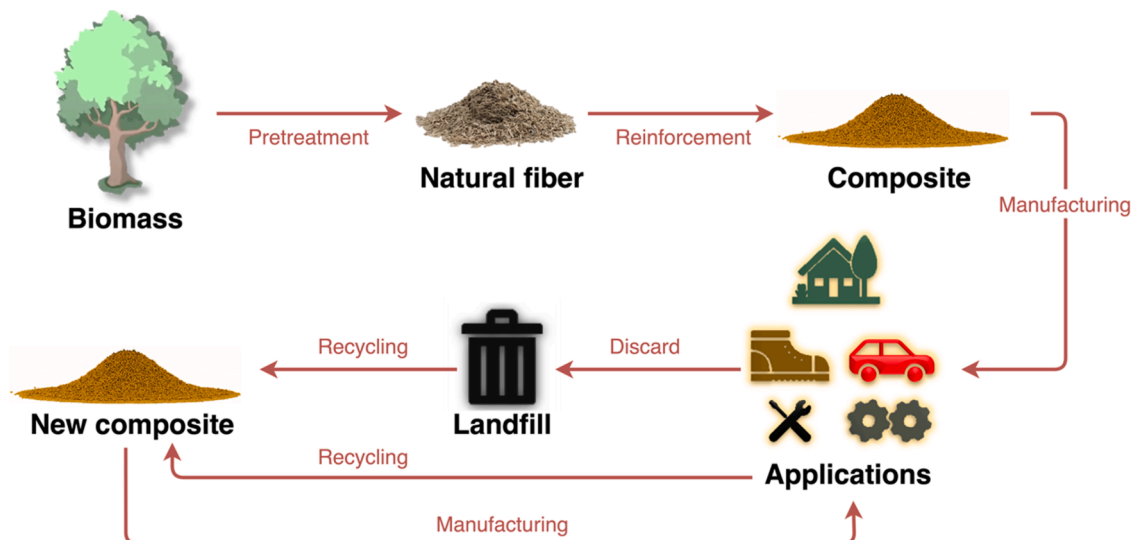

Fig. 1. A flow diagram of recycling natural fiber composites toward a circular economy in which future manufacturing processes will be designed to minimize and remove waste from the system. Natural fibers derived from biomass are used as reinforcement to produce composites, which will be expected to be reused and/or repurposed after completing their service life.

internally in the same production line as is done for waste of defective products. Finally, the terms "chemical recycling" and "mechanical recycling" are used to differentiate processes in which the main components of the composite material are broken down to their chemical constituents or are retained in their original form but reprocessed into a similar product, respectively. Recycling provides different benefits attributable to the good performance of the composites after certain processing cycles and the cost savings of an additional production phase (Bourmaud et al., 2016). Depending on the final application for recycled material, composites may have different recycling potential (Faraca and Astrup, 2019). A primary recycling method for post-consumer plastic waste is mechanical recycling, in which the plastic waste is ground, washed, and reprocessed to form new secondary plastic materials that maintain their original chemical structure. However, for composite recycling, the composite type and application play a critical role in the recycling efficiency (Faraca and Astrup, 2019). Some composites such as wood/PP and flax/PP maintain good stability regarding mechanical properties after recycling (Bourmaud et al., 2016).

As manufacturers transition toward circular economy practices (Huysman et al., 2017), it will be important to have a complete understanding of the impact of recycling and reprocessing on material properties of NFRPCs as they are reused in subsequent product applications. Research on recycling and reprocessing of NFRPCs has been reported over the past several decades and is the focus of this review. The scope of the paper focuses on thermoplastic matrices, and the effects of reprocessing/recycling on the material properties (e.g., mechanical, thermal, hygroscopic, viscoelasticity) of NFRPCs. The mechanisms behind these property changes are discussed. Mitigation approaches to overcome degradation of material properties during recycling are discussed in detail, and the main challenges in these approaches are discussed. An effort is made to provide a rationale for reprocessing/recyclability assessment from practical considerations to obtain materials, as well as from a use perspective for subsequent product applications and outlook.

2. Effects of recycling/reprocessing on material properties

2.1. Mechanical properties

The mechanical performance of polymer composites is one of the most important factors examined in determining their recyclability. The effect of reprocessing on the properties of neat polymers depends on many factors, including the method of processing (e.g., thermo-mechanical, chemical, aging), processing parameters (e.g., temperature, time, speed and shear rate, humidity, UV exposure), and, of course, the molecular makeup of the polymer itself. Fig. 2 shows the chemical structures of some representative polymer resins, such as poly(3-hydroxybutyrate-co-2-hydroxyvalerate) (PHBV) and poly(butylene

succinate) (PBS). When reinforcing agents, such as particles, fibers, or other fillers, are added to the polymer, the relationships between recycling processes and resulting material performance become even more complex. Fig. 3 shows the digital photos of representative natural fibers.

A summary of the mechanical properties of a variety of NFRPCs is given in Table 1. The following sections discuss the effects of recycling on the tensile, flexural, and impact performance of a wide variety of polymer composites investigated in recent years. Researchers commonly simulate recycling by repetitive thermo-mechanical reprocessing using combinations of extrusion, melt compounding, compression molding, injection molding, and grinding or shredding (Bakkal et al., 2012; Soroudi and Jakubowicz, 2013). Some researchers have also investigated solvent-assisted recycling in which a polymer and fiber filler are dissolved in a suitable solvent, cast to films, and re-extruded, or processed in some other method. Finally, aging and the effects of long-term use on the recyclability of composites are occasionally considered by researchers. Aging can be simulated by exposing the material to prolonged elevated temperature, humidity, UV exposure, or biological agents such as bacteria or fungi (Bakkal et al., 2012; Soroudi and Jakubowicz, 2013).

2.1.1. Tensile properties

The tensile properties of a composite, including tensile strength, modulus, failure stress, and failure strain before and after recycling, are highly dependent on the aforementioned competing factors, including fiber aspect ratio and dispersion, compatibility between the fibers and polymer matrix, and the molecular weight of the polymer. Therefore, these properties have been shown to both improve and worsen with increased reprocessing cycles. For example, Bodur et al. (Bakkal et al., 2012; Bodur et al., 2011) recycled LDPE samples reinforced with waste cotton textile pieces. The tensile strength and modulus of the composites first increased and then decreased with subsequent recycling steps. Meanwhile, the tensile strength of neat LDPE decreased with recycling, which indicated polymer degradation and chain scission. The initial increase in the tensile performance of the composites could be attributed to an increased aspect ratio of the cotton textiles and a better-quality dispersion upon recycling. The fiber orientation also affected the tensile performance of the composites: the samples parallel to the extrusion direction had a 51% higher tensile strength than the samples perpendicular to the extrusion direction. The reduction in tensile performance attributable to polymer degradation after six recycling steps eventually outweighed the positive contributions given by the fiber reinforcements upon reprocessing. Similarly, Beg and Pickering (Beg and Pickering, 2008b) and Uitterhaegen et al. (Uitterhaegen et al., 2018) observed a similar increase (after recycling two times) and subsequent decrease during repeated reprocessing of wood fiber/PP (Beg and Pickering, 2008b) and LDPE composites filled with coriander straw fiber and wood fiber (Uitterhaegen et al., 2018). These trends were attributed to an
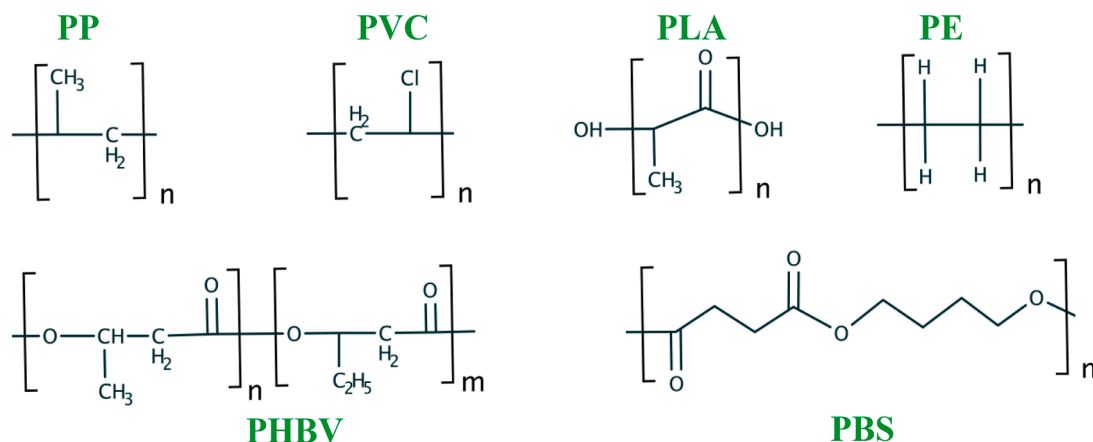

Fig. 2. Chemical structures of representative polymer resins (Fernández-Bravo et al., 2013; Ke et al., 2017; Li et al., 2020; Mohanan et al., 2020; Xu et al., 2019).

Fig. 3. Digital photos of representative natural fibers: jute (reproduced with permission from Sai Shravan Kumar and Viswanath Allamraju, 2019), sisal (reproduced with permission from Sai Shravan Kumar and Viswanath Allamraju, 2019), kenaf (reproduced with permission from Sai Shravan Kumar and Viswanath Allamraju, 2019), copyright © 2019 Elsevier Ltd., flax (reproduced with permission from Mohanty et al., 2018), wood pulp (reproduced with permission from Mohanty et al., 2018), bagasse (reproduced with permission from Mohanty et al., 2018), copyright © 2018 American Association for the Advancement of Science, pine (reproduced with permission from Zhao et al., 2020a), copyright © 2020 American Chemical Society, cellulose (reproduced with permission from Li et al., 2021), copyright © 2021 Elsevier Ltd., and bamboo (reproduced with permission from Maier et al., 2020).

enhanced dispersion and fibrillation of the fibers in their thermoplastic matrices, which was overcome by degradation of the fibers and polymers after repeated thermomechanical cycling.

Conversely, Nadali et al. (2018) observed a decrease and subsequent increase in the tensile modulus upon repeated cycles of extrusion and compression molding of wood fiber/HDPE composites. The wood fiber decreased in size, and the molecular weight of the polymer was reduced with reprocessing, which initially reduced the composite's modulus. However, the subsequent rise in modulus with continued recycling was likely because of combined polymer crosslinking and increased crystallinity as a result of the respective chain scission and smaller, better-dispersed particles. Similarly, Lila et al. (2018) observed a decrease and successive increase in tensile strength and modulus of bagasse fiber/PP composites upon recycling. Although both decreased after reprocessing, the reduction in diameter was larger than the reduction in the length. Therefore, the aspect ratio of the fibers increased during recycling, which corresponded to the observed increase in the composite's tensile properties.

Reprocessing has also been shown to recover or "regenerate" mechanical performance diminished after aging. Soccalingame et al. (Soccalingame et al., 2016, 2015b) produced wood fiber/PP composites that were artificially or naturally aged. Artificial aging was achieved by exposing the samples to prolonged UV irradiation and intermittently spraying them with water, whereas other samples were naturally aged by leaving them outdoors for one year before recycling. In both cases, the UV exposure led to degradation of both the polymer matrix and the wood fiber filler. The artificially aged samples displayed a higher tensile modulus and lower yield strain. This was attributed to a higher degree of crystallization in the polymer, facilitated by its shortened molecular chains due to chain scission. Similarly, the tensile strength of the naturally aged samples drastically decreased. Both types of aged samples were ground and injection molded five times after aging, and the tensile properties of each returned to nearly 100% of their original values. This restoration of mechanical performance was attributed to an improvement in the dispersion of the wood fiber upon reprocessing, as well as increased crystallization and possible crosslinking after chain scission.

Although the recycling studies discussed previously showed that the tensile properties of NFRPCs can fluctuate substantially upon reprocessing, a simple decrease in mechanical performance after recycling is also very common. Researchers have conducted studies on PLA or PLA blends filled/reinforced with sisal, bamboo, cellulose, flax or wood fibers (Åkesson et al., 2016b; Bajwa and Bhattacharjee, 2017; Bourmaud et al., 2016; Chaitanya et al., 2019; Fazita et al., 2015; Graupner et al., 2016; Lagazzo et al., 2019; Ngaowthong et al., 2019). In each study, the tensile strength of the composite samples was drastically reduced upon repeated recycling. This reduction in performance was attributed to fiber fracture as well as degradation of the PLA matrix. PLA is known to be particularly susceptible to hydrolytic degradation at high temperatures (e.g., >50 °C). Natural fibers, such as sisal and cellulose, are hygroscopic and can retain a high degree of moisture even after rigorous drying, which exacerbates the degradation of PLA during reprocessing. Graupner (Graupner et al., 2016) showed this theory by comparing a cellulose/PLA composite recycled three times with a composite of cellulose fiber/virgin PLA, chopped such that their length distribution matched. The mechanical performance of the cellulose/virgin PLA composite far exceeded that of the recycled composite, proving that fiber breakage is not the sole factor contributing to degradation in the composite's tensile properties after reprocessing. Furthermore, an increased amount of wood fiber or cellulose filler was found to degrade PLA more rapidly because a greater percentage of fibers contain more moisture (Åkesson et al., 2016b; Bajwa and Bhattacharjee, 2017).

Several studies have seen similar decreases in the tensile properties of natural fiber/PP composites after recycling. Dickson (Dickson et al., 2014), Abbassi (Ezzahra El Abbassi et al., 2019), Evens (Evens et al., 2019), and Voltz (Rosenstock Voltz et al., 2020) attributed losses in the tensile properties of medium density fiber board (MDFB)/PP, alfa fiber/PP, flax fiber/PP, and wood fiber/PP composites, respectively, to a reduction in the length of the fibers and degradation of both the PP matrix and reinforcing fibers after repeated recycling. Interestingly, the MDFB/PP samples retained their tensile properties better than glass fiber/PP composites.

Additionally, a decrease in the tensile properties of natural fiber-

Table 1

Mechanical property changes in natural fiber composites after recycling (HDPE: high density polyethylene; PLLA: poly(L-Lactic acid)). The reinforcements are assumed as randomly oriented fibers before and after recycling. Typically, natural fiber composites can be recycled approximately 4–6 times until their mechanical performance decreases. PP is a promising polymer that is amenable to multiple reprocessing cycles.

Resin	Fiber (loading: wt%)	Reprocessing methods (number of recycling steps)	Tensile strength and modulus changes	Flexural strength and modulus changes	Other property changes	Ref.
PP	Hemp (30)	Grinding and injection molding (6)	No change	–	Failure strain: no change	(Bourmaud and Baley, 2007)
		Grinding and injection molding (4)	Decrease	Modulus: −10%		(Ezzahra El Abbassi et al., 2019)
	Flax (20)	Grinding and injection molding (10)	No change	Modulus: −10%	Impact strength: increase	(Evens et al., 2019)
	Sugarcane bagasse (20)	Extrusion and injection molding (4)	–	Strength: −14% Modulus: −19%	–	(Correa-Aguirre et al., 2020)
	Sisal (30)	Extrusion and injection molding (4)	No change	–	–	(Ngaowthong et al., 2019)
		Grinding and injection molding (6)	Strength: decrease Modulus: −10%	–	–	(Bourmaud and Baley, 2007)
	Bagasse (20)	Extrusion and injection molding (4)	Modulus: no net change	Strength: no net change	–	(Lila et al., 2018)
PLA	Wood (30)	Extrusion and compression molding (4)	Strength: no change Modulus: decrease	–	Failure strain: no change	(Morreale et al., 2015)
	Sisal (30)	Extrusion and injection molding (2)	Strength: −58% Modulus: −5%	–	–	(Ngaowthong et al., 2019)
		Extrusion and injection molding (8)	Decrease	Decrease	–	(Chaitanya et al., 2019)
		Compounding and compression molding (3)	Strength: decrease	Modulus: increase	Failure strain: decrease	(Lagazzo et al., 2019)
	Cellulose (30)	Shredding and injection molding (3)	Strength: −45%	–	Impact strength: −36%	(Graupner et al., 2016)
	Cellulose (~14)	Extrusion and injection molding (6)	Strength: −30% Modulus: no change	–	–	(Åkesson et al., 2016b)
	Bamboo (35)	Granulation, extrusion, pelletization, and injection molding (1)	Strength: −7%; Modulus: −17%	Strength: +8%; Modulus: +22%	Failure strain or impact strength: decrease	(Fazita et al., 2015)
PLLA	Flax (30)	Extrusion and injection molding (5)	Strength: −70%; Modulus: −8%	–	Failure strain: −78%	(Le Duigou et al., 2008)
PBS/ PLLA	Flax (30)	Grinding and injection molding (6)	Modulus: decrease	–	–	(Bourmaud et al., 2016)
LDPE	Cotton textile (25)	Granulation and extrusion (6)	No net change	–	–	(Bakkal et al., 2012; Bodur et al., 2011)
HDPE	Wood (10)	Grinding and injection molding (6)	Strength: decrease Modulus: no change	Strength: decrease Modulus: no change	Failure strain or impact strength: increase	(Åkesson et al., 2016a)
	Wood (60)	Grinding and injection molding (1)	Strength: decrease Modulus: no change	Strength: −13% Modulus: increase	–	(Shahi et al., 2012)
	Cellulose (48)	Grinding and injection molding (5)	Strength: −50%; Modulus: −37%	–	Failure strain: −28%	(Fonseca-Valero et al., 2015)
	Poplar (40)	Grinding and injection molding (6)	Strength: −15%	Strength: −12%; Modulus: −30%	Impact strength: −27%	(Zhuo et al., 2019)
PHBV	Sisal (30)	Compounding and compression molding (3)	Strength: decrease	Modulus: no change	Failure strain: decrease	(Lagazzo et al., 2019)
	Coffee bean (25)	Solvent dissolution and casting (1)	Decrease	–	Failure strain: decrease	(T et al., 2019)
PA11	Flax (21)	Grinding and injection molding (7)	Strength: −17% Modulus: −28%	–	–	(Gourier et al., 2017)
PVC	Wood (40)	Extrusion and injection molding (19)	–	Strength: +40% Modulus: no change	Failure strain or impact strength: no change	(Augier et al., 2007)

reinforced composites has been noted with HDPE as the matrix polymer. Fonseca-Valero et al. (2015) found a similar diminishing effect of compatibilizer in cellulose/HDPE composites processed with and without maleic anhydride. The tensile modulus of the composites decreased strongly with reprocessing, and the difference in tensile performance between compatibilized and non-compatibilized composites was almost imperceptible after five recycling steps. Scanning electron microscopy and Fourier transform infrared spectroscopy analyses confirmed that the reduction in the tensile modulus was attributed primarily to breakage and oxidative degradation of the cellulose fibers. Bhattacharjee (Bhattacharjee and Bajwa, 2018), Bajwa (Bajwa and Bhattacharjee, 2017), Shahi (Shahi et al., 2012), and Åkesson (Åkesson et al., 2016a) all studied the effect of recycling on wood fiber/HDPE composites. Each found a decrease in the tensile strength of the composites, which was attributed to the damage of both the wood fiber and HDPE matrix.

Although there are numerous examples of degraded tensile performance caused by repeated thermomechanical processing, some researchers have seen an improvement in these properties upon recycling.

For example, Soccalingame et al. (Soccalingame et al., 2015a) noticed a slight increase (~3–6% increase) in the tensile strength of wood fiber/PP composites both with and without maleic anhydride compatibilizer after repeated reprocessing cycles. This enhancement was posited to be because of crosslinking of the PP matrix after chain scission. Morreale et al. (2015) also studied a PLA/copolyester blend reinforced with wood fiber. The tensile properties of the neat polymer remained largely unchanged after recycling, and the tensile performance of this composite far exceeded that of the samples that were recycled with the fibers included from the start. This composite (neat polymer recycled four times plus wood fiber) was then recycled once, after which its elastic modulus increased more than 10%. This further improvement was thought to be attributable to a higher degree fiber dispersion in the polymer matrix, which eases stress transfer between the polymer and high aspect ratio fibers. In both studies, it is unknown whether further recycling would continue to improve the tensile properties, or if they would begin to decrease because of polymer or fiber degradation, fiber breakage, or some other mechanism.

Different types of changes in the tensile properties of NFRPCs have

been discussed, but some researchers have also found properties to remain constant throughout multiple reprocessing cycles. In most cases, this is thought to be attributable to a combination of competing and ultimately neutralizing influences that would improve or degrade the tensile properties if unaccompanied. For example, Ngaowthong et al. (Ngaowthong et al., 2019) found that reprocessing sisal fiber/PP composites had no net effect on their tensile strength or modulus. The aspect ratio of the sisal fibers in this study remained constant because their diameter and length reduced at the same rate during recycling. Similarly, Kaci et al. (Kaci et al., 2009) saw no change in the tensile moduli of wood fiber/PP and flax fiber/PP composites. The authors hypothesized that lignin in the wood acts as a radical scavenger in the presence of the degrading PP and stabilizes the composite from further degradation. Furthermore, the dispersion quality of the wood fiber in the PP matrix was seen to improve upon reprocessing.

2.1.2. Flexural properties

The flexural properties of NFRPCs can display similar trends in response to reprocessing as the tensile properties. However, the stress distribution and failure mode in flexure or bending is different from that in tension. During a tensile test, the entirety of the sample is under constant axial stress, and failure will occur by propagating from the largest defect in the sample. However, during a flexural test, the maximum stress occurs at the upper and lower surface of the specimen, where shear stress is at a minimum. If the largest defect in the sample is not located on these surfaces, its influence on the failure mechanism and flexural properties of the material may be minimal (Correa-Aguirre et al., 2020).

For example, Uitterhaegen et al. (2018) found that the impact of multiple reprocessing cycles on the flexural properties of PP and PE composites reinforced with coriander straw fiber was consistently lower than that on the tensile properties. Furthermore, aging the samples through exposure to UV irradiation and temperature cycles at a high humidity increased the flexural strength of the PP composites by 30%, whereas little effect was seen on the tensile properties. This effect is likely due to photo- and/or thermo-oxidation of the polymer, resulting in chain scission that leads to a lower molecular weight and the formation of carbonyl groups. Reduction in the molecular weight of a polymer is typically detrimental to its mechanical properties, but it was thought that the adhesion between the polymer matrix and fiber filler was greatly improved after aging because crosslinking likely occurred between the newly formed carbonyl groups and the lignin fraction of the fiber fillers. As UV irradiation and oxidation largely affect the surface of exposed samples, the core of the polymer, which largely dominates the tensile properties, may remain unchanged. Upon reprocessing of these aged samples, the flexural properties of the composites showed an increasing and subsequent decreasing trend, as was seen in many studies on the tensile properties of recycling composites. Again, this fluctuating performance is attributable to competing consequences of recycling. Bundles of fibers initially present in the polymer were broken up and dispersed more evenly upon recycling, but the fiber length was also reduced, and the polymer matrix and fibers themselves began to degrade with successive reprocessing cycles.

Correa-Aguirre (Correa-Aguirre et al., 2020) and Fazita (Fazita et al., 2015) observed fluctuations in the flexural properties of bagasse fiber/PP and bamboo fiber/PLA composites, respectively, upon recycling. An increase and subsequent decrease in the flexural modulus of both composites after repeated reprocessing was attributed to better dispersion of fibers in the polymer matrices and increased crystallinity because of this dispersion and decreased molecular weight. The detrimental effect of the decreased molecular weight likely outweighed the positive effects of recycling on the flexural performance after multiple cycles in both cases. Similarly, Zhuo et al. (Zhuo et al., 2019) found that the flexural strength and modulus of 40 wt% poplar fiber/HDPE composites first increased and then decreased with successive grinding and injection molding cycles. The flexural strength increased by ~13% and

then decreased by ~11%. The initial improvement in the flexural properties was attributed to an enhanced distribution of the poplar fibers in the HDPE matrix, but this effect was eventually diminished by degradation and fracture of the poplar fibers and polymer chains. Finally, Augier et al. (Augier et al., 2007) documented a 40% increase in the flexural strength of pine fiber/PVC composites after 20 extrusion and injection molding cycles. Thermal degradation in the PVC was thought to dechlorinate its backbone, leading to crosslinking between the chains and a perceived increase in molecular weight. Additionally, the aspect ratio of the fibers remained constant throughout reprocessing, whereas their dispersion in the polymer matrix improved.

Although the flexural and tensile property changes in response to reprocessing can differ, numerous authors have found both sets of properties to decline with repeated reprocessing. Beg (Beg and Pickering, 2008b), Wang (Wang et al., 2019), Evens (Evens et al., 2019), Abbassi (Ezzahra El Abbassi et al., 2019), Srebrenkoska (Srebrenkoska et al., 2008), and Lila (Lila et al., 2018) all saw the tensile and flexural performance of PP composites reinforced with fibers (e.g., wood, flax, alfa, rice hull, kenaf, bagasse) decline with successive mixing and molding cycles. These losses were attributed to numerous factors, including a reduction in the molecular weight of PP, as well as degradation and shortening of the reinforcing fibers.

Chaitanya (Chaitanya et al., 2019) and Bhattacharjee (Bhattacharjee and Bajwa, 2017) investigated PLA composites reinforced with wood fibers and sisal fibers, respectively. The flexural strength and modulus of the composites declined with repeated extrusion and injection molding attributable to fiber length reduction and degradation of the PLA matrices. Similarly, Shahi (Shahi et al., 2012) and Bhattacharjee (Bhattacharjee and Bajwa, 2018) found the flexural performance of wood fiber/HDPE composites to decline upon recycling. Interestingly, Shahi reported that the wetting of HDPE on the wood fiber particles increased upon recycling (grinding and injection molding), whereas Bhattacharjee found the adhesion between polymer and filler to degrade after recycling (extrusion and injection molding). The samples in Bhattacharjee's study were recycled six times, whereas Shahi performed only one reprocessing cycle; and Bhattacharjee's samples contained 30 wt% wood fibers, while Shahi's contained 60 wt%. These factors, as well as processing temperatures, speeds, and the sources and initial properties of the PP and wood fiber used in either study, can affect the mechanical properties before and after recycling and could be responsible for this discrepancy.

2.1.3. Impact strength

Impact strength can also be affected during recycling of NFRPCs. For example, Uitterhaegen (Uitterhaegen et al., 2018), Gourier (Gourier et al., 2017), Evens (Evens et al., 2019), Soccalingame (Soccalingame et al., 2015b), and Correa-Aguirre (Correa-Aguirre et al., 2020) all noted a significant increase in the impact strength of PP composites filled with coriander straw fibers, flax fibers, wood fibers, and bagasse fibers after recycling. Increases in the impact strength of an NFRPC after recycling can be attributed to a reduction in the fiber length, which renders the composite more ductile. In this manner, a decrease in the modulus and strength in tension and flexure can often accompany an increase in impact strength, as well as strain at break. Additionally, a degradation of the interface between the fiber and polymer matrix after reprocessing likely improves its impact properties. Fibers that are able to debond from their polymer matrices upon impact are thought to allow the composite to absorb more energy before failing.

Antithetical to these findings, some researchers have documented a decrease in the impact strength of natural fiber-reinforced composites after reprocessing. Graupner (Graupner et al., 2016) and Fazita (Fazita et al., 2015) noted deterioration of the impact strengths of PLA composites reinforced with cellulose and bamboo fibers, respectively, after reprocessing. Interestingly, Wang et al. (Wang et al., 2019) noted a decrease in impact strength of wood fiber/PP composites with recycling, whereas Soccalingame et al. (Soccalingame et al., 2015b) reported an

increase in the impact strength of the recycling of these composites. Soccalingame et al. used PP with either 10 or 30 wt% of wood fibers, whereas Wang prepared PP composites with 60 wt% wood fibers. Both studies used maleic anhydride-grafted PP (MAPP) as a compatibilizer, and the composites were subjected to a similar number of recycling steps (five in Soccalingame's study versus seven in Wang's). Wang noted significant damage to the wood fiber in the composites after recycling, which was not reported by Soccalingame et al. The significantly higher filler content in the composites studied by Wang may have accelerated their degradation during reprocessing because of higher shear forces during mixing.

2.2. Thermal properties

As discussed in previous sections, thermomechanical or chemical reprocessing of composites can damage both the polymer matrix and the reinforcing agent or filler. This typically results in a decrease of the polymer's molecular weight and shorter, but more well-dispersed, fibers throughout the matrix. Changes in the size, distribution, and morphology of reinforcing fibers, as well as the length of polymeric chains, have a strong effect on a material's thermal properties, including crystallinity, thermal stability, and thermal transition temperatures such as glass transition (T_g), crystallization (T_c), and melting (T_m) temperatures. These thermal properties are most often measured using differential scanning calorimetry and thermogravimetric analysis and can directly affect the material's mechanical performance, as well as its overall recyclability (the degree to which a material might maintain its performance with respect to a given application over the course of multiple reprocessing cycles) (Beg and Pickering, 2008b; Bhattacharjee and Bajwa, 2017; Lagazzo et al., 2019; Lila et al., 2018; Soroudi and Jakubowicz, 2013; T et al., 2019). A summary of the changes in thermal properties for a variety of NFRPCs discussed in this section is provided in Table 2.

2.2.1. Crystallization upon cooling and heating

In addition to the degree of crystallinity in a sample, changes in the structure of the polymer matrix and reinforcing fibers of a composite after reprocessing can also alter the crystallization behavior and kinetics upon heating and cooling. Crystallization in thermoplastics is most commonly thought of as a phenomenon occurring while cooling from a melt, when molecular chains can diffuse into lower energy, ordered regions as they lose thermal energy and mobility. The temperature at which this crystallization occurs depends on the energy required for polymer chains to diffuse through the melt. A reduction in the molecular weight of a polymer because of degradation during recycling typically increases the polymer chains' mobility and, accordingly, their ability to crystallize. Numerous researchers have found that the crystallization temperature of natural fiber-reinforced thermoplastic composites shifts to lower values as the material is reprocessed because the degraded, shorter polymer chains are more mobile at lower temperatures. For example, Åkesson et al. (Åkesson et al., 2016b) saw a drop of almost 20 °C in the crystallization of PLA reinforced with ~14 wt% cellulose fibers after six cycles of extrusion and injection molding. The drop in crystallization temperature was attributed to the significant reduction in the molecular weight of the PLA, which was reduced from 29 to 13 kDa after recycling, as determined by nuclear magnetic resonance measurements. Similarly, Dhar (Dhar et al., 2018) and Chaitanya (Chaitanya et al., 2019) noted reductions in the molecular weight of PLA in their studies on the recycling of composites reinforced with cellulose nanocrystals and sisal fibers. Reductions in the crystallization temperatures upon recycling have also been reported in cotton/LPDE (Bakkal et al., 2012; Bodur et al., 2011), sisal fiber/PP (Ngaowthong et al., 2019), and wood fiber/HDPE (Bhattacharjee and Bajwa, 2018) composites.

Crystallization can also be observed upon heating of a sample and usually occurs if a material was cooled too rapidly to crystallize; this phenomenon is commonly referred to as cold crystallization. As a material is heated above its glass transition temperature, the chains gain mobility and can diffuse to a lower energy crystalline state before they are again melted at a higher temperature. Some researchers have also

Table 2

Thermal property changes in natural fiber composites after recycling (T_d: degradation temperature). Typically, natural fiber composites can be recycled approximately 4–6 times until their thermal property changes.

Resin	Fiber (loading: wt%)	Reprocessing methods (number of recycling steps)	T_m change	T_g change	T_d change	T_c change	Crystallinity change	Ref.
PP	Hemp (30)	Grinding and injection molding (6)	–	–	–	–	No change	(Bourmaud and Baley, 2007)
	Sugarcane bagasse (20)	Extrusion and injection molding (4)	No change	No change	Decrease	–	No change	(Correa-Aguirre et al., 2020)
	Sisal (30)	Extrusion and injection molding (4)	No change	–	–	Decrease	+2.5%	(Ngaowthong et al., 2019)
		Grinding and injection molding (6)	No change	–	–	No change	–	(Bourmaud and Baley, 2007)
	Bagasse (20)	Extrusion and injection molding (4)	–	–	No change	–	Increase	(Lila et al., 2018)
PLA	Sisal (30)	Extrusion and injection molding (2)	Decrease	–	–	Increase	+20%	(Ngaowthong et al., 2019)
		Extrusion and injection molding (8)	Decrease	Decrease	Decrease	Decrease	–	(Chaitanya et al., 2019)
	Cellulose (~14)	Extrusion and injection molding (6)	–	Decrease	–	Decrease	–	(Åkesson et al., 2016b)
	Bamboo (35)	Granulation, extrusion, pelletization, and injection molding (1)	No change	–	–	–	Decrease	(Fazita et al., 2015)
PLLA	Flax (30)	Extrusion and injection molding (5)	–	Decrease	–	–	–	(Le Duigou et al., 2008)
PBS/ PLLA	Flax (30)	Grinding and injection molding (6)	–	−20%	–	–	+80%	(Bourmaud et al., 2016)
LDPE	Cotton textile (25)	Granulation and extrusion (6)	Increase	–	–	Decrease	Increase	(Bakkal et al., 2012; Bodur et al., 2011)
HDPE	Wood (10)	Grinding and injection molding (6)	Decrease	–	–	–	Increase	(Åkesson et al., 2016a)
	Wood (60)	Grinding and injection molding (1)	–	–	Decrease	–	–	(Shahi et al., 2012)
	Poplar (40)	Grinding and injection molding (6)	Decrease	–	–	–	Increase	(Zhuo et al., 2019)
PHBV	Sisal (30)	Compounding and compression molding (3)	Decrease	–	–	–	–	(Lagazzo et al., 2019)
	Coffee bean (25)	Solvent dissolution and casting (1)	Decrease	–	Decrease	Decrease	–	(T et al., 2019)
PA11	Flax (21)	Grinding and injection molding (7)	No change	–	–	No change	–	(Gourier et al., 2017)
PVC	Wood (40)	Extrusion and injection molding (19)	–	Increase	Decrease	–	–	(Augier et al., 2007)

reported shifts in the cold crystallization of recycled composites to lower temperatures with increased reprocessing. Beltran et al. (Beltran et al., 2018) noted a 5 °C decrease in the cold crystallization temperature after re-extruding and compression molding a neat PLA sample, while Ngaowthong et al. (Ngaowthong et al., 2019) reported a constant decrease in the cold crystallization temperature of sisal fiber/PLA composites with increased fiber loading. This indicates that a reduction in molecular weight facilitates cold crystallization, and the presence of fibers - which serve as nucleation sites - can aid crystallization upon heating. With increased fiber loading, the cold crystallization exotherm, as noted by differential scanning calorimetry scans, broadened and shifted to lower temperatures until it disappeared altogether, indicating that with increased fillers, the composite was able to sufficiently crystallize upon cooling because of the nucleating ability of the fibers. The composites also showed a similar trend upon reprocessing in which the cold crystallization exotherm shifted to lower temperatures with an increasing number of recycling steps until it disappeared completely.

2.2.2. Glass transition and melting

Because the glass transition and melting behavior of a polymer are closely tied to the mobility of its molecular chains, both the glass transition and melting temperatures of composites are affected by changes in its structure after reprocessing. Many researchers have reported a reduction in T_g and T_m after reprocessing, which can be understood through the same mechanism by which the crystallization temperatures were noted to decrease. Bourmaud et al. (Bourmaud et al., 2016) noted a 7% decrease in the glass transition temperature of neat PLA after seven cycles of extrusion and injection molding. Upon the addition of flax fiber at 30 wt%, the T_g of the composite was observed to decrease 23% when subjected to the same recycling methods. Similarly, Chaitanya (Chaitanya et al., 2019), Åkesson (Åkesson et al., 2016b), and Le Duigou (Le Duigou et al., 2008) reported drops in the T_g of PLA composites reinforced with sisal, cellulose, and flax fibers after multiple cycles of extrusion and injection molding. Similar results have been noted after recycling of wood fiber/PP composites (Wang et al., 2019). In each case, a reduction in T_g after recycling was attributed to a rise in the mobility of molecular chains attributable to polymer degradation. Polymer degradation can increase the number of chain ends (Gewert et al., 2015). Another possible reason for T_g reduction is some extractives (e.g., wax) can detach from natural fibers and act as a plasticizer.

In another study on sisal fiber/PLA composites, Ngaowthong et al. (Ngaowthong et al., 2019) reported a stable value for T_g after reprocessing, but a notable drop in T_m. A reduction in the melting temperature of composites after recycling has been reported by others for sisal fiber/PLA composites (Chaitanya et al., 2019), as well as for sisal fiber/PHBV (Lagazzo et al., 2019), wood fiber/PP (Rosenstock Voltz et al., 2020), and poplar fiber/HDPE (Zhuo et al., 2019) composites, among others. Some researchers, however, have detailed an increase in T_m after composite recycling. Bhattacharjee (Bhattacharjee and Bajwa, 2018), Bodur (Bodur et al., 2011), and Bakkal (Bakkal et al., 2012) studied the effect of recycling on the thermal properties of wood fiber/HDPE and cotton/LDPE composites, finding that the T_m of the composites rose 1–2 °C after six recycling steps. Bhattacharjee attributed this behavior to a severe degradation and possible charring of the fibers after repeated thermomechanical cycling. Interestingly, Bhattacharjee's study also reported an increase in the crystallinity of the wood fiber/HDPE composites after recycling, which was attributed to fiber degradation.

Finally, researchers have reported the appearance of a double endothermic melting peak in natural fiber-reinforced composites after reprocessing. In studies of neat PLA and flax fiber/PLLA/PBS composites (Beltran et al., 2018; Bourmaud et al., 2016), the appearance of double endothermic peaks after recycling was attributed to polymer degradation, which led to the formation of imperfect crystals at lower temperatures during cooling. Upon heating, these crystals can melt at lower temperatures, reorganize into more stable crystals, and then melt again

at higher temperatures.

2.2.3. Thermal stability

The thermal stability of a composite after reprocessing is often an important factor in assessing its recyclability. A material is generally more thermally stable if it can withstand higher temperatures before degrading, which is typically measured using thermogravimetric analysis. Reprocessing of natural fiber-reinforced composites can increase or decrease the thermal stability of the materials depending on the extent of degradation of the polymer, fibers, and their interface. As discussed previously, this seemingly adverse degradation may lead to increased crystallinity, crosslinking, and/or a stronger reinforcing effect because of enhanced dispersion and wetting of the fibers by the matrix polymer.

Beg and Pickering (Beg and Pickering, 2008b) reported an increase in the degradation temperature of wood fiber/PP composites after reprocessing, which was attributed to an increase in crystallinity of the PP after its molecular weight was reduced because of degradation. A similar result was noted by Bhattacharjee and Bajwa (Bhattacharjee and Bajwa, 2018) in a study on the recycling of wood fiber/HDPE composites in which the composite decomposition continuously shifted to higher temperatures throughout six cycles of extrusion and injection molding. Srebrenkoska et al. (Srebrenkoska et al., 2008), however, associated higher degradation temperatures of kenaf fiber/PP composites after recycling to an apparent increase in the polymer's molecular weight after crosslinking between molecular chains, as well as molecular chains and fiber surfaces. Crosslinking can occur because of reprocessing-induced degradation when free radicals are released from severed chains. Another explanation for an increase in the thermal stability of natural fiber-reinforced composites after reprocessing was given by Nadali et al. (Nadali et al., 2018). The shift in degradation temperatures of wood fiber/HDPE composites after four cycles of extrusion and compression molding was likely influenced by the reduction in hemicellulose groups in the wood fiber after reprocessing. Hemicelluloses are one of the most hydrophilic constituents (Wang et al., 2021) of wood fiber; they hold a substantial number of hydroxyl groups, which also makes them highly susceptible to thermal degradation. A reduction in hemicellulose within wood fiber, therefore, can lead to a less hydrophilic nature and result in a more thermally stable composite. Zhuo et al. (Zhuo et al., 2019) analogously attributed an increase in the thermal degradation temperature of poplar fiber/HDPE composites after recycling to a loss of volatile small molecule substances in the poplar fibers within the first or second stages of recycling, and saw higher shifts after reprocessing composites with higher fiber loadings.

Conversely, many researchers have found reprocessing to negatively affect the thermal stability of natural fiber composites. A shift in the onset of thermal degradation toward lower temperatures after recycling was reported in multiple types of composites, including those prepared with PLA, PP, HDPE, and PVC (Augier et al., 2007; Chaitanya et al., 2019; Correa-Aguirre et al., 2020; Ngaowthong et al., 2019; Rosenstock Voltz et al., 2020; Shahi et al., 2012). In most cases, this loss of stability was attributed to severe degradation of the polymer matrix and/or the reinforcing fibers, indicating again that recycling-induced degradation can be beneficial to a certain degree, but can often begin to degrade the composite's mechanical and/or thermal performance after continuous reprocessing.

2.2.4. Thermal expansion and heat deflection behavior

Volumetric or conformational changes in bulk composite materials after reprocessing are less often studied than more traditional thermal properties, such as glass transition and crystallization, but they are nonetheless important in practical applications. The expansion of a material during heating, defined by its coefficient of thermal expansion (CTE), is particularly important when composites are used in close contact or when adhered to other materials subjected to elevated temperatures. Differing thermal expansion behavior between two materials in contact can lead to debonding and cracking. Bhattacharjee

(Bhattacharjee and Bajwa, 2018) and Bajwa (Bajwa and Bhattacharjee, 2017) both reported increases in the CTE of wood fiber/HDPE composites with repeated extrusion and injection molding. The increased CTE was associated with a reduction in the fiber lengths, which reduced the restriction of the polymer expansion upon heating. Additionally, the researchers theorized that oxidation of the polymer during reprocessing could lead to weaker intermolecular bonds and a higher degree of thermal expansion. Bhattacharjee et al. also reported a decrease in the heat deflection temperature (HDT) of their composites with increased recycling, meaning the composites began to deform under load at lower temperatures after reprocessing. Again, this was likely attributable to degradation of the polymer, fibers, and their interface, which gave the composite increased molecular mobility with a lower thermal energy contribution.

2.3. Degree of crystallinity

Bodur (Bodur et al., 2011), Bakkal (Bakkal et al., 2012), and Zhuo (Zhuo et al., 2019) each reported an increase in the crystallinity of cotton/LDPE and poplar fiber/HDPE composites after six reprocessing cycles. In the study by Zhuo (Zhuo et al., 2019), poplar fiber was added to composites at a range of 10 to 40 wt% of HDPE. The increase in crystallinity upon recycling was larger in higher loaded composites, indicating that an increased number of fibers provides more nucleation sites for crystal growth within a polymer melt. Furthermore, a higher filler content is known to increase the shear forces within the composite during reprocessing, accelerate the degradation of the polymer and hasten the fracture of the reinforcing fibers. A lower molecular weight polymer and larger number of fibers due to breakup can increase the crystallinity in the composite after reprocessing.

Similarly, numerous authors (Beg and Pickering, 2008b; Correa-Aguirre et al., 2020; Lila et al., 2018; Rodrigues et al., 2013; Soccalingame et al., 2015b; Srebrenkoska et al., 2008) have reported increases in crystallinity after multiple reprocessing cycles of PP composites reinforced with wood fibers, rice hulls, kenaf fibers, and bagasse fibers. Every study connected the increased crystallinity to an enhanced nucleation effect of the fibers after better dispersion in the composite and/or an increased molecular mobility because of polymer degradation. Additionally, Rodrigues et al. theorized that oxidation of the polymer chains during reprocessing could increase the molecules' polarity, which would then increase the rate of crystal nucleation. In the study conducted by Correa-Aguirre et al. (Correa-Aguirre et al., 2020), bagasse fibers were treated with NaOH and silanized before being incorporated into a PP composite and recycled. The crystallinity after recycling with the silanized fiber composites was greater than that observed after recycling composites filled with untreated fibers or those treated only with NaOH, suggesting that the silanization improved the nucleating effect of the bagasse fibers.

Other authors (Bourmaud and Baley, 2007; Ngaowthong et al., 2019; Soccalingame et al., 2015a) studying the crystallization after recycling of PP composites reinforced with sisal, hemp, and wood found that there was no change in the crystalline content of the composites after recycling. Each study used a relatively low filler content (between 10 and 30 wt%) and noted no significant changes in the tensile or flexural properties of the composites after reprocessing. It is likely that a combination of factors including processing methods and parameters, the use of compatibilizers or fiber pretreatments, and the initial properties of the polymer and fibers such as molecular weight and aspect ratio, respectively, allowed these composites to be recycled multiple times without presenting any degradation in their properties. Based on the findings of other researchers, it is likely that after some number of recycling steps in each case, the properties of the composites would begin to change. Eventually the fibers or the polymer matrix start to degrade, which can raise or lower the crystallinity depending on each polymers' ability to crosslink and the degree of fracture and dispersion of the fibers.

In another study, Bourmaud et al. (Bourmaud et al., 2016) found that

crystallization of a PBA/PLLA blend reinforced with 30 wt% of flax fibers increased more than 20% after five reprocessing cycles. In a similar study of PLLA reinforced with 20 or 30 wt% of flax fibers conducted by Le Duigou et al. (Le Duigou et al., 2008), the crystallinity of the 20 wt% flax fiber composite increased continuously with recycling. However, the crystallinity of the more highly loaded composite was found to increase and then decrease. At a higher fiber volume fraction, the quantity of polymer of the composite could reduce, leading to a decrease in crystallinity (Husnil et al., 2019). The same trend was seen in the melting enthalpy of this composite measured by differential scanning calorimetry. The authors were unable to give a complete explanation of this phenomenon but theorized that either the size or dispersion of fibers in the more highly loaded composite hindered crystallite growth. It was also possible that the polymer was too heavily degraded after multiple recycling steps to fully crystallize.

A decrease in the crystallinity of natural fiber/polymer composites after reprocessing has been documented by authors, although it seems to be a rare occurrence. Fazita et al. (Fazita et al., 2015) reported a decrease in crystallinity from 11% to 4% in bamboo fiber/PLA composites after just one cycle of granulation, extrusion, and injection molding. Similar to the aforementioned work with flax/PLLA composites, they attributed the drop in crystallinity to a limitation of crystallite growth due to the enhanced dispersion of fibers in the polymer matrix, theorizing that the longer polymer chains were unable to mobilize adequately to crystallize. A comparison in the molecular weight of the matrix polymers and the fiber lengths at different stages during recycling may be enlightening as to the apparent chain mobility and possible impediment to crystallization caused by the fibers. However, such a comparison is not available. Finally, Bhattacharjee and Bajwa (Bhattacharjee and Bajwa, 2018) documented a decrease in the crystallinity of wood fiber/HDPE composites after seven cycles of extrusion and injection molding. Extensive degradation of the wood fibers was thought to be responsible for the drop in crystalline content, suggesting that fibers in a composite could break down to a degree after which they no longer favor crystal nucleation and growth.

2.4. Hygroscopic behavior

A major application of NFRPCs is in outdoor infrastructural building and construction materials, such as railing, decking, and fencing. Because of the hydrophilic nature of natural fibers, they can absorb/desorb moisture/water, which causes either swelling or shrinking. Hence, NFRPCs experience dimensional instability, which will result in physical and aesthetical (warping) downgrading. The hygroscopic behavior also lowers the mechanical properties and durability of the NFRPCs (Tajvidi and Takemura, 2010). There are mainly three moisture penetration paths: (1) gaps/flaws at the interface of fibers and polymers, (2) pores in the fibers, and (3) micro-voids in the polymer matrix (Beg and Pickering, 2008c). Factors such as natural fiber or polymer type, fiber morphology and surface characteristics, recycling methods, interfacial compatibility, and matrix crystallinity all are important characteristics in determining the hygroscopic behavior of recycled NFRPCs.

One method to characterize the hygroscopic behavior of reprocessed NFRPCs is the water absorption test (Tajvidi and Takemura, 2010). During such a test, NFRPCs are submerged in distilled water and weighed periodically. The water absorption percentage is calculated as follows:

$$Water\ absorption\ percentage = \frac{W_t - W_0}{W_0} \times 100\% \qquad (1)$$

where, W_t is the weight of a specimen at a given time and W_0 is the dry weight of a specimen.

Another method is to measure the thickness swelling of NFRPCs after being submerged in water for a certain period (Tajvidi and Takemura, 2010). The thickness swelling percentage is calculated as follows:

$$\text{Thickness swelling percentage} = \frac{T_t - T_0}{T_0} \times 100\% \qquad (2)$$

where T_t is the thickness of a specimen at a given time and T_0 is the dry thickness of a specimen at the beginning of the test. Swelling may be anisotropic if the fibers lie predominantly in plane (Rafsanjani et al., 2014).

Although thickness swelling is caused by water absorption, the relationship between the two is not linear. Thickness swelling was reported to be fiber-type dependent. For example, composites containing rice hulls have been found to swell less than those containing wood fiber (Tajvidi and Takemura, 2010). Rice hulls contain a high silica content that contributes to this behavior.

Numerous articles have reported that reprocessing NFRPCs can inhibit their hygroscopic behavior (Table 3) (Beg and Pickering, 2008b, 2008c; Tajvidi and Takemura, 2010, 2011; Viksne and Rence, 2008; Zhuo et al., 2019). For example, reprocessing using equipment with high fiber dispersion capacity, such as extrusion, was reported to decrease the water absorption percentage significantly (Viksne and Rence, 2008). The researchers attributed the reduced water absorption percentage of reprocessed NFRPCs to the improved dispersion of wood fiber. Since more fibers were separated and protected by polymer wrapping on their surface, moisture cannot access the fibers extensively. In another two studies, the authors attributed the reduced water absorption percentage/thickness swelling percentage to factors such as increased density of the NFRPCs by reducing the defects in the fibers and increasing the crystallinity of the polymer matrix, and improved interfacial compatibility between the fibers and the matrix (Beg and Pickering, 2008b, 2008c). Tajvidi and Takemura also performed experiments on reprocessed natural fiber/polyolefin composites and discovered a similar trend in hygroscopic behavior change (Tajvidi and Takemura, 2010, 2011). A better fiber dispersion was reported after recycling NFRPCs (Fig. 4A and B). Additionally, reprocessing can lower the equilibrium moisture content of NFRPCs and cause them to lose moisture faster during the water

desorption test (Tajvidi and Takemura, 2010, 2011), which is speculated to be related to the increased hydrophobicity of materials after recycling. Such a change is associated with the reduction of the most hydrophilic component of NFRPCs, hemicellulose, during reprocessing because of thermal degradation (Hammiche et al., 2016). On the contrary, Shahi et al. (Sarabi et al., 2014; Shahi et al., 2012) reported a decrease in water resistance and density after only one reprocessing step of 60% wood/HDPE composites. Authors noticed significant decomposition of the NFRPCs at the reprocessing stage with thermogravimetric analysis measurements, which explained the change. The cause of the decomposition may be the slow extrusion speed (2 rpm).

Few researchers have investigated the hygroscopic behavior of NFRPCs manufactured from already-recycled plastics without further reprocessing. Turku et al. (Turku et al., 2017) produced wood-filled PP/PE composites out of construction and municipal plastic waste. The ratio of PP to PE was varied to examine its effect on the product's performance. They reported water absorption percentage and thickness swelling percentage of all formulations seemed to be comparable to NFRPCs processed from virgin plastics.

2.5. Viscoelastic properties

One of the environmental conditions NFRPCs experience during outdoor applications is temperature fluctuation. Similar to other polymer composites, NFRPCs undergo property changes when the temperature changes. It is important to understand such behavior to help scientists/engineers design sufficient products to guarantee service life. Dynamic mechanical analysis is often used to characterize the viscoelastic properties of NFRPCs. During this test, the elastic modulus and loss modulus are recorded as a function of temperature. The loss factor, tan δ, is calculated from the ratio of loss modulus to elastic modulus. The T_g of the tested material can then be defined as the temperature corresponding to the peak of the loss factor curve, although T_g can also be calculated from the elastic modulus curve.

Table 3

Summary of hygroscopic behavior of NFRPCs after recycling/reprocessing. In general, the fiber length, -OH density, and thickness swelling decrease after recycling, while the matrix crystallinity increases. The water absorption ($\geq$200 h) decreases after recycling, likely because of the composites' hydrophilic substances reduction.

Composite composition (wt%)	Reprocessing methods (number of recycling steps)	Fiber length (mm)	-OH density	Composite density (g/cm³)	Matrix crystallinity (%)	Water absorption (%)	Thickness swelling (%)	Ref.
Wood flour (50) MAPP (5) PP (45)	Batch mixing (5) Extrusion (5)	– –	– –	– –	– –	3.5→1.0 (400 h) 1.4→1.3 (400 h)	– –	(Viksne and Rence, 2008) (Viksne and Rence, 2008)
Wood fiber (40) MAPP (4) PP (56)	Pelletizing and injection molding (8)	2.4→0.4	–	1.05→1.1	44→45	4.0→2.5 (50 days)	3.0→2.3	(Beg and Pickering, 2008b, c)
Wood flour (50) MAPP (2) PP (48)	Pelletizing, mixing and compression molding (5)	0.5→0.2	1.4→1.0	0.86→1.04	–	23→9 (200 h)	6.8→5.9 (200 h)	(Tajvidi and Takemura, 2010)
Wood flour (50) MAPE* (2) HDPE (48)	Pelletizing, mixing and compression molding (5)	0.5→0.2	0.9→0.8	1.01→1.10	–	17→6 (200 h)	7→5 (200 h)	(Tajvidi and Takemura, 2011)
Wood flour (60) HDPE (40)	Pelletizing and profile extrusion (1)	–	–	1.13→1.08	–	3.1→4.5 (24 h)	–	(Shahi et al., 2012)
Wood sawdust (60) HDPE (40)	Pelletizing and profile extrusion (1)	–	–	1.13→1.09	–	3.1→4.6 (24 h)	–	(Sarabi et al., 2014)
Wood fiber (40) HDPE (60)	Grinding and injection molding (6)	–	–	–	61→74	1.9→1.2 (10 days)	–	(Zhuo et al., 2019)
Wood flour (65) PVC (30) Additive (5)	Batch mixing and compression molding (4)	0.5→0.2	1.6→1.3	–	–	7.5→5.5 (~200 h)	–	(Hammiche et al., 2016)
Wood flour (54) MAPE (3) Recycled PP/PE (40) Lubricant (3)	Extrusion and injection molding (0)	–	–	–	–	12 (28 days)	2 (28 days)	(Turku et al., 2017)

* MAPE: Maleated PE.
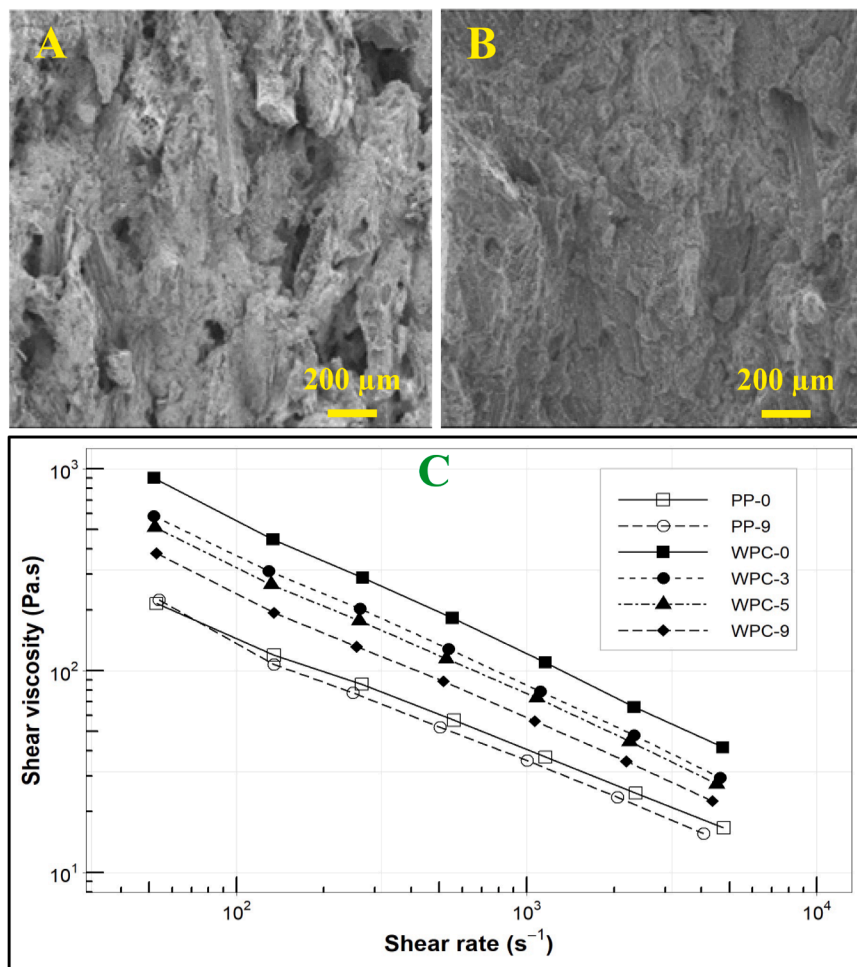
Fig. 4. The surface morphology and shear viscosity of materials before and after recycling. Scanning electron microscopy images of the fracture surface of wood fiber/PE composites: (A) before recycling and (B) after being recycled five times. The fiber dispersion was found to be improved after the recycling (reproduced with permission from Tajvidi and Takemura, 2011). Copyright © 2011 John Wiley and Sons. (C) Variation of shear viscosity with shear rate for PP and wood-fiber thermoplastic composite (WPC) before and after recycling (after the 3rd, 5th, and 9th recycling steps; graph reproduced via the WebPlotDigitizer software). The reduction in viscosity with recycling steps could be attributed to polymer degradation and fiber shortening (reproduced with permission from Rosenstock Voltz et al., 2020).

Morreale et al. studied the dynamic mechanical properties of wood fiber-filled PLA composites after reprocessing (Morreale et al., 2015). After one reprocessing step, the elastic modulus of wood fiber (15 wt %)/PLA improved from 286 to 296 MPa, and the loss modulus improved from 22 to 25 MPa. Such a change mainly came from the improved fiber dispersion after reprocessing. For reprocessed wood fiber (65 wt%)/PVC composites after three cycles, T_g (from tan δ) moved toward higher temperatures (from 45 to 50 °C), which indicated a stronger interaction between fibers and polymers at the segmental chain level (Hammiche et al., 2016). Bhattacharjee and Bajwa, on the other hand, discovered that the elastic modulus (at 30 °C) of wood (30 wt%)/PLA composite decreased from 6.4 to 5.7 GPa after six cycles of reprocessing by pelletizing, extrusion, and injection molding (Bhattacharjee and Bajwa, 2017). The authors attributed the performance decrease to a few factors, including fiber/polymer degradation, increased number of micro-voids, and poor interfacial adhesion. It appears that the effects of reprocessing on the dynamic mechanical properties are directly related to the number of reprocessing cycles. Indeed, Chaitanya et al. found that the elastic modulus of wood (30 wt%)/PLA composites improved after two reprocessing steps, while a gradual decline of elastic modulus occurred after the third reprocessing step until up to eight reprocessing cycles (Chaitanya et al., 2019). The loss modulus of the wood/PLA composite increased each time during three reprocessing cycles and gradually decreased after the fourth reprocessing cycles. The tan δ of NFRPCs was reported to increase as reprocessing continued in most cases, indicating that reprocessing magnified the viscous tendency of the NFRPCs (Chaitanya et al., 2019; Hammiche et al., 2016; Morreale et al., 2015; Nadali and Naghdi, 2020). Therefore, reprocessing or recycling will

likely negatively impact the long-term creep performance of NFRPCs. Aydemir et al. (Aydemir et al., 2015) addressed the reprocessing of foam NFRPCs produced by reactive extrusion of wood fiber and styrene maleic anhydride. The elastic modulus and tan δ of the composite foam both decreased significantly after just one reprocessing cycle (Aydemir et al., 2015).

Other studies have shown that using slightly recycled polymers (e.g., recycled < 5 times) to produce NFRPCs does not seem to degrade their viscoelastic properties. For example, the elastic modulus of the wood fiber (15 wt%)/recycled PLA composite increased from 286 to 296 MPa, and the loss modulus increased from 22.1 to 23.4 MPa, compared with wood fiber (15 wt%)/virgin PLA (Morreale et al., 2015). Similarly, an array of natural fiber (5–20 wt%)/recycled PP composites, including waste paper, cardboard, and wood fiber, showed very slight changes in tan δ and elastic modulus as compared with the composites made from virgin PP (Zander et al., 2019).

2.6. Rheological properties

Rheological properties play an important role in determining the melt processability of fiber reinforced composites. Processing parameters such as temperature, shear rate, and time affect rheological properties like viscosity, storage and loss modulus, and melt flow index of these composites. In addition, filler type (bio-based source or species), morphology, loading, and filler pre-treatment influence the composite's flow properties and processability. Reprocessing or recycling fiber reinforced composites can alter their melt rheological properties by simultaneously affecting both the polymer matrix and the filler.

Typically, reprocessing causes thermal and mechanical degradation of the matrix, thereby leading to reduced average molecular weight and melt viscosity attributable to chain scission. Additionally, the reduction in fiber length upon reprocessing NFRPCs typically leads to a drop in viscosity (da Costa et al., 2005; Ramzy et al., 2015; Rosenstock Voltz et al., 2020).

Voltz et al. (Rosenstock Voltz et al., 2020) conducted studies on recycling of PP and WPC, consisting of 40 wt% thermomechanical pulp, 60 wt% PP, and a small amount of MAPP. Here, the recycling process was simulated by passing the PP and WPC pellets through a twin-screw extruder up to nine times. The variation of viscosity with shear rate, measured using a capillary rheometer, was reported for the chosen materials before recycling, as well as after 1st, 3rd, 5th, 7th, and 9th recycling steps, as shown in **Fig. 4**C. Both PP and WPC exhibited shear-thinning behavior (i.e., a decrease in viscosity with increasing shear rates), which is typically beneficial for melt processing. For PP, a comparison of unrecycled material (PP-0) and PP after nine recycling cycles (PP-9) indicated recycling to cause a slight decrease in viscosity. This was attributed to the degradation process occurring during reprocessing, leading to a decrease in molecular weight and shear viscosity. On the other hand, for WPCs, a greater degree of viscosity drop was observed with an increase in the number of reprocessing cycles. Although matrix degradation takes place in WPCs similar to the observations in PP, this steep decrease in viscosity was attributed mainly to shortening of the fibers. This led to an increase in the number of fibers with shorter length, which facilitates their alignment in the flow direction in the capillary rheometer (Rosenstock Voltz et al., 2020). A similar trend in reduction in viscosity with increasing the number of reprocessing cycles was observed for sisal/PP and hemp/PP composites, attributed to matrix degradation and fiber shortening from multiple reprocessing cycles (Bourmaud and Baley, 2007). Wood flour composites from recycled PP also showed a similar behavior owing to chain shortening after recycling (Ares et al., 2010).

The melt flow index, defined as the mass of material flowing through a die in ten minutes at a certain temperature, is another rheological property which is often used as a simple and quick method to measure the ease of flow of polymer melts. Recycling typically has been shown to increase the melt flow index of composites. In a study by Bhattacharjee and Bajwa (Bhattacharjee and Bajwa, 2018) on composites containing 30 and 50 wt% wood fiber in HDPE with 3 wt% MAPE, multiple reprocessing cycles (up to six cycles via extrusion) led to an increase in melt flow index for both composites by ~38% and ~250%, respectively. This observation, indicating a decrease in viscosity, was once again attributed mainly to fiber shortening with reprocessing (Bhattacharjee and Bajwa, 2018).

Ramzy et al. (Ramzy et al., 2015) conducted studies on recycling of sisal and hemp fiber (30 wt%) reinforced PP (with coupling agent and stabilizer) using injection molding as the reprocessing method. Flow-ability measurements were performed using a spiral flow length test, a method used to predict a material's mold filling behavior in the injection molding process. Increasing the number of recycling times increased the flowability of these composites, especially for the first few cycles, in which the fiber breakage is typically the highest.

2.7. Weathering resistance

When NFRPCs are used in outdoor applications, their durability becomes an inevitable issue. Outdoor conditions (e.g., microbes, moisture and temperature change, UV exposure) all become detrimental to the long-term service of NFRPCs. Accelerated aging tests have been conducted by a number of researchers to evaluate the durability of NFRPCs, which include freeze-thaw cycling tests, UV weathering tests, or even long-term natural condition tests (Adhikary et al., 2009, 2010; Homkhiew et al., 2014; Moreno et al., 2018). The freeze-thaw cycling test starts by immersing the NFRPCs in water for a certain number of hours, freezing the wet NFRPCs, and finally thawing them at higher

temperatures before evaluation (Adhikary et al., 2010). The UV light weathering test exposes NFRPCs to UV light in a testing chamber at a controlled humidity, temperature, and UV light irradiance (Moreno et al., 2018). Compared with neat plastics, NFRPCs were reported to have lower performance degradation after weathering tests because natural fibers preferentially absorb incident UV radiation, which protects the plastic matrix (Moreno et al., 2018).

Adhikary et al. (Adhikary et al., 2009) investigated the effect of material composition on the aging resistance of NFRPCs made from wood fiber and recycled PP with or without MAPP. In general, UV weathering degraded the performance of NFRPCs (increased hygroscopic behavior and color change and reduced flexural properties). Composites with lower wood fiber content experienced lower flexural strength and modulus decreases, as well as lower water absorption percentage and thickness swelling percentage increase, after the UV light weathering test. The addition of MAPP lowered the degradation in flexural strength and modulus by 50% and 27%, respectively, of weathered NFRPCs compared with composites without MAPP. MAPP also lessened the color change of NFRPCs during the UV weathering test. The effectiveness of MAPP in reducing the weathering effect lies in its ability to improve the interface between wood fiber and recycled PP. Similar results have been reported regarding the effect of MAPP on the hygroscopic, mechanical, and thermal properties of wood fiber/recycled PP or recycled HDPE composites after freeze-thaw cycling tests (Adhikary et al., 2010). In these studies, using NFRPCs from recycled plastics did not seem to result in significantly more performance degradation after weathering when compared with NFRPCs from virgin plastics. Homkhiew et al. (Homkhiew et al., 2014) further studied the use of a UV stabilizer to meliorate the effect of weathering on the performance of rubberwood fiber/recycled PP composites under natural weathering conditions. The addition of 1 wt% UV stabilizer did not seem to improve the performance of rubberwood (45 wt%)/recycled PP composites during a 360-day weathering test.

2.8. Durability under UV exposure

Photodegradation studies show a loss of tensile, flexural, and impact strength of neat thermoplastics and wood plastic composites upon exposure to light (**Table 4**). UV (100–400 nm) in sunlight can cause cracking on exposed surfaces of thermoplastics as well as a lighter appearance of WPC surfaces attributable to bleaching of the wood fiber. Lignin present in wood fibers can act as an antioxidant (Meng et al., 2020) which reduces loss of strength; however, it can undergo bleaching after long UV exposure. Bleaching in wood is caused by a generation of free radicals from the oxidation of lignin to para-quinone chromophore structures, which are further reduced to hydroxyquinones upon extended UV exposure. These reactions can lead to a lightening of wood fiber and yellowing of the polymer matrix. Color analysis can be done using CIE $L*a*b*$ system, where surface color change is defined as $\Delta E = (\Delta L*^2 + \Delta a*^2 + \Delta b*^2)^{1/2}$, in which $\Delta L*$ denotes the change in lightness value and $\Delta a*$ and $\Delta b*$ are changes in chromaticity values. After natural exposure to sunlight and accelerated UV aging, ΔL values are higher in wood and lignin composites than neat thermoplastics such as PP because bleaching of lignin causes fading of the composite (Peng et al., 2014).

Soccalingame et al. carried out artificial UV aging and natural aging of 10 and 30 wt% spruce wood flour-PP (WF10/PP and WF30/PP) injection molded composites containing 1 wt% MAPP and then reprocessed them by injection molding. Artificial aging for 14 days was performed according to ISO 4892–2 standard using xenon arc light of 300–400 nm. These conditions are equivalent to 2–3 months of natural UV-only (300–400 nm) exposure. Natural aging was also carried out for one year by placing panels at 45° facing south. Natural aging showed more severe weathering because surfaces had more cracks and bleaching than artificially aged samples. In both cases, the drop in molecular weight of PP was lower in WF30/PP than WF10/PP, and the intensity of the carbonyl peak in the composite Fourier transform infrared

Table 4

Effect of UV weathering on mechanical property of natural fiber composites. Typically, UV weathering decreased the mechanical performance of natural fiber composites.

Resin	Reinforcing agent (wt%)	Light weathering conditions	Tensile strength and modulus changes	Other property changes	Ref.
PP	Poplar or cellulose or lignin (40)	Total time: 960 h 12 h cycles: 8 h UV + 4 h condensation	–	Flexural modulus (GPa): 2.9→1.6 (poplar) 2.6→0.7 (cellulose) 2.3→1.8 (lignin)	(Peng et al., 2014)
PP	Spruce wood (10 & 30) MAPP (1)	Total time: 14 days 120 min cycles: 102 min UV + 18 min water spraying	–	Impact strength (KJ/m^2): >100 no break → 75 (neat PP) 21→12 (10% wood) 13→10 (30% wood)	(Soccalingame et al., 2015a)
PP	Spruce wood (10 & 30) MAPP (1)	Natural sunlight aging: 1 year	Strength (MPa): 39→16 (neat PP) 39→32 (10% wood) 43→37 (30% wood)	Elongation at yield (%): 7→1 (neat PP) 4→3 (10% wood) 2→2 (30% wood) Impact strength (KJ/m^2): >100→1 (neat PP) 17→2 (10% wood) 9→6 (30% wood)	(Soccalingame et al., 2016)
PP	Bleached and unbleached pine (40) Maleated PP (3)	Total time: 1000 h 1 h irradiation + 1 min water spray + 2 h condensation	Strength (MPa): 25→23 (neat PP) 39→35 (un-bleach) 41→35 (bleach) Modulus (GPa): 1.5→1.5 (neat PP) 4.5→4.0 (un-bleach) 4.5→4.0 (bleach)	Impact strength (KJ/m^2): 10.5→3.3 (neat PP) 5.6→6.4 (un-bleach) 6.1→5.0 (bleach)	(Beg and Pickering, 2008a)
PLA	Untreated and alkaline treated hemp (30)	Weathering: 1000 h 1 h UV + 1 min water spray + 2 h condensation	Strength (MPa): 48→45 (neat) 60→9 (untreated) 82→36 (treated) Modulus (GPa): 4.9→2.7 (neat) 7.6→1.2 (untreated) 11→5 (treated)	–	(Islam et al., 2010)
LDPE	Pine wood waste (30) PE-grafted maleic anhydride (2.5)	Total time: 4032 h 22 h UV + 2 h water vapor condensation	Strength (MPa): 12→2 (neat LDPE) 12→9 (pine)	Elongation at break (%): 575→10 (neat LDPE) 25→10 (pine)	(Moreno et al., 2018)
HDPE	Eucalyptus, acacia or pine (40) Maleated PP (3) Wax (1) PE wax (1)	Weathering: 2000 h 12 h cycles: 8 h UV + 4 h condensation	–	Flexural strength (MPa): 68→66 (eucalyptus) 70→68 (acacia) 70→67 (pine)	(Nguyen et al., 2020)
PP- ethylene vinyl acetate	Eucalyptus or pine (30)	Natural weathering: 270 days	Strength (MPa): 37→12 (neat) 28→13 (eucalyptus) 28→13 (pine) Modulus (GPa): 0.9→0.2 (neat) 0.6→0.4 (eucalyptus) 0.5→0.3 (pine)	Izod unnotched impact strength (J/m): 1290→150 (neat) 220→160 (eucalyptus) 210→150 (pine)	(Catto et al., 2017)

spectroscopy spectra at 1715 cm^{-1} attributable to oxidation was lower in WF30/PP than WF10/PP, indicating that a higher wood content protects WF/PP panels from photodegradation. As discussed previously, after reprocessing the artificially and naturally aged panels through injection molding, the tensile and impact strength was recovered, although the viscosity of the panels was reduced, likely because of chain scission (Soccalingame et al., 2015a, 2016).

In PP and other thermoplastics, the elongation at break during tensile loading decreases upon photodegradation and recovers upon reprocessing (regeneration), following a zig zag pattern upon repeated aging and reprocessing. This pattern is possibly attributable to redistribution of excised chains because only a thin surface layer is degraded by aging. It has also been hypothesized that only the amorphous fraction of the polymer matrix significantly degrades during aging, as diffusion of oxygen in crystalline fraction is very low (Jansson et al., 2003).

Crystallinity has been found to increase in both wood flour filled PP and HDPE upon photodegradation, as the excised shorter chains can crystallize because of higher mobility (Du et al., 2010; Soccalingame et al., 2015a). A comparison of 40 wt% poplar wood flour, cellulose, or alkali lignin in PP concluded that cellulose was better than lignin for color stability, but cellulose-filled PP had severe loss of flexural modulus and developed many microcracks on the surface on UV aging (Peng et al., 2014). In one recent study, *Acacia mangium*-filled HDPE had less surface degradation on accelerated UV aging than *Eucalyptus urophylla* and *Pinus caribaea* wood filled HDPE (Nguyen et al., 2020). Rubberwood fiber/PP composites had higher bleaching on UV exposure when recycled PP was used instead of virgin PP (Homkhiew et al., 2014). Overall, addition of wood fibers to thermoplastic matrices reduces the loss of mechanical strength of the composites upon exposure to light. Finally, the addition of UV absorbers such as hindered amine light stabilizers, carbon black, ferric oxide, titanium dioxide, and nano-silica protects WPCs from bleaching and degradation (Du et al., 2010).

3. Mitigation approaches

3.1. Functional additives

During initial processing of virgin plastic and its first service life,

irreversible mechano-chemical, chemical, or irradiative changes take place in the polymer chain and natural fiber filler (Pfaendner, 2010). In these chemical degradation processes, often induced by radical mechanisms, new chemical groups are often formed in the polymer structure, changing the polymer composition. The concentration of these new structures increases with the service time and the environment of exposure, but can be reduced by including proper additives (Pfaendner, 2010). In any case, these structural changes result in recyclates (ground recycled composites) that are more sensitive to further degradation than the virgin material.

In addition to changes in the composite structure from its first service life, reprocessing NFRPCs can damage and/or degrade additives from these composites. First, heating and drying the composite, as necessary for reprocessing, can embrittle the reinforcing fibers and consume the chemical additives in the plastic prior to recycling (Cestari et al., 2019; Pfaendner, 2010). Additionally, reprocessing can result in further mechanical degradation of the natural fiber fillers and the polymer matrix. Recyclate containing natural fibers without further or pre- treatment, therefore, is usually less mechanically and thermally stable to further processing than virgin material (Hahladakis et al., 2018). This reduction in properties is one of the biggest barriers to more widespread recycling of NFRPCs, along with economic barriers as described above; however, it is often still material- and application-dependent.

Although many studies have uncovered the effects of recycling on the properties of NFRPCs, there is significantly less research in mitigation of those effects. However, over the past decade, there has been growing industrial interest in understanding mitigation approaches, especially for composites containing bio-based fiber fillers, which have seen growing demand in many applications including automotive, construction, and packaging (Cestari et al., 2019; Roy et al., 2019). In this section of the review, the most recent developments to combat property reduction caused by recycling of NFRPCs will be outlined. The first major strategy for combating property degradation of recycled material is the use of functional additives. There are many different types of functional additives utilized to achieve this. They are summarized in Table 5 and will be discussed thoroughly in the following sections.

3.1.1. Compatibilizers and coupling agents

Inherent differences between highly polar lignocellulosic fibers and non-polar polymer matrices, along with the high degree of intermolecular interactions in lignin molecules, can result in poor fiber dispersion and fiber-matrix interactions in NFRPCs (Yang et al., 2019). This often results in aggregation of lignin in bio-based composites due to pi-pi stacking (referring to noncovalent interaction that involves aromatic groups containing pi bonds (Zhuang et al., 2019)), hydrogen bonding, and van der Waals attractions between the polymer chains, which reduces the composites' mechanical properties and dimensional stability (Yang et al., 2019). Overcoming this energetic barrier to achieve compatibility is critical to enhancing the resulting properties of NFRPCs. Research aimed at enhancing the performance of NFRPCs with virgin polymer matrices have largely focused on three areas. The first involves chemical modification of the fibers by washing with NaOH (also known as bleaching), alcohol, or benzene. Washing with NaOH likely falls short of mercerization because mercerization typically involves the treatment of fibers with a high concentration of NaOH (e.g., 10–25%) at 5 °C (Modibbo et al., 2009). A second highly studied topic is surface modification of fibers using stearic acid, maleic anhydride, glycidyl methacrylate, silane, and isocyanate. Finally, polymer modification using anhydrides, epoxides, and even amines has been demonstrated by various researchers (Chand and Fahim, 2008; Pracella et al., 2010; Yang et al., 2019). However, during subsequent recycling and drying processes, the effects of polymer and/or fiber pretreatments are often reduced and sometimes even eliminated (Benoit et al., 2018). As such, there is often a need for additional additives to enhance the compatibility between the phases of the composite.

Currently one of the most common methods to overcome the energetic barrier between polymers and natural fibers in NFRPCs is the use of coupling agents and/or compatibilizers. Table 6 and Table 7 list the mechanical and thermal property changes in NFRPCs (with the addition of coupling agents) after recycling. Compatibilizers and coupling agents are used to promote interfacial adhesion between polymers or phases which are otherwise incompatible. Compatibilizers are usually block copolymers, where each block is compatible with one of the phases and serves to help reduce the surface energy at the interface. These molecules tend to concentrate at the interfaces and stabilize them, allowing finer dispersion and increased compatibility. Coupling agents are chemicals which improve the interfacial properties of fillers and polymers, usually by reacting with the filler surface. They include at least one side group that can either react with the polymer matrix or is at least compatible with it.

Table 5

Functional additives utilized to combat mechanical properties reduction for recycled natural fibers in polymer composites. These functional additives are mainly used to improve the composite compatibility, processing, and performance.

Type of additive	Examples	Function	Ref.
Coupling agents, compatibilizer	Silane MAPP Triazine Isocyanate Paraffin wax Bio-based, grafted co-polymer	Improve compatibility of polymer phase Improve mechanical and physical properties	(Chand and Fahim, 2008; Pfaendner, 2010; Pracella et al., 2010; Xie et al., 2010; Yang et al., 2019)
Processing aids	Salt Wax Stearic acid	Higher melt flow Higher throughput Improved processing Reduced melting temperature	(Pfaendner, 2010; Xu et al., 2019; Xu et al., 2018)
Stabilizers (antioxidants)	UV absorber Phosphite antiacid Phenolic antioxidant Hindered amine stabilizer compound	Improved processing Extended long-term heat stability	(Hamad et al., 2013; Leão et al., 2015; Pfaendner, 2010)
Chain extenders	Epoxide Oxazoline, oxazolone, oxazine Isocyanate, anhydride, acyllactam Maleimide, phosphonite, cyanate, alcohol Carbodiimide and ester preferably with catalyst	Increase molecular weight Increase mechanical properties	(Pfaendner, 2010; Xu et al., 2018)
Other strengthening agents	Talc Clay Wood flour	Flame retardant Strength additives	(Lei et al., 2007; Pfaendner, 2010; Viksne et al., 2004)

Table 6
Mechanical property changes in natural fiber composites (coupling agent added) after recycling. Typically, natural fiber composites with appropriate coupling agent incorporation can be recycled approximately 5–6 times until their mechanical performance decreases.

| Resin	Fiber (loading: wt%)	Coupling agent	Reprocessing methods (number of recycling steps)	Tensile strength and modulus changes	Flexural strength and modulus changes	Other property changes	Ref.
PP	Wood (40)	MAPP	Granulation and injection molding (8)	Strength: −25% Modulus: −16%	Strength: −30% Modulus: −20%	Failure strain: +160% Impact strength: −48%	(Beg and Pickering, 2008b)
	Wood (20)	MAPP	Extrusion and injection molding (7)	Strength: no change Modulus: decrease	–	Failure strain: increase	(Soccalingame et al., 2015a)
	Wood (30)	MAPP	Artificial weathering, extrusion, and injection molding (5)	No change	–	Failure strain or impact strength: increase	(Soccalingame et al., 2015b)
	Wood (30)	MAPP	Natural weathering, extrusion, and injection molding (5)	Strength: −14%	–	Failure strain: no change	(Soccalingame et al., 2016)
	Wood (20)	EBAGMA	Compounding and compression molding (6)	No change	–	Failure strain: increase	(Kaci et al., 2009)
	Wood (40)	MAPP	Extrusion and injection molding (9)	Strength: −20%	–	Failure strain: decrease	(Rosenstock Voltz et al., 2020)
	Wood (60)	MAPP	Pelletization and extrusion (6)	Strength: −30%	Strength: −27%	Impact strength: −33%	(Wang et al., 2019)
	Wood (60)	MAPP + antioxidant		Strength: −33%	Strength: −27%	Impact strength: decrease	
	Hemp (30)	MAPP	Grinding and injection molding (6)	No change	–	Failure strain: +22%	(Bourmaud and Baley, 2007)
	Flax (30)	Not specify	Extrusion and injection molding (5)	Strength: −30% Modulus: −17%	Strength: −15% Modulus: −6%	Failure strain: +98% Impact strength: −58%	(Dickson et al., 2014)
	Flax (21)	MAPP	Grinding and injection molding (7)	Strength: −18% Modulus: no change	–	–	(Gourier et al., 2017)
	Sugarcane bagasse (20)	NaOH	Extrusion and injection molding (4)	–	Strength: −4% Modulus: −4%	–	(Correa-Aguirre et al., 2020)
		NaOH + silane		–	Strength: +4% Modulus: +14%	–	
	Coriander straw (40)	MAPP	Extrusion and injection molding (5)	Modulus: no net change	Modulus: no net change	Impact strength: +90%	(Uitterhaegen et al., 2018)
	Alfa (30)	Salt + NaOH	Grinding and injection molding (4)	Decrease	Strength: −7% Modulus: −11%	–	(Ezzahra El Abbassi et al., 2019)
	Rice hull (30)	MAPP	Grinding and compression molding (2)	–	Strength: −10% Modulus: no change	–	(Srebrenkoska et al., 2008)
	Kenaf (30)	MAPP		–	Strength: −5% Modulus: +20%	–	
	MDFB (40)	MAPP	Extrusion and injection molding (5)	Strength: −21% Modulus: −20%	Strength: −14% Modulus: −14%	Failure strain: +132%	(Dickson et al., 2014)
PLA	Wood (50)	MA	Extrusion and injection molding (5)	Decrease	Decrease	Impact strength: decrease Failure strain: increase	(Bhattacharjee and Bajwa, 2017)
		MA	Extrusion and injection molding (6)	Strength: −42% Modulus: decrease	Strength: −70% Modulus: decrease	Impact strength: −40%	(Bajwa and Bhattacharjee, 2017)
LDPE	Coriander straw (40)	MAPE	Extrusion and injection molding (5)	Strength: no net change	Strength: no net change	Impact strength: +200%	(Uitterhaegen et al., 2018)
HDPE	Wood (50)	MAPE	Extrusion and injection molding (6)	Strength: −10% Modulus: decrease	Strength: −15% Modulus: −24%	Failure strain: increase	(Bhattacharjee and Bajwa, 2018)
	Wood (65)	MAPE	Extrusion and compression molding (4)	Modulus: no net change	–	–	(Nadali et al., 2018)
	Wood (50)	MA	Extrusion and injection molding (6)	Decrease	Strength: −10% Modulus: decrease	Impact strength: −24%	(Bajwa and Bhattacharjee, 2017)
	Cellulose (48)	MAPP E-43	Grinding and injection molding (5)	Strength: −37% Modulus: −30%	–	Failure strain: −22%	(Fonseca-Valero et al., 2015)
		MAPP G-3002	Grinding and injection molding (5)	Strength: −46% Modulus: −53%	–	Failure strain: −9%	

Figs. 5 and 6 exhibit the mechanical property changes in natural fiber composites after recycling: without and with coupling agent, with different types of coupling agents/polymers/fibers/reprocessing methods. The addition of MAPP or MAPP E-43 (Epolene E-43, which exhibits an acid number of 45 and a number average molecular weight of 3900, available from Eastman Chemical Products, Tennessee, USA) (Fonseca-Valero et al., 2015) is helpful to improve the failure strain of the recycled NFRPCs. In addition, MAPP E-43 can promote the tensile strength and tensile modulus of the recycled NFRPCs. The addition of NaOH is helpful to improve the flexural strength (by 10%) and flexural

modulus (by 15%) of the recycled NFRPCs, which can be further improved via further addition of silane. Different types of coupling agents including MAPP, MAPP + antioxidant, NaOH, NaOH + silane, MAPE, and MA have been used for natural fiber composites. Results indicate that the combination of NaOH and silane is a promising coupling agent. Polymer matrix type and reprocessing method have a significant effect on the mechanical performance of natural fiber composites after recycling. HDPE is a promising polymer that is amenable to multiple reprocessing cycles. Different types of natural fibers have been studied for the recycling of natural fiber composites, finding that

Table 7
Thermal property changes in natural fiber composites (coupling agent added) after recycling. Typically, natural fiber composites with appropriate coupling agent incorporation can be recycled approximately 5–6 times until their thermal property changes.

Resin	Fiber (loading: wt%)	Coupling agent	Reprocessing methods (number of recycling steps)	T_m change	T_d change	T_c change	Crystallinity change	Other property changes	Ref.
PP	Wood (40)	MAPP	Granulation and injection molding (8)	Decrease	–	–	+3%	–	(Beg and Pickering, 2008b)
	Wood (20)	MAPP	Extrusion and injection molding (7)	–	–	–	No change	–	(Soccalingame et al., 2015a)
	Wood (30)	MAPP	Artificial weathering, extrusion, and injection molding (5)	–	–	–	No change	–	(Soccalingame et al., 2015b)
	Wood (40)	MAPP	Extrusion and injection molding (9)	Decrease	Decrease	–	–	–	(Rosenstock Voltz et al., 2020)
	Wood (60)	MAPP	Pelletization and extrusion (6)	–	–	–	+3%	T_g: decrease	(Wang et al., 2019)
	Wood (60)	MAPP + antioxidant		–	–	–	+8%	T_g: decrease	
	Hemp (30)	MAPP	Grinding and injection molding (6)	–	–	No change	No change	–	(Bourmaud and Baley, 2007)
	Flax (21)	MAPP	Grinding and injection molding (7)	No change	–	–	–	T_g: no change	(Gourier et al., 2017)
	Sugarcane bagasse (20)	NaOH	Extrusion and injection molding (4)	No change	Decrease	No change	Increase	–	(Correa-Aguirre et al., 2020)
		NaOH + silane		Decrease	Decrease	Increase	Decrease	–	
	Rice hull (30)	MAPP	Grinding and compression molding (2)	–	Decrease	–	–	–	(Srebrenkoska et al., 2008)
	Kenaf (30)	MAPP		–	Increase	–	–	–	
PLA	Wood (50)	MA	Extrusion and injection molding (5)	–	Increase	–	Decrease	CTE: increase	(Bhattacharjee and Bajwa, 2017)
		MA	Extrusion and injection molding (6)	–	–	–	–	CTE: −41%	(Bajwa and Bhattacharjee, 2017)
HDPE	Wood (50)	MAPE	Extrusion and injection molding (6)	Increase	Increase	Decrease	Decrease	HDT: decrease	(Bhattacharjee and Bajwa, 2018)
		MA	Extrusion and injection molding (6)	–	–	–	–	CTE: +50% HDT: decrease	(Bajwa and Bhattacharjee, 2017)
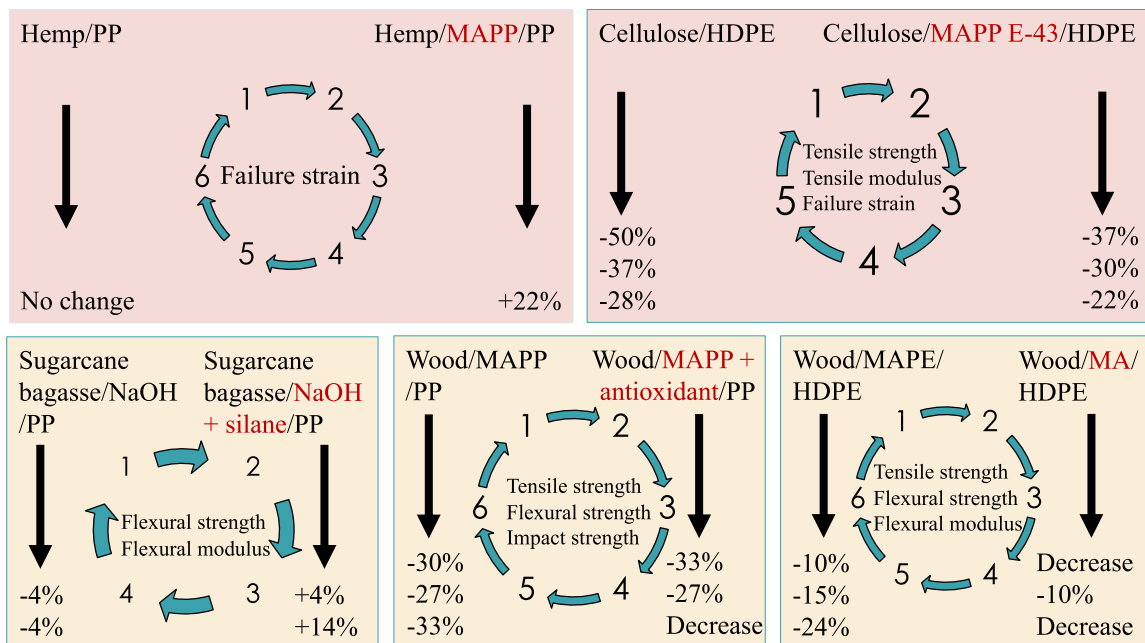
Fig. 5. Mechanical property changes in natural fiber composites after recycling (the circle symbol means recycling and the number means recycling step): without coupling agent, with coupling agent, and with different types of coupling agents (data derived from (Bajwa and Bhattacharjee, 2017; Bhattacharjee and Bajwa, 2018; Bourmaud and Baley, 2007; Correa-Aguirre et al., 2020; Fonseca-Valero et al., 2015; Wang et al., 2019)). The addition of coupling agents (e.g., MAPP, NaOH, and silane) can promote the mechanical property of the recycled NFRPCs. The combination of NaOH and silane is a promising coupling agent for maintaining the mechanical performance of the natural fiber composite after recycling.
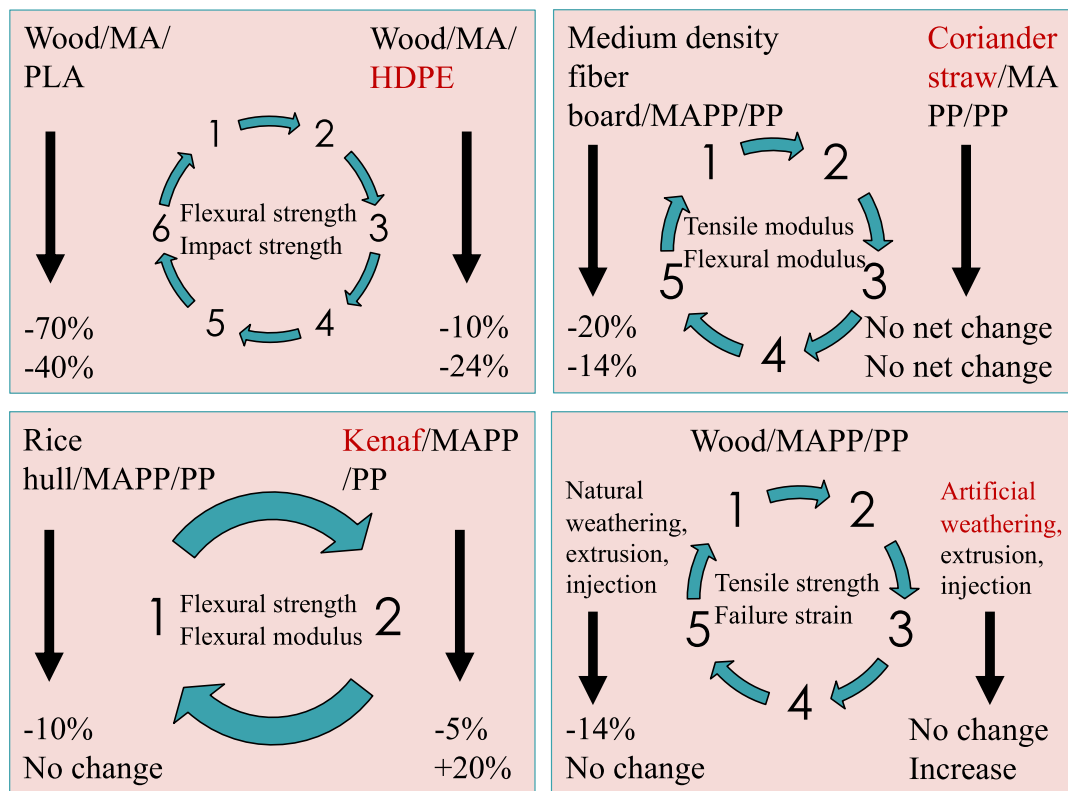
Fig. 6. Mechanical property changes in natural fiber composites after recycling: with different types of polymers, fibers, and reprocessing methods (data derived from (Bajwa and Bhattacharjee, 2017; Beg and Pickering, 2008b; Bhattacharjee and Bajwa, 2017; Dickson et al., 2014; Rosenstock Voltz et al., 2020; Soccalingame et al., 2016, 2015b; Srebrenkoska et al., 2008; Uitterhaegen et al., 2018)). The types of polymer matrix, fiber, and reprocessing method have a significant effect on the mechanical performance of natural fiber composites after recycling. Coriander straw is a promising fiber for maintaining the mechanical performance of the natural fiber composite after recycling.

coriander straw is a promising fiber. In summary, the addition of a coupling agent can improve the compatibility between the polymer matrix and fiber, which can promote the mechanical properties of the recycled NFRPCs.

Many researchers have studied the use of MAPP as a coupling agent for recycled polymer systems within the past decade, and it shows continued promise as a coupling agent for natural fiber-reinforced PP (Zheng et al., 2017). More recently, researchers have studied the use of acrylic acid-grafted PP coupling agents in recycled PP filled with Kenaf fiber (Suharty et al., 2016). The addition of acrylic acid-grafted PP coupling agent increased the tensile strength of the composites by 18%, flexure strength by 28% and the impact toughness by 27%. Other researchers have studied the use of MAPE, carboxylated PE and a titanium-derived mixture as coupling agents for recycled HDPE and bagasse fiber composites (Lei et al., 2007). A titanium-derived mixture, obtained from Kenrich Petrochemicals Inc., is an organometallic inter-face chemical that contains titanium. Two examples of titanium-derived coupling agents are titanium IV 2-propanolato, tris(dioctyl) phosphato-O, and titanium IV 2,2(bis 2-propenolatomethyl)butanolato, tris neodecanoato-O (Elshereksi et al., 2017; Lei et al., 2007). Results from the study showed that MAPE, carboxylated PE, and titanium-derived mixture improved the compatibility between the fiber and the recycled HDPE. As more MAPE (until reaching an optimum level) was added to the composites, the modulus, tensile strength, and impact strength increased in correlation, largely attributed to enhanced compatibility. Carboxylated PE had no significant influence on the mechanical properties of the composites except the tensile strength, largely attributed to the decrease in resulting compatibility compared with MAPE-containing composites. Finally, titanium-derived mixture improved the mechanical properties of the composite slightly, but

largely served as a lubrication agent rather than a coupling agent and resulted in voids in the composite system at higher loading levels. This study highlighted the importance of considering the makeup of both the polymer matrix and reinforcing agent when selecting a coupling agent for a composite.

Bourmaud (Bourmaud and Baley, 2007) and Wang (Wang et al., 2019) studied PP with hemp fibers and wood fibers, respectively, with the use of MAPP as a compatibilizer. The use of a compatibilizer enhanced the tensile performance of the composites due to increased stress transfer between the polymer and well-adhered fibers. However, after recycling, the compatibilizers were seen to degrade quickly, and the tensile performance of compatibilized and non-compatibilized composites were similar after multiple reprocessing cycles. The hemp/PP composite displayed a slight loss in its tensile modulus and a notable decrease in the failure strain, which was attributed to fiber breakup and poor adhesion between the fibers and polymer matrix upon recycling, while a decrease in the tensile strength of the wood fiber/PP samples was ascribed to wood fiber damage endured during reprocessing.

More recently, researchers have expressed concerns with the use of synthetic compatibilizers and coupling agents in green materials and over the past several years there is a growing wealth of research designing bio-based coupling agents. For example, researchers studied the use of ionic liquid-treated lignin as a bio-based coupling agent in recycled 60 wt% bagasse fiber-reinforced PP (Younesi-Kordkheili and Pizzi, 2020). The cost of the ionic liquids is still high, but it will decrease with continuous research and development on the new ionic liquids and scale-up. One example of development is the commercialization of the ionic liquid-based catalyst. In 2021, Chevron and Honeywell announced the start-up of world's first commercial ionic liquid alkylation unit

(Chevron and Honeywell, 2021). Even though this unit is not used for biomass process, the commercialization of ionic liquid for biomass processing will become realistic. More importantly, most of the ionic liquids used in the biomass processing can be recycled (Wang et al., 2012), which can further lower the cost of ionic liquids. The ionic liquid-treated lignin's functionality was compared side-by-side with MAPP. Generally, the composites with modified lignin exhibited a higher flexural and tensile strength than those with unmodified lignin. The addition of lignin up to 5 wt% dramatically enhanced both tensile and flexural strengths (47% and 59% increase, respectively) of the composites based on recycled PP. Another group looked at the use of starch gum as a coupling agent for wood fiber/recycled PP composites in comparison to MAPP controls (Rocha and Rosa, 2019). The micrographs of the composite samples from virgin polymer and recycled polymer are shown in Fig. 7. Virgin composites showed a limited improvement on the elastic modulus upon the addition of starch gum in comparison to those with MAPP. A 3D construction and proposed mechanism of MAPP compatibilization in virgin composites are shown in Fig. 8. After recycling of both composites, however, the samples containing starch gum presented ~26% higher values for the elastic modulus than those with MAPP, indicating starch gum showed an enhanced interaction with oxidized structures compared with MAPP, resulting in decreased size and number of voids in the composite – although voids were still present - and improved compatibility between composite phases.

3.1.2. Processing aids

Natural fibers begin to thermally degrade at temperatures between 200 and 250 °C, at which the thermal degradation of hemicellulose, cellulose, lignin, and pectin begin to occur. Cellulosics are reported to start to degrade at approximately 180–220 °C (Peng et al., 2013). The extent of thermal degradation of natural fibers is presumably a function of time at a certain temperature (Khanlou et al., 2017). PLA suffers from thermal degradation during the compression molding process (Khanlou et al., 2017). Time and temperature are major process parameters for controlling thermal degradation in both natural fibers and polymer matrices (Khanlou et al., 2017). For example, the optimum mechanical properties of flax/PLA composites can be achieved at the shortest molding time (5 min) (Khanlou et al., 2017). Approximately 6%–10% of natural fiber damage (e.g., mass loss) is acceptable at certain processing temperatures (El-Sabbagh et al., 2014; Khanlou et al., 2017). In one

study, the mass loss of flax fibers at isothermal conditions for 15 min at different temperatures was as following: 0.3% (170 °C), 1% (180 °C), 2% (200 °C), 3% (220 °C), 4% (230 °C), 6% (240 °C), and 11% (260 °C) (El-Sabbagh et al., 2014). Many thermoplastic matrices used in NFRPCs have melting and processing temperatures in excess of 250 °C, and therefore processing at these temperatures or above result in irreversible degradation of the fibers (Pfaendner, 2010). The resulting structural changes to the fibers from thermal degradation make them more brittle and cause a decrease in the overall modulus of the NFRPC systems. Additionally, adding too much fiber can make the composite too stiff for processing. The use of plasticizers can help soften the system, enabling higher loading levels of filler and ideally better properties of recycled polymer.

More recently, researchers have demonstrated that using plasticizers or modifiers can reduce the melting point of plastics, resulting in a reduction of the processing temperatures required during recycling, and thus a reduction in fiber degradation. Researchers have shown the use of salts, such as LiCl, can lower the melting temperature of polyamide 6 and polyamide 6-HDPE blends (Xu et al., 2019; Xu et al., 2018). Differential scanning calorimetry analysis indicated the use of 3 wt% LiCl lowered the melting temperature of polyamide 6 from 220 to 191 °C. Hydrogen bonding between the salt and the nylon chains encouraged wrapping, thereby altering the crystallization behavior of the polymer chains. However, the addition of LiCl caused a brown color change of the solution that increased with an increase in processing temperature (Xu et al., 2018). Similarly, researchers observed a reduction in melting temperature (from 212 to 194 °C) and change in solution color after a 3 wt% LiCl addition as a modifier for PA/HDPE blends. Moreover, the addition of LiCl increased the impact strength of the solution by 36% and assisted with interfacial adhesion of the composite (Xu et al., 2019). Other researchers studied the effects of addition of natural rubber (up to 7 wt%) as a plasticizer in kenaf fiber/recycled PP composites (Suharty et al., 2016). The natural rubber was used to soften the polymer and resulted in higher filler loading and an increase flame resistance. The researchers found that a 3 wt% addition of the recycled natural rubber resulted in the best mechanical properties.

3.1.3. Thermal stabilizers

The use of antioxidants as thermal stabilizers in plastics dates back to the 1960s, where their use was predominantly to stabilize plastics, such
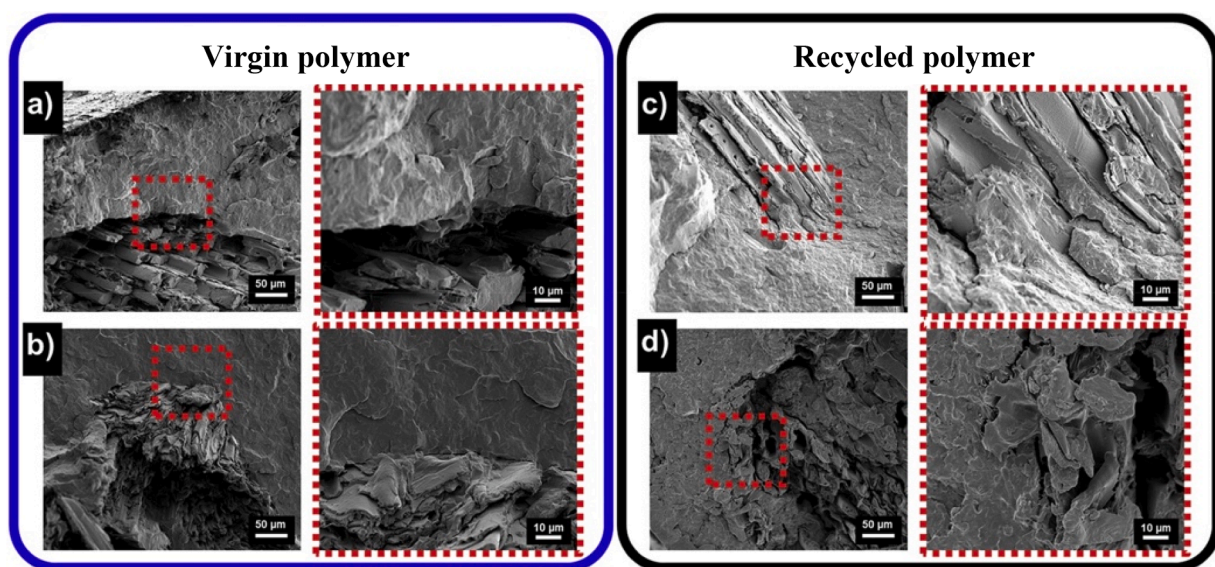

Fig. 7. Scanning electron microscopy images of the composite samples from virgin polymer and recycled polymer: (a) 5% starch gum/virgin polymer, (b) MAPP/virgin polymer (interface was compatible with good interaction), (c) 5% starch gum/recycled polymer (interface was compatible with good interaction), and (d) MAPP/recycled polymer. MAPP shows good miscibility with virgin polymer, but starch gum shows good miscibility with recycled polymer (reproduced with permission from Rocha and Rosa, 2019). Copyright © 2019 Elsevier Ltd.
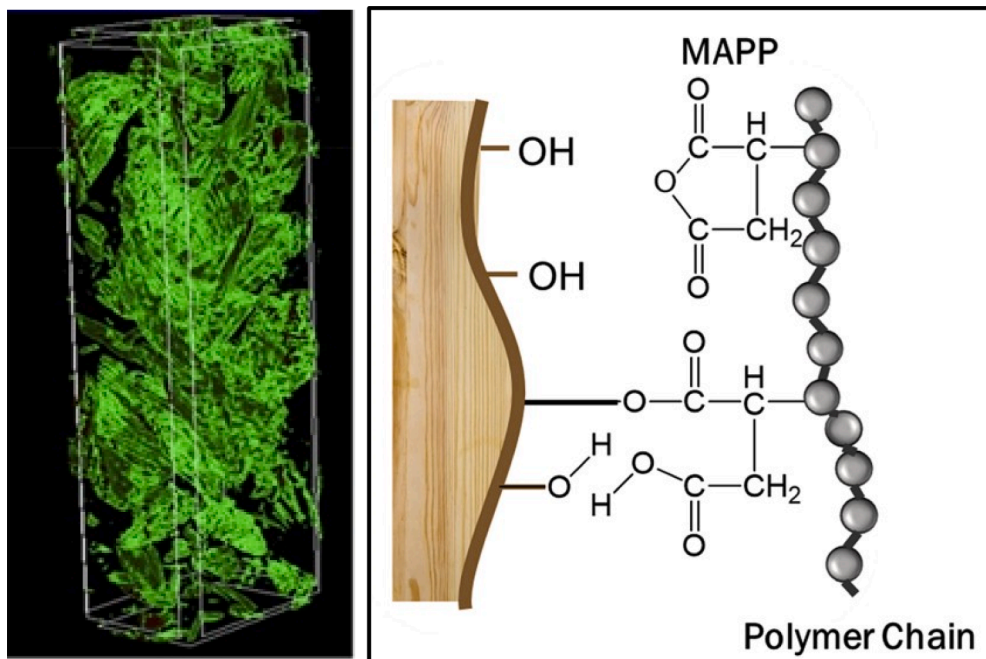
Fig. 8. The 3D construction (left) with matrix suppressed and a highlight of the reinforcement and (right) proposed mechanism of MAPP/virgin polymer composites. There is a proper distribution of fibers in the composite, but no signal of orientation. MAPP has good miscibility with the matrix due to its chain of PP, and promotes a chemical bonding with fiber surface due to its maleic anhydride (reproduced with permission from Rocha and Rosa, 2019). Copyright © 2019 Elsevier Ltd.

as polyolefins, and other organic materials, such as hydrocarbon fuels and rubbers, against degradation caused by oxygen or ozone to enhance shelf-life (Meltsner, 1971). Stabilizers protect recyclates against both oxidative and photooxidative damage and as a result, preserve some properties both during processing and product life cycle. However, researchers have been using thermal stabilizers to add more environmental protection for virgin engineering thermoplastics such as polyamides, polyolefins, and polyesters (Leão et al., 2015). Their use in recycled NFRPCs, however, is limited. Any stabilizer residues from the composite's first life will retain some protective function after recycling, but their efficacy is often limited. Although plastics originally used in single use applications, such as in packaging, typically contain small amounts of stabilizers and require further stabilizer addition during recycling. Overall, stabilization of most recyclates is typically achieved through phenolic antioxidants, phosphites, and antiacids, hindered amine stabilizers, or UV absorbers (Pfaendner, 2010).

In UV aging (315–400 nm) of 30 wt% pine wood waste-LDPE waste composites containing 2.5 wt% maleic anhydride grafted PE for 48–4320 h, considerable changes to surface and bulk properties occurred in neat LDPE after 720 h, whereas they occurred in pine wood filled samples only after 4320 h. The tensile strength in unfilled LDPE dropped from 12 to 2 MPa on 4032 h of UV exposure, whereas the 30% pine wood filled LDPE composites only changed from 12 to 9 MPa (Moreno et al., 2018). Wang et al. (Wang et al., 2019) used a commercial antioxidant in their study with wood fiber/PP composites. After seven cycles of extrusion and pelletization, the flexural strength was found to decrease less than in composites processed without the antioxidant.

Researchers have shown that recycled, melt mixed PP composites, reinforced with natural fibers treated using various methods, exhibited decreases in tensile strength relative to the original composites. However, when thermal stabilizers were added, the mechanical properties were maintained or increased (Hamad et al., 2013). Other researchers studied bagasse/PP composites containing mixed metal salt blends and phosphorus-based thermal stabilizer additives. With the addition of either thermal stabilizer, there was a maintained or increased tensile strength of the composite compared with the original composite and the recycled, un-stabilized composite (Leão et al., 2015). More recently,

researchers showed that low concentrations (<0.2 wt%) of thermal stabilizers can yield more recyclable polyolefins, like HDPE (Knauer, 2019). The low concentration of stabilizers helped preserve the molecular weight of polymers for up to five passes, retain color through recycling passes, and maintain mechanical properties.

3.1.4. Chain extenders

Chain extenders are low molecular weight molecules that react with the ends of polymer chains and increase their overall molecular weight. Thermal and mechanical degradation due to re-processing and/or drying of recyclate can reduce the molecular weight of polymer chains. As the molecular weight of a polymer has a significant effect on the strength of the material, degradation during recycling can affect the material properties. Using chain extenders during recycling can reduce and even eliminate the impacts of recycling on the material properties. Some research has even shown that chain extenders can increase the mechanical properties of the recyclate to even higher than that of virgin composites (Pfaendner, 2010). A number of chemical compounds have been suggested such as epoxides, oxazolines, oxazolones, oxazines, isocyanates, anhydrides, acyllactams, maleimides, phosphonites, cyanates, alcohols, carbodiimides, and esters, preferably in combination with a catalyst (Pfaendner, 2010). More recently, researchers have explored 2,2′-(1,4-phenylene)bis(2-oxazoline) as a chain extender in conjunction with a LiCl thermal stabilizer (Xu et al., 2018). The addition of the chain extender significantly increased the molecular weight of the polymer chains in the system, resulting in enhanced mechanical properties.

3.1.5. Other methods

The alfa fibers in Abbassi's work (Ezzahra El Abbassi et al., 2019) were treated with either saltwater alone or saltwater and NaOH. The alkaline treatment improved the mechanical performance of virgin composites in comparison to that of saltwater-treated or untreated fiber composites. This treatment seemed to have a lasting effect, as well, because the flexural strength of the saltwater-treated fiber composites dropped 15% after five cycles of extrusion and injection molding, whereas the flexural strength of saltwater and alkali-treated fibers

dropped only 7% after the same recycling processes.

Inspired by others who used clays as reinforcement and flame retardant agents for plastic composites, researchers have added montmorillonite, a type of clay, to recycled PP composites reinforced with natural fibers to enhance their flame retardance (Suharty et al., 2016). The results indicated that the montmorillonite additive enhanced the flame retardancy by undergoing an endothermic reaction which can absorb the heat produced during the combustion process, ultimately improving the mechanical properties of the recycled material. Other groups have explored the addition of wood dusts or flours as strengthening additives in addition to natural fibers. One such group looked at the addition of sawdust treated with paraffin for mechanical strengthening of wood fiber reinforced recycled LDPE (Viksne et al., 2004). The researchers noted an increase of Young's modulus and decrease of tensile strain with an increase of modified sawdust content in the blend.

3.2. Addition of virgin plastic

As has been discussed, a loss of mechanical properties is generally observed after several reprocessing steps attributable to polymer chain scission and thermal degradation of fillers. However, the dispersion of the fillers is improved because of the extensive mixing. Addition of virgin plastics into the reprocessed composites can compensate for the loss of mechanical property in reprocessed NFRPCs (Åkesson et al., 2016b; Beg and Pickering, 2008b; Petchwattana et al., 2012; Viksne and Rence, 2008). Petchwattana et al. (Petchwattana et al., 2012) investigated the effect of the addition of virgin plastic on the mechanical properties of the recycled wood fiber/PVC composites, as the recycled material did not pass an imposed quality control. As shown in the Fig. 9A, different mixing ratios of recycled and virgin WPC were investigated using profile extrusion. Compared with the virgin composite (fresh-feed), they found the composites made from WPC scrap: fresh-feed at 30:70 kept the flexural strength and improved the flexural modulus. After considering other factors (e.g., impact energy and material cost), the ratio of 30:70 was also found to be the most appropriate mixing ratio. Beg and Pickering (Beg and Pickering, 2008b) studied the reprocessing of wood fiber/PP composites using a twin-screw extruder. They mixed the reprocessed composites with virgin composite at a ratio of 50:50 and found the tensile strength matched the virgin composite.

In addition to petroleum-based polymer matrices, recycled biobased composites have been studied to mitigate the mechanical properties loss

due to processing. Compared with PP and PVC, biobased polymers (e.g., PLA) are more sensitive to reprocessing and tend to degrade at high temperatures. Akesson et al. (Åkesson et al., 2016b) investigated the mechanical recycling of PLA filled with a high level of cellulose fibers (70 vol%), and found these composites are not suitable for multiple processing (compression molding) due to the high fiber loading. Instead, they processed the ground recycled composites (recyclate) with virgin PLA through compression molding. As shown in Fig. 9B, the recyclate had a relatively good reinforcing effect on PLA. The tensile strength and Young's modulus increased by 54% and 136% (compared with neat PLA), respectively, at a mixing ratio of 50:50, attributable to the fiber reinforcement from the recyclate. However, the authors did not compare the properties of the new composite made from virgin PLA and composite recyclate with composite itself prior to recycling, and it is hard to conclude the effect of adding virgin PLA to the mechanical properties.

Similar to adding virgin plastic to the reprocessed NFRPCs to mitigate the property loss, adding extra fillers can have a similar effect. During multiple reprocessing cycles, the aspect ratio of the fillers in a NFRPC is typically reduced significantly (Bourmaud and Baley, 2009). Addition of new, high aspect ratio fibers can reinforce reprocessed NFRPCs and achieve better properties. However, no literature information has been found exploring this option so far.

4. Emerging applications

The global market for biocomposites is expected to reach $41 billion net worth by 2025 (Zwawi, 2021). NFRPCs have been widely used and have potential for a variety of applications such as 3D printing (Bhagia et al., 2021), automotive (Keskisaari and Kärki, 2018), tribological, packaging, and biomedical applications. For example, Li et al. (Li et al., 2019) investigated the cellulose nanocrystal reinforced poly(ethylene glycol) diacrylate composites for 3D printing use. Cellulose nanocrystal has high strength, high Young's modulus, renewability, and biocompatibility (Li et al., 2019; Wang, L. et al., 2020). The authors (Li et al., 2019) used the cellulose nanocrystal composites to 3D print a disk structure and an octet-truss lattice structure, shown in Fig. 10. Conventional composite processing technologies have limitations in efficiently fabricating composites with different and complex shapes. However, 3D printing can easily fabricate different and complex parts via computer aided design (Li et al., 2019; Zhao et al., 2020). Jiang and Raney (Jiang and Raney, 2019) studied the 3D printing of cotton fiber
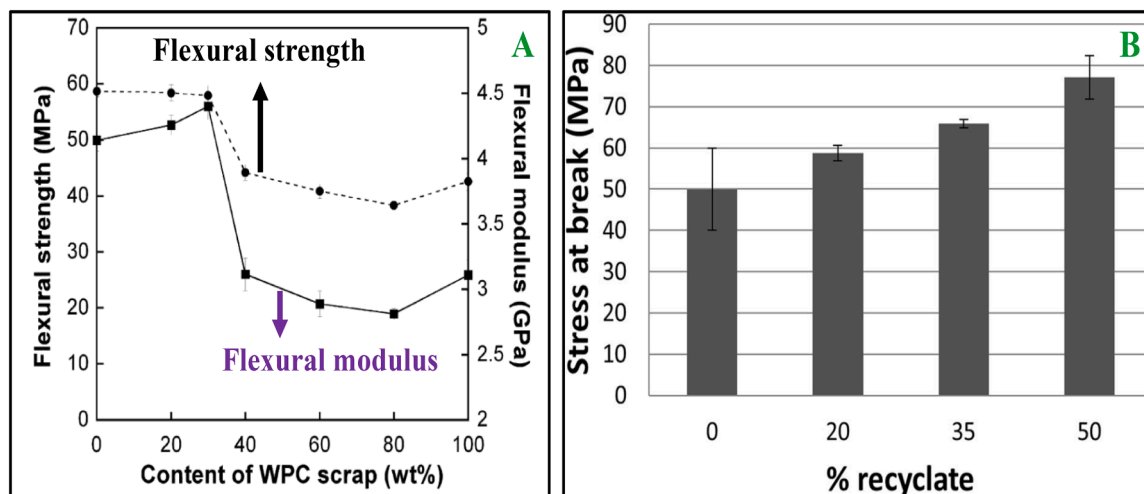

Fig. 9. (A) Flexural strength and flexural modulus of the WPC profiles extruded from WPC fresh-feed from various mixtures of WPC scrap: fresh-feed at 20:80, 30:70, 40:60, 60:40, 80:20 and 100:0. The composites made from WPC scrap: fresh-feed at 30:70 (optimum mixing ratio) kept the flexural strength and improved the flexural modulus (reproduced with permission from Petchwattana et al., 2012). Copyright © 2012 Elsevier Ltd. (B) Stress at break for composites with different loading of the recyclate, suggesting the recyclate had a relatively good reinforcing effect on PLA (reproduced with permission from Åkesson et al., 2016b). Copyright © 2016 Springer Nature.
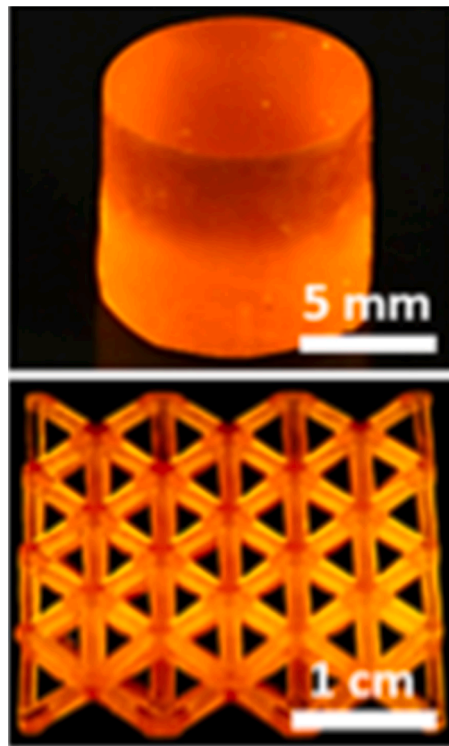
Fig. 10. The 3D-printed disk structure (top) and the octet-truss lattice structure (bottom) using cellulose nanocrystal composites (reproduced with permission from Li et al., 2019). Copyright © 2019 Springer Nature.

reinforced amylopectin composites to produce mortars in building. Amylopectin, a primary component of sticky rice, is a branched polymer with a high molecular weight. The cotton fiber reinforced amylopectin composites were more resilient than common thermoplastic composites (wood fiber/PLA composites) toward heat, flame, and ultraviolet light (Jiang and Raney, 2019). In a previous study (Zhao et al., 2019), poplar fiber was used to reinforce PLA for large-scale 3D printing application. A podium base, shown in Fig. 11, was successfully 3D printed using the poplar fiber reinforced PLA composites (Zhao et al., 2019).
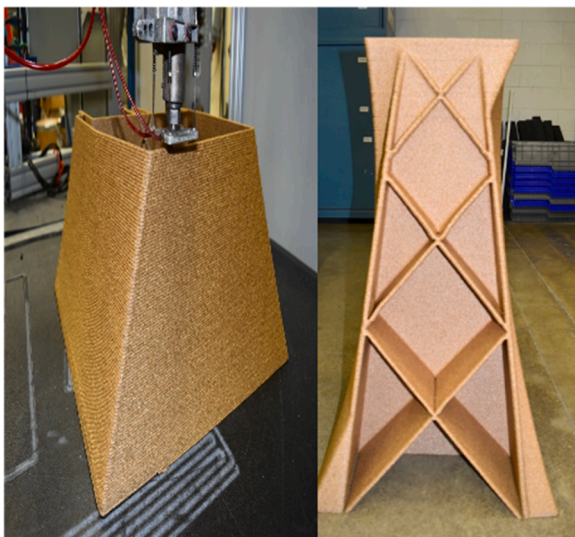
Fig. 11. Large-scale 3D printing of poplar/PLA composites. (Left) 3D printing process and (right) back of the 3D-printed podium base) (reproduced with permission from Zhao et al., 2019). Copyright © 2019 American Chemical Society.

Furthermore, Sullins et al. (Sullins et al., 2017) investigated hemp fiber reinforced PP composites. They stated that NFRPCs have been used in automotive applications because of their good modulus and strength, recyclability, and low carbon footprint (Sullins et al., 2017). Naeem et al. (Naeem et al., 2020) studied banana peel fiber reinforced epoxy composites, which have potential for applications in automotive seat backs, floors, bolsters, and other non-loaded-bearing applications. Soccalingame et al. (Soccalingame et al., 2015b) investigated the spruce wood flour reinforced PP composites, which are commonly used in outdoor decking applications. Batu et al. (Batu et al., 2020) studied natural fiber (ramie, curaua, or pine apple) reinforced polymer (epoxy) composites. The composites can be used for wind turbine blade applications (Batu et al., 2020).

Unfortunately, very little literature specifically discusses the use of recycled NFRPCs for these applications. There are many challenges that need to be addressed for NFRPCs to be more widely used, recycled, and for the recyclate to be utilized commercially. Lau et al. (Lau et al., 2018) reported that the high moisture content, flammability, property inconsistency, and swelling effect of natural fibers, as well as poor bonding properties between fiber and polymer (hydrophilic fiber and hydrophobic polymer), and uneven dispersion of natural fibers in the products are the major challenges. Researchers have worked to recycle NFRPC products from applications such as packaging, but these recycled composites have not been retested in products, only characterized for thermomechanical properties (Fazita et al., 2015). As discussed throughout this review, recycled NFRPCs suffer from variations in resultant properties, especially after repeated recycling. These differences can limit their applications. For instance, recycled fibers often display higher levels of crystallinity, all of which results in property variation, making it difficult to use commercially. However, researchers have shown that additional materials, such as stabilizers, could help mitigate property loss and increase use of recycled materials in these applications (Leão et al., 2015). Overall, further research is needed to develop commercially viable recycling methods that can produce useable materials for these applications.

5. Challenges and future directions

There is a research gap regarding the safety, durability, and especially recyclability of NFRPCs. Safety hazards might exist when recycling composites that contain formaldehyde-based adhesives that might give off volatile organic compounds. Durability will be an issue if the composites are used in exterior applications where they are not protected from weather, biological attack, and so on. Natural fiber composites, by their nature, are recyclable. The major technical challenges for recycling and application of NFRPCs that need to be addressed include polymer and fiber degradation, high moisture content, flammability, variation in the natural fiber composition, and poor bonding between hydrophilic fibers and hydrophobic polymers. Understanding how to improve the properties of NFRPCs toward targeted applications before and after recycling is crucial for their increased use.

(1) Fundamental research on the fiber-matrix interface to improve the interfacial adhesion and mechanical properties of NFRPCs is needed. Some physicochemical methods, such as mechanical hotpressing and using acids or epoxies as modifiers, have been developed to improve the properties of the composites (Zhao et al., 2020a). A novel class of natural fiber composites is surging, which comprises the incorporation of graphene-based materials to act as reinforcement or to improve the interfacial adhesion between hydrophilic natural fibers and hydrophobic polymer matrices (da Luz et al., 2020). Graphene-based materials have been shown to significantly improve several properties (e.g., toughness and stiffness) of NFRPCs (da Luz et al., 2020). However, the mechanisms behind this enhancement and fabrication

optimization to further improve their properties have not been thoroughly investigated.

(2) Durability, sustainability, and cost reduction need to be further explored for wider applications of NFRPCs and their recyclates. Long-term durability and recyclability of composites can be reached via emerging mitigation approaches during recycling to obtain high performance. Sommerhuber et al. (Sommerhuber et al., 2017) investigated the life cycle assessment of wood plastic composites. Wood is essential for sustainable economic growth, but thermoplastic derived from fossil resources can potentially cause severe environmental impacts along the life cycle of composites. After the end of life, wood plastic composites have mainly been treated as waste to be incinerated, leading to value loss and environmental issues. Recycling of wood plastic composites would be an ecologically preferable pathway. Wood plastic composites produced from recycled plastics are environmentally better than those produced from virgin plastics. Wood plastic composites produced from recycled composite materials are a technically and ecologically superior alternative. However, the recycled composite materials may need further separation and particle processing, which contributes to the overall environmental impacts. For example, adding additives is a potential method for improving the durability of composites, but additives can cause concerns for human health and the environment along the life cycle (Sommerhuber et al., 2017). Additionally, the sustainability of such composites can be improved by developing new high-performance biobased polymers derived from biomass. Finally, a low cost can be maintained by developing cost-effective processes and reusing waste fiber, polymer, and composites.

(3) Further research into the effects of processing parameters on recycled NFRPCs' properties would be quite beneficial to the field. Processing technologies including compounding, mixing, extrusion, injection molding, and compression molding are generally used to prepare and recycle NFRPCs, and these parameters can significantly influence the properties of all polymer composites (Faruk et al., 2014). Additionally, the parameters available to alter can vary for different processing technologies. For example, during twin-screw extrusion, one can alter the screw configuration, screw speed, extrusion temperature, and flow rate. However, there have been no detailed studies on the effects or optimization of these parameters on the reprocessing of the NFRPCs.

(4) Wide applications of thermoset composites indicate that a market exists for the recycling of used thermoset composites. However, little literature specifically discusses the recycling of natural fiber reinforced thermoset composites. Dinu et al. (Dinu et al., 2021) prepared composites from humins based thermoset resin along with chicken feathers and vegetal non-woven fibers (linseed flax and jute). Mechanical recycling of the thermoset composites was performed. Humins based resin becomes flexible because of structural and chemical rearrangements upon exposure to heat. This resin can be reprocessed into the desired shape under heat and pressure, making it acceptable for mechanical recycling. The authors mentioned that there was no significant influence of recycling on the thermal stability of the composites. However, the storage modulus of recycled linseed flax composites, recycled chicken feather composites, and recycled jute composites was approximately 66%, 48%, and 36%, respectively, less than that of the composites before recycling. On performing the morphological analysis of recycled composites, authors found that fiber distribution was non-directional but uniform. In addition, poor fiber-matrix adhesion and the presence of cracks and voids on the fiber-matrix interface were observed (Dinu et al., 2021). Because of the high chemical cross-linking of networks in cured thermosets, recycling thermosets is still a challenge (Yue et al., 2021). Cost-effective recycling of thermoset composites will be needed.

(5) A major challenge for external recycling of NFRPCs is the contamination and immiscibility of polymers during recycling. Effective physical presorting (e.g., sorting by size, weight, or color) of components in the mixtures can be developed.

6. Conclusions

Natural fibers have been increasingly used to reinforce polymers. Compared with synthetic fibers, such as glass or carbon fibers, NFRPCs are a sustainable, cost-effective alternative in many applications with low to moderate strength and stiffness requirements. Repetitive reprocessing using combinations of granulation, grinding, shredding, mixing, melt compounding, extrusion, pelletization, compression molding, and injection molding has been used to recycle NFRPCs. However, the commercial recycling of NFRPCs is limited by technical feasibility and cost-effective processing. In addition, there is a research gap regarding the safety of recycling NFRPCs.

This article systematically reviewed the effects of reprocessing/ recycling on the material properties of NFRPCs and discussed the mitigation approaches to overcome degradation of the material properties. For example, hygroscopic behavior should not be a concern for recycled NFRPCs because it is often improved after recycling. The fiber dispersion can be improved after the recycling. However, the dynamic mechanical properties of NFRPCs are more sensitive to reprocessing/recycling if the material is reprocessed beyond a certain number of cycles. NFRPCs can typically be recycled approximately 4–6 times until their mechanical performance decreases. The reduction in viscosity after recycling can be attributed to fiber shortening and polymer degradation. Meanwhile, increasing the fiber content in NFRPCs worsens the weathering resistance of the composites because natural fibers degrade under weathering conditions. The types of fiber, polymer, and reprocessing method significantly affect the mechanical performance of NFRPCs after recycling. Coriander straw is a promising fiber for maintaining the mechanical performance of NFRPCs after recycling. PP and HDPE are promising polymers that are amenable to multiple reprocessing cycles.

Using coupling agents, compatibilizers, and chain extenders can improve the interface between natural fibers and polymer matrices, reducing the chances for degradation occurring and improving the overall composite properties after recycling. The combination of silane and NaOH is a promising coupling agent for maintaining the mechanical performance of NFRPCs after recycling.

Future work needs to be performed to investigate more effective additives (e.g., epoxy and graphene) to further improve composite properties. Fundamental research on the fiber-matrix interface for improving the interfacial adhesion is necessary. Other recycling methods (e.g., pyrolysis and gasification) can be developed for the applications of used NFRPCs.

Declaration of Competing Interest

The authors declare no competing financial interest.

Acknowledgments

(http://energy.gov/downloads/doe-public-access-plan).

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