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Review Article

Hybrid biocomposites

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Biocomposites are being explored by research communityfor the past few decades due to rising environmental concerns. Moisture uptake, poor interface and moderate to lower mechanical properties have proved to be a bottleneck for biocomposites to partially or fully replace synthetic composites. The concept of hybridising, i.e. incorporating two or more reinforcements instead of single reinforcement in a matrix, has the potential to overcome this bottleneck by exploiting the desired contribution of both the reinforcing phases. Although hybrid composites have already extensively explored for high-performance fibres, the studies on hybrid composites using biodegradable constituents still lag behind. This review addresses studies conducted on hybrid composites using at least one biodegradable component and the findings in terms of properties and factors affecting them are discussed. The analysis on synergistic effect reported by researchers along with the scope of ongoing research and prospects for hybrid biocomposites have also been discussed in detail.

Keywords: Biocomposite, Green composite, Hybrid effect, Hybridisation, Natural fibres, Surface treatments

1 Introduction

There is a continuous demand for lightweight materials that have superior strength and toughness, for various applications. Polymer matrix based composite materials came into existence to fulfil this demand. Composites find application in different fields, such as automobiles, aircrafts, sports accessories, helmets, household furniture, etc. Biocomposites are a class of composites where the matrix or the reinforcing phase is biodegradable. Such biocomposites having both the phases (reinforcing phase and matrix phase) biodegradable are termed as Green Composites. The biodegradable components in biocomposites are usually obtained from nature. Frequently reported natural fibres and fillers used as reinforcements arejute[1–6], banana[5,7–9], coir[10–15], sisal[16–21], hemp[22–27,] rice straw[28–30], clays[31,32], wood flour[5,33–35], coir shell[12,36] and natural biodegradable matrices such as starch[29,37], polylactic acid[28,38,39], soy[40,41] etc.

Hybridisation of composites with fibres of different nature was among one of the various strategies that were applied to carbon fibre composites to reduce their brittleness and achieve higher toughness properties[42]. The combination of reinforcement in hybrids is selected in such a way that they inherently possess different properties of each component. The outcome of this approach ultimately reflects in the final composite that was earlier limited by the properties of one reinforcement only. This approach also provides a wider window to choose different combinations of reinforcements that will result in a desired set of properties. Hybrid composites demonstrate certain deviation from the expected mechanical properties. The deviations may occur due to the synergistic effect, i.e. positive hybrid effect or negative hybrid effect (less than the expected).

Biocomposites, having two or more reinforcing phases in a single matrix, are called hybrid biocomposites. The reinforcements maybe present in different physical forms. Various possible combinations are fibres with other fibres, fibres with particulate fillers, two different types of particulate fillers, layered fibrous mats, etc. An overview of hybrid biocomposites among other classes of composites is represented in Fig. 1.

The hybridisation phenomenon and the hybrid composites for high-performance fibres, such as carbon, Kevlar and basalt, have been widely studied but biodegradable reinforcement based hybrid biocomposites have been explored to a lesser extent. Researchers have tried natural fibres and fillers in combination with man-made reinforcements. Very few researchers have reported studies where both the reinforcing phases consist of biodegradable fibres.

This paper aims to review hybrid biocomposites, i.e. composites that use at least one biodegradable fibre or filler as the reinforcement. This review

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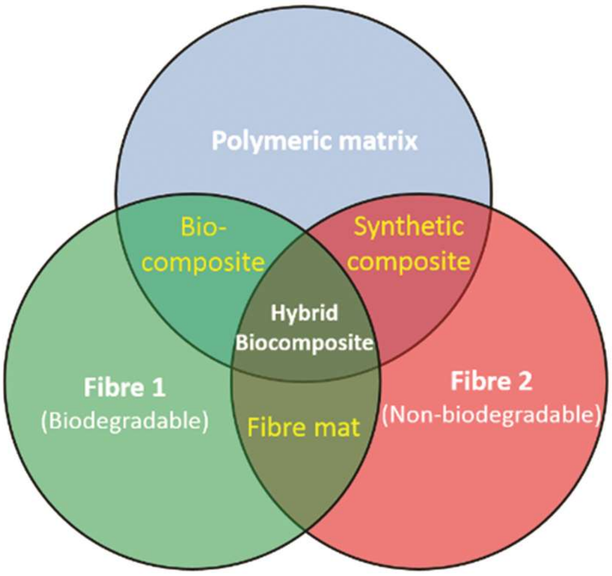

Fig. 1 — Representation of biocomposites and hybrid biocomposites

consists of eight sections out of which the first section is about the introduction and background of biocomposites and hybrid composites. Next section will focus on various types of biodegradable fibres and fillers that are reported in hybrid composites. This section will also discuss different types of matrices that are used in the studies of hybrid composites consisting of above-mentioned bioreinforcements. The third section will deal with chemical surface treatments that enhance the interface between dissimilar surface of polar and hydrophilic natural fibres with nonpolar hydrophobic matrices. Studies on mechanical properties of hybrid biocomposites are mentioned in the fourth section. Fifth section consists of factors, such as total fibre volume fraction, ratio between fibre volumes, length of fibres, orientation of mats, layering sequence, dispersion of fibres, etc. and their effect on the end properties. This section also includes parameters which were found crucial for a set of desired properties. Sixth section is about the studies carried out on the phenomena of hybridisation and the positive or negative hybrid effect demonstrated by researchers in the case of hybrid biocomposites. The next section (conclusion) of the review discusses the gaps in ongoing research in natural fibre or filler based hybrid composites. Finally, the future scope with emerging fields of study in hybrid biocomposites and new possibilities are mentioned in the eighth section.

2 Reinforcements and Matrices

2.1 Reinforcing Phases

Researchers have reported the use of various types of biodegradable reinforcements in hybrid biocomposites. Almost all the reported reinforcements are obtained from nature and majority have cellulose as the main constituent. Most researchers reported hybrid composites having two reinforcing phases. There are very few published papers with more than two reinforcing phases. Studies with three reinforcing phases are reported by some researchers[43–45]. It has also been observed that the use of particulate fillers in hybrid biocomposites is limited. Fibres, in the form of fibrous webs, woven and nonwoven have been mostly used as reinforcements by researchers.

Various types of natural fibres are explored in combination with man-made or natural fibres, although studies on hybrid biocomposites with both the reinforcements from natural resources are fairly outnumbered by those having one natural fibre and another man-made fibre. A fair proportion of these man-made fibres, used as one of the constituents, is glass fibres in different physical form. Studies on particulate filler based hybrid biocomposites are also reported. Wood flour was used with coir[46] and kenaf fibres[34,47]. Fly ash was used as filler with jute fibres in epoxy resin[45]. Carbon black was used in natural rubber matrix with pineapple leaf fibres[48]. However, the share of studies reported with particulate based fillers is relatively less. Different type of fibres and fillers used as reinforcements are mentioned in Table 1.

It can be noticed from Table 1 that majority of literature on hybrid biocomposites involves fibres, majority of which involves one reinforcement as glass in fibrous form. Limited studies on use of particulate fillers have been reported in hybrid biocomposites.

2.2 Matrix Phase

Thermoset matrices have been predominantlyused with majority of the above-mentioned reinforcements. Polyester and epoxy based resins are widely reported in literature of natural fibre based hybrid composites. However, other thermoset resins such as phenol formaldehyde and novolachave also beenreported. It is generally observed that thermoset resin can impregnate the fibres efficiently and thus results in better penetration into the fibrous web and layers. It may possibly be the reason why natural fibre based hybrid composites have so far used thermoset resins to such a large extent. A few studies with thermoplastic resins such as polypropylene[34], polyethylene[21], thermoplastic natural rubber[51] and soybean oil[41] are also available. Table 2 mentions different types of matrices used in hybrid biocomposites and the studies in which they were reported.

Table 1 — Different reinforcements used in hybrid biocomposites

Reinforcements		References
Fibres	Glass	2,4,18,21–24,26,27,36,38,41,43,45,49–88
	Jute	1,3–6,15,16,18,25,43,64,65,68,73, 75–78,80,83,86,89–93
	Flax	14,27,41,57,66,71,80,82,94–100
	Coir	8,11,12,14,15,88
	Hemp	22,23,25–27,33,101,102
	Banana	5,7,8,19,90,91,103–106
	Kenaf	34,38,47,67,69,72,74,79,81,89,107,108
	Sisal	7,11,16–21,52,84,85,89,103,109–111
	Oil palm fibre	6,20,31,50,56,58,70,89,109
	OPEFB	51,108
	Cotton	17,110
	Silk	35,54
	Bamboo	36,59,77,112
	Pineapple	60
	Lyocell	101
	Seaweed	37
	Palmyra	55
	Bagasse	87,113
Particulate fillers	Wood flour	34,35
	Coconut shell	33
	Fly ash	45
	Clay	31,32
	Cornhusk flour	114
	Silica	19

Table 2 — Different matrices used in hybrid biocomposites

Matrices		References
Thermoset	Epoxy resin	3,4,6,16,18,19,27,33,36,43,45, 47,50,52,54,57,64,65,67,68,71, 75–77,80,82,88,90–93,96–100,102,106,111,112,115–118
	Modified epoxy resin	14,15,26,58,107
	Polyester	2,5,7,8,17,22,25,30,38,53,55, 60,69,72,78,81,86,87,103,104, 110,119
	Natural rubber	11,20,48,51,89,109,120
	Phenolic resin	1,56,66
	Soybean oil matrix	41
Thermoplastic	Polypropylene	12,23,32,34,70,74,85,105
	Polylactic acid	101,114
	Polyethylene	21,31

Table 2 shows that thermoset resins have predominantly been used as compared to thermoplastic polymers as matrix in hybrid biocomposites. Epoxy based resins are most widely used as matrix phase. Epoxy resins are a popular class of matrix used for thermoset composites. The proper selection of resin, modifiers, and cross-linking agent actually enables properties of cured epoxy resin to be tailored to achieve specific performance characteristics. There are various properties for which epoxies are favoured as a resin.

Epoxies have very high chemical resistance, particularly to alkaline environments. It has outstanding adhesion to a variety of substrates. As far as mechanical properties are concerned, they have a combination of high tensile, compressive, and flexural strengths. Shrinkage in thermoset resins is often a cause for concern. The epoxies have shown very low shrinkage on cure. For applications where electrical insulation is important, epoxies play a good role. The epoxies also show good resistance to corrosion. The fatigue strength is superior when compared to other thermoset materials.

3 Surface Treatments

As already mentioned in the introduction section, hybrid biocomposites have at least one reinforcement which is biodegradable. Most of these reinforcements are lignocellulosic and have different content of lignin, cellulose and hemicellulose. Lignocellulosic fibres and fillers have polar groups and are hydrophilic. When incorporated in synthetic matrices which are non-polar, absence of desired interfacial adhesion leads to inferior mechanical properties of biocomposites. Various chemical and physical surface treatments are explored for cellulosic fibre based reinforcements for composites. However, there are no reports of using physical surface treatments like Corona and plasma discharge for hybrid biocomposites. There are many chemical surface treatments which have been reported for biocomposites, but out of these, only a few are reported in hybrid biocomposites as well.These chemical treatments are discussed below.

3.1 Alkali Treatment

Various studies, as shown in Table 3, reports chemical treatment of fibres or fibrous structures prior to their incorporation in matrix. Alkali treatment using NaOH for fibre surface treatment in hybrid biocomposites, is most widely reported by the

Table 3 — Details of alkali treatment carried out by researchers

NaOH conc., %	Temperature of treatment, °C	Fibre	Duration of treatment h	Processes subsequent to alkali treatment	Ref.
0.5-4	-	Sisal-oil palm	1	0.1 % acetic acid	89
0.5-10	Room temp.	Sisal-oil palm	1	NIL	109
1.5	Room temp.	Sisal, rice husk, sugarcane bagasse	48	NIL	44
2	-	Flax	0.25	Dipped in acrylonitrile for 3 h and washed with 0.5 N acetic acid for grafting of –CN group	41
2	-	Coir	1	NIL	13
2-10	300	Coir	1	NIL	53
2-12	-	Sisal	0.5	Dilute HCl#	21
4	-	Banana	1	NIL	104
5	-	Hemp	24	NIL	33
5	-	Kenaf-oil palm	1	NIL	108
5	-	Sugarcane bagasse	72	NIL	87
5	70	Coir	2	NIL	121
5	Room temp.	Hemp	1	2% glacial acetic acid	22
5	-	Cotton-kapok	2	NIL	30
5	-	Jute	4	Dilute acetic acid#	15
5	30	Jute	8	NIL	92
5	-	Sisal-Jute	0.5	Dilute HCl#	16
5,10	30	Pineapple-sisal	1	NIL	52
5,10	70	Jute-coir	2.5	Neutralized with acid#	122
6	-	Kenaf	3	NIL	115
6	-	Kenaf	24	NIL	67
6	Room temp.	Kenaf	3	NIL	38
6.4	Room temp.	Oil palm	48	5% acetic acid	31
6.4	-	Coir	48	acetic acid#	12
10	-	Pineapple	1	Dilute acid#	60

(-)Indicates that the temperature of treatment has not been mentioned.
# Indicates concentration of acid/alkali has not been mentioned.

researchers. The concentration of NaOH from0.5% to 10%has been reported depending upon the fibre type and time of treatment, as mentioned in Table 3. Alkali treatment takes away excess lignin from the surface of lignocellulosic fibres and exposes the rough cellulosic surface to interact with the matrix to form an improved interface. Alkali treatment was reported to reduce the silica content of rice husk particles, resulting in improved adhesion[44]. NaOH treatment also reduced agglomeration of glass fibres in hybrid biocomposites and led to uniform distribution of fibres, ultimately resulting in higher mechanical properties. However, the explanation for reducing the agglomeration of glass fibres by alkali treatment was not discussed by the author[21]. Increase in storage modulus and decrease in damping properties of natural rubber-sisal-oil palm hybrid biocomposites after treatment of fibres with 4% NaOH was reported and the improved interface between fibres and matrix was thought of the reason behind the observation[89]. Similar trend and explanation were alsoobserved with the same hybrid composite after the silane treatment[124].

As evident from Table 3, the concentration of sodium hydroxide treatment varies from 0.5% to 10% and 4-6% sodium hydroxide solution is mostly reported in the literature. However, the temperature of alkali treatment and after treatments have not been reported in many studies. Duration of alkali treatment also has large variations, as can be seen in the table.

3.2 Silane Treatment

Silane treatment is the second most popular surface treatment of natural fibres, reported in hybrid biocomposites. In a study by Mehta *et al.*[22], a solution of ethanol and water was used to dissolve ϒ-MPS. The concentration of solution was 1%ϒ-MPS and *p*H was maintained at 4 using 2% glacial acetic acid. Fibre mats were dipped in the solution for one hour and later drained, dried and cured in an air oven. Same research group followed a similar procedure to treat jute and hemp fibres[25]. Another study reported the treatment of alkali treated sisal fibres using heated reflux process by heating fibres in a reflux vessel using a mixture of 5% silane (by weight of fibres), carbon tetrachloride and dicumyl peroxide (DCP) 2.5% (by weight of fibres). Fibres were finally filtered and dried until constant weight was attained[21]. This study proposed that vinyl group in silane under goes polymerisation in the presence of dicumyl peroxide and forms a chain of hydrophobic polymer on the surface of sisal fibres. This modified hydrophobic surface of the fibre resulted in the formation of Van der Wall bonds with the hydrophobic polyethylene matrix. The research team proposed that silane acted as a compatibiliser and connected two incompatible surfaces. Reduction in water absorption by silane treatment of sisal and oil palms fibres due to strong hydrogen bonding between hydroxyl groups of cellulosic fibres and highly electromagnetic fluorine atoms was reported by Jacob *et al.*[123].

3.3 Other Chemical Treatments

Apart from alkali and silane treatments, few studies on other chemicals were also reported by researchers. Acrylonitrile treatment of hemp fibres was reported by Mehta *et al.*[22] along with alkali and silane treatment (Fig. 2). Hemp fibre mats were drained and dried after treating with 3% acrylonitrile, 0.5% DCP and 96.5% of ethanol for 15 min. Rout *et al.*[53] reported acrylonitrile grafting for alkali treated coir fibres. The grafting was carried out in an aqueous solution having Cu^{2+} - IO^{-4}initiator. Bleaching of coir fibres using 0.7% textone (sodium chlorite) solution at *p*H 4 and temperature 65°-85°C for a duration of 2 h was also discussed in this study. Bleaching of fibres at 65°C was found to be optimum for tensile and flexural properties. It was found that bleaching reduced the lignin content which resulted in less stiff and flexible fibres. The fibres had better tensile properties.

In a study by Idicula *et al.*[60], banana, sisal and pineapple were refluxed in 5% solution of PSMA (Polystyrene maleic anhydride)in toluene. This treatment with PSMA resulted in improved compatibility between fibres and polyester matrix. The interface in these composites was enhanced by chemical treatment. It was proposed in the research that PSMA reacted to hydroxyl groups of fibres, resulting in a hydrophobic surface. The hydrophobic surface formed by chemical modification resulted in chemical interaction with the hydrophobic centres of the polyester matrix. Fibrillation of the fibres in the microscopic image of the fracture surface of composites confirmed the hypothesis that the researchers proposed. The chemical treatment of fibres improved contact between fibre and matrix. This led to increased density of banana-sisal hybrid biocomposites minimising thermal resistance[60]. A hypothetical representation of the surface of PSMA treated fibres is shown in Fig. 3.

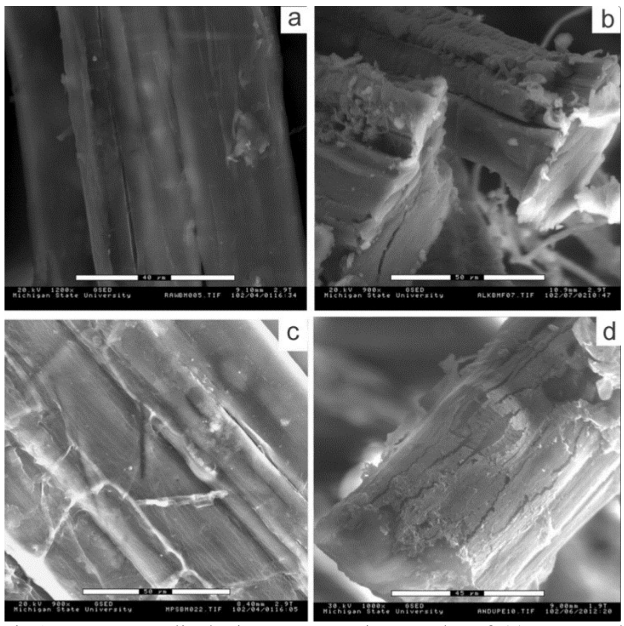

Fig.2 — Images displaying ESEM micrographs of (a) untreated (×1200), (b) alkali treated (×900), (c) silane treated (×900), and (d) acrylonitrile treated (×1000), hemp fibre[22]

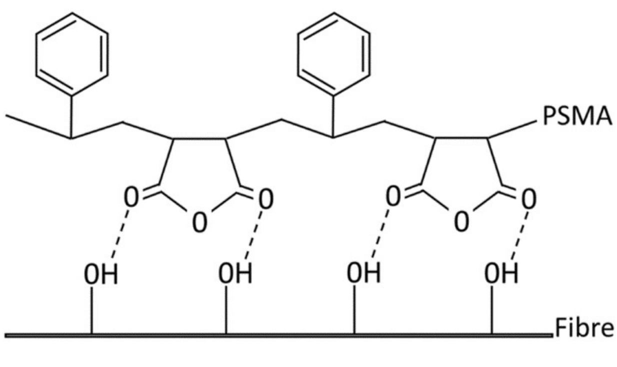

Fig.3 — Proposed model of interaction of polyester and PSMA treated fibre (redrawn on the basis of Idicula *et. al.*[60])

Study by Kalaprasad *et al.*[21]elaborately discussed various chemical treatments for sisal fibres. Effect of alkali treatment using sodium hydroxide, acetylation using acetic acid, permanganate treatment, stearic acid treatment, peroxide treatment and silane treatment on tensile properties of sisal-glass hybrid composites has been reported. Alkali and silane treatments have already been mentioned in previous section. For permanganate treatment of sisal fibres, $KMnO_4$ was dissolved in acetone to produce solutions with various concentrations ranging from 0.02% to 0.2%. Alkali treated sisal fibres were dipped in this solution for two minutes followed by drying. It was proposed that highly reactive MnO_4^- ion initiated the grafting of polyethylene on surface of sisal fibres and thus resulted in increased tensile strength of the composites. Tensile strength improved because adhesion between fibres and matrix was enhanced by grafting. However, the enhancement in tensile properties was observed only till 0.06% concentration of permanganate during the treatment. Further increase in concentration led to degradation of cellulosic sisal fibres. Similarly, the treatment of acetylation and stearic acid imparted hydrophobicity to the fibre surface and led to enhanced adhesion between fibre and matrix.

Similarly, acetylation, permanganate treatment using0.06% $KMnO_4$, stearic acid treatment, peroxide treatment with a combination of 1% dicumyl peroxide, peroxide treatment using0.8% benzoyl peroxide and matrix treatment by 8% maleic anhydride grafted polyethylene were found optimum for sisal-glass hybrid composites[21]. The study also proposed the order of effectiveness of chemical treatments as NaOH < acetic anhydride < stearic acid < KMnO4 < maleic anhydride < silane-A < DCP < BPO for the sisal and glass fibres. Figure 4 shows the effect of various chemical treatments of fibres on Young's modulus of hybrid biocomposites. Interestingly, it is the only reported study where glass fibres were chemically treated. Glass fibres were treated with silane (0.5% by weight) in 0.1 M acetic acid solution followed by drying at 100°C for 20 min[21].

3.4 Matrix Modification

Besides the surface treatment of fibres, a study on modification of matrix has also been reported in literature for hybrid biocomposites. In this research work, maleic anhydride was used for modifying polyethylene and blended with LDPE at varying concentrations from 1% to 14% weight of LDPE. Increase in tensile strength was reported up to 8%

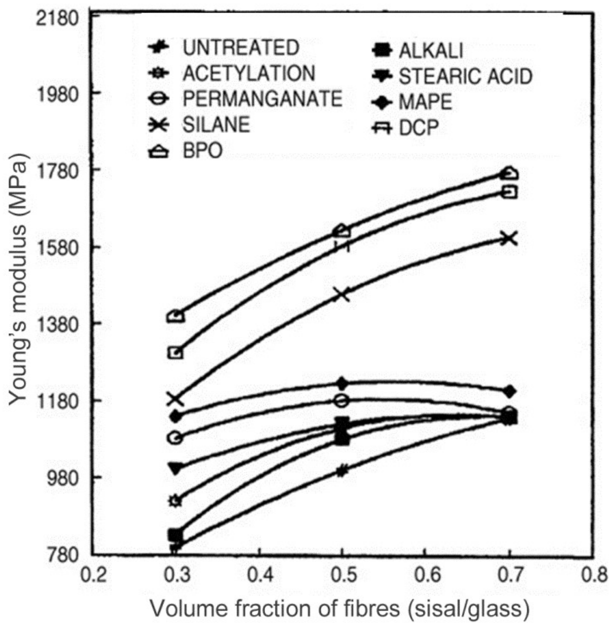

Fig.4 — Effect of various chemical treatments on Young's modulus of sisal/glass hybrid biocomposites at varying fibre ratios[21]

concentration and it was proposed that there may be dipolar interactions between anhydride groups of maleic anhydride grafted polyethylene and hydroxyl groups on cellulosic and glass fibre surfaces. Hypothesis was backed by FTIR analysis which displayed a strong peak at 1640 cm^{-1} and matrix stuck to fibre's fracture surface as detailed in the microscopic image. In this case the matrix was modified to interact better with fibres. Microscopic images of fibre pull out in fracture surface verified the improved adhesion. Peroxide induced grafting of glass and sisal fibre was also carried out and following chemical mechanism starting from peroxide free radical was proposed by the authors[21]:

PO-OP → 2PO•

PO• + PE-H →POH+ PE•

PO• + glass-H →POH+ glass•

PO• + sisal (cellulose-H) →POH+ cellulose

PE• + cellulose• + glass• → Cellulose-PE-glass

PE• + PE• → PE-PE

Treating natural fibres for surface modification is not very often reported in case of hybrid biocomposites. In the available literature, several studies did not undergo any surface treatment for the cellulosic fibres or fillers. Alkali and silane treatments were mostly reported chemical surface treatment technique used for lignocellulosic fibres for the hybrid biocomposites in the remaining studies. There are few dedicated studies on the effect of various chemical treatments in context of hybrid biocomposites. It was observed that all chemical treatments modified the hydrophilic surface of cellulosic fibres to hydrophobic, thus improving their wettability with hydrophobic matrices. Improved wettability of initially dissimilar surfaces resulted in stronger and improved interface, as well as better penetration of resin into the fibre bed. All the hybrid biocomposites that incorporated fibres treated with different chemical processes, reported improvement in mechanical properties. Figure 5 provides a pictorial representation of the share of various reinforcements, matrices and chemical treatments reported for hybrid biocomposites based on the literature cited in this paper.

4 Mechanical and Physical Properties

Hybrid biocomposites have been studied for various properties. Mechanical properties, such as tensile, flexural and impact, are the most widely studied properties. From the available literature, it can be observed that different approaches to manufacture hybrid biocomposites and study of different parameters, such as chemical treatments, fibre volume fraction and layering pattern, are usually directed towards achieving improved mechanical properties.

The effect of these approaches and parameters on tensile flexural and impact properties are widely investigated by the research community. Several other properties have also been studied by researchers for hybrid biocomposites and are discussed in the subsequent sections.

4.1 Tensile Properties

Tensile strength and tensile modulus are widely studied in hybrid biocomposites. It has been observed that incorporation of glass fibres or glass fibre laminates in natural fibre based composites resulted in improved tensile strength and modulus. The increase in tensile strength and modulus depends upon the type of fibre andthe fabrication process of composites. Incorporation of 10% of glass fibres in 30% hemp fibres leads to an increment of 76% in tensile strength and 34% in tensile modulus[22]. Addition of 10% glass fabric by weight results in 4.52% increment in tensile strength of silk composites[54]. The chemical surface treatments reported in various studies for natural cellulosic fibres report improvement in tensile properties for resulting hybrid composites[15,21,22,25,35,53]. The enhancement in tensile

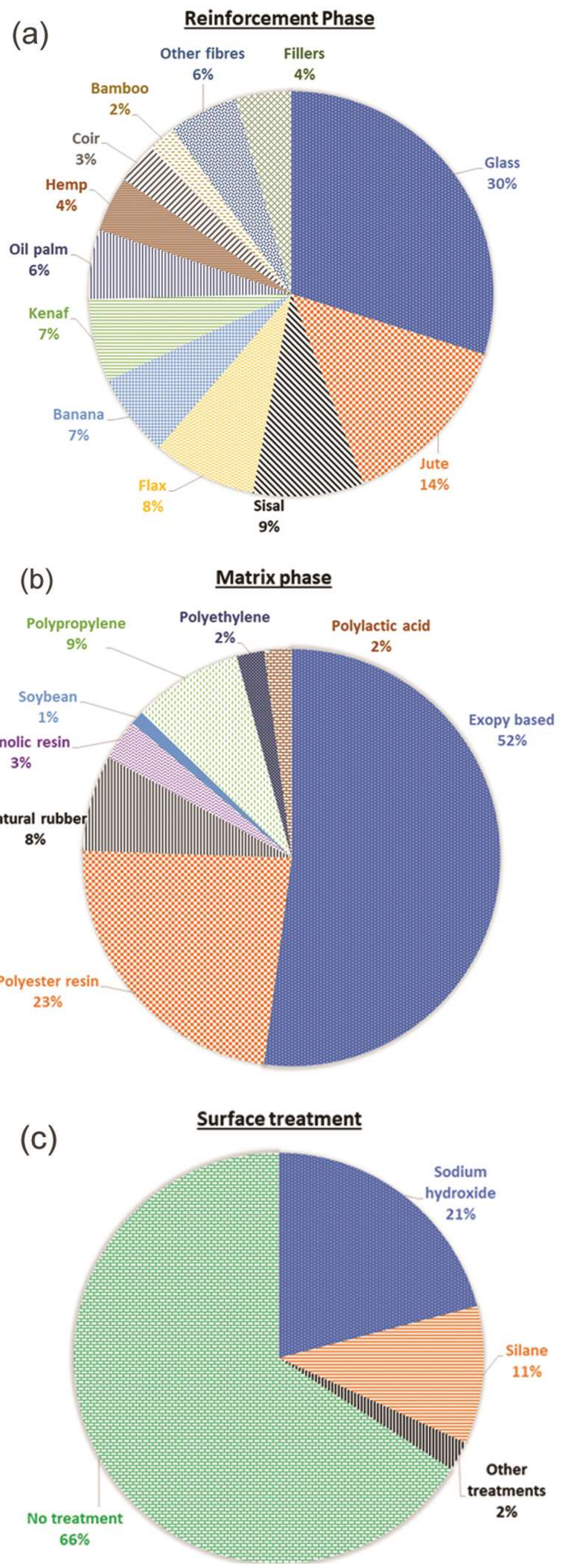

Fig. 5 — Percentage share of (a) various reinforcement, (b) matrix phases and (c) chemical surface treatments as per the literature cited in this review paper

strength was due to a stronger interface between modified fibre surface - matrix and better homogeneity in the composites. The improvement in properties was observed due to enhanced wettability of fibre that allowed matrix to reach fibres which was inaccessible before the chemical treatments. Tensile modulus also improved due to the deeper strengthening of matrix and its increased stiffness.

A study of layering pattern and mixing in sisal-banana based hybrid composites and its effects on mechanical properties was carried out. The pattern of layering did not yield any significant effect on the tensile strength. It was observed that the composite samples with intimately mixed fibres transferred the load efficiently between different types of fibres, which was not possible in the case of laminated composites. The tensile modulus showed improvement with the bilayer arrangement due to the initial stress being taken up by stiffer banana fibres at same value of strain[7]. Comparision of experimental values of tensile properties were carried out with various theoritical models, such as rule of hybrid mixture (ROHM)[50,117], parallel model[10,49] and Hirsch model[7,49].

4.2 Flexural Properties

Flexural properties have been the part of research in majority of mechanical testing of hybrid composites. Various approaches to produce hybrid biocomposites also discuss their effect on flexural properties. Different mechanism such as tensile, compression and interlaminar shearing occur simultaneously during flexural testing[49]. Failure takes place after the bending and shear failure[22,52].

In a study reported by Mehta *et. al.*[22], flexural strength of hemp composites increased by 83% and modulus of elasticity by 66% when glass fibres were incorporated. The bending modulus of glass- hemp hybrid biocomposites was found to be 7% higher than the pure glass composites. However, the explanation of enhancement of flexural properties was provided only for treated hemp fibre hybrid composites and not on account of the synergistic effect occurring due to hybridisation of composites[22]. In another study, 10 %loading of glass and 30 %of hemp fibres were used in hybrid biocomposites. Flexural strength improves 62% at 4% of maleic anhydride coupling agent in PP and there was a marginal improvement in flexural modulus at the same coupling agent content when compared to samples with no coupling agent. The reason for enhancement of flexural strength was not discussed by the author[23]. In glass and OPEFB (oil palm empty fruit bunch) fibre hybrid biocomposites, 0.55 volume fraction of OPEFB fibres resulted in optimum flexural strength, whereas 0.76 volume fraction gives better flexural modulus property at total fibre loading of 40% due to optimum glass fibre content at these volume fractions, which resists the shear developed in the biocomposites[52]. In pineapple leaf fibre and glass hybrid composites, the incorporation of glass from 4.3% to 12.9% increased the flexural strength from 85.61MPa to 101.25 MPa. The increase of flexural strength, in this case, was almost steady. At 30% total fibre loading, the sisal-glass fibre hybrids, the incorporation of 2.8% of glass fbres increases the flexural strength of hybrid composites by 25%. Surface treatment of sisal fibres by cynoethylation resulted in the maximum flexural strength of 151.57 MPa. It was discussed that β-cyanoethyl group on the fibre surface is chemically bound to polyester matrix to give superior interface properties, which ultimately leads to better flexural strength composites[52]. Similarly, surface treatment of coir by bleaching resulted in 20% improvement in flexural strength when compared to untreated hybrid composites of coir and glass. Here, the composition of coir and glass was13% and 7% respectively. The improvement in flexural strength by bleaching was found to be more effective as compared to alkali treatment or acrylonitrile treatment of coir fibres. The bleaching treatment resulted in loss of lignin that reduced the stiffness of fibres, ultimately imparting flexibility to the composites[53].

Various other studies on surface treatment of lignocellulosic fibres have also been reported, resulting in improvement of flexural properties of hybrid biocomposites. Acrylonitrile grafting on surface of hemp fibres displayed better improvement in flexural strength than alkali or silane treatment in the case of hemp-glass hybrid biocomposites due to covalent bond formation with highly polar –CN functional groups. It was also observed that flexural strength does not essentially follow the trend as observed for tensile properties in the hybrid biocomposites. This was due to the pattern of layering and mixing of two fibres or fillers in the hybrid composites[22]. The effect of mixing of fibres and layering pattern on the hybrid effect has been discussed in a study by *al.*[21]. It was stated that biocomposites having intimately mixed fibres displayed better tensile properties and positive hybrid effect because the interaction between fibres of one type is reduced after incorporation of another fibre. Optimum interaction between matrix and the fibres, as well as between two types of fibres, is achieved at a particular ratio. In case of laminated hybrid composites, such interaction is missing and stress concentrations are generated, leading to variable strain in different layers, and delamination and failure during tensile loading. This also results in a negative hybrid effect in tensile properties.

Tensile behaviour of hybrid biocomposites does not always conform to the flexural or impact behaviour. Flexural properties depend upon the tensile as well as compression behaviour of the material simultaneously. Samples with intimately mixed fibres displayed poor flexural properties. The samples with sandwich type stacking of different layers of fibreswere found to have better flexural properties and displayed positive hybrid effect, depending upon the layer facing tension or the compression during bending. Stiffer fibre layers on the extreme faces resulted in positive hybrid effect for flexural properties of laminated hybrid biocomposites. It was observed that ASTM D790 involving three point bending method was the most popular method of testing the flexural properties of the hybrid biocomposites[1–4,8,16–18,22–25,27,30,34–38,41,47,49,52,53,55,58,61–65,67,69,71,77,90,91,99,100,107,116,122,125] and was frequently used by the research community.

4.3 Impact Properties

Impact strength of a composite is directly related to its overall toughness and is determined by its ability to resist fracture under high-speed stress. Fibrous network arrests crack propagation and restricts continuous matrix towards catastrophic fracture. Tensile and flexural properties are more relevant in such applications[23]. The impact properties are dependent upon interlaminar and interfacial properties[52].

Investigation of impact properties in hybrid biocomposites has been carried out by various researchers and the effect of (a) hybridising, (b) surface treatments, and (c) nature of testing have also been discussed. Izod impact testing of hemp fibres hybridised by two different fibres glass and jute displayed 9% and 693% (ref. 24) improvement in impact strength respectively when compared with pure hemp composites. At a total fibre loading of 40%,the incorporation of glass fibres into the OPEFB composites resulted in drastic improvement in the impact strength[49]. Maximum enhancement in the impact properties was observed at 0.74 volume fraction of OPEFB fibres. At thiscomposition, the impact strength was even better than the hundred percent glass fibre composites. The reason for this enhancement was not mentioned by the author and may be a subject of further investigation. This study also discussed the mechanism of impact fracture using impact factography. It was established that combined effect of debonding and fibre pull-out during impact fracture led to higher energy absorption. The pullout of oil palm fibres was lesser than that of glass, and thus it was also concluded that glass fibres had poor interaction with phenolic resin. This was the reason why the hybrid composites of oil palm -glass fibres showed better impact strength than fully glass composites.

Surface treatment of hemp and glass hybrid biocomposites by alkali, silane, acrylonitrile grafting, etc., resulted in a reduction of impact strength. However, the authors did not provide any explanation for reduction in impact strength by this chemical surface treatment[22].A study for glass and hemp hybrid composites having 10% and 30% weight composition of respective fibres, discussed the effect of interface between fibres and matrix[23]. This study found that the matrix with maleic anhydride as coupling agent improved interfacial adhesion up to a concentration of 4% by weight of polypropylene matrix. It was found that the coupling agent played a vital role in determining the impact strength of hybrid biocomposites. Strong interfaces led to brittle failure of biocomposites, while a poor interface which caused due to absence of coupling agent resulted in easy fibre pull out, leading to a subsequent decrease in impact strength. It was concluded that the interaction between fibre and matrix surface should be optimum to attain better impact properties. This study also discussed the notched and unnotched impact strength of hybrid composites.

In a study by Rout *et al.*[53], using total fibre loading of 25% by weight, PALF- glass hybrid composites displayed highest impact strength of 127.38 J/m for 8.6% of glass fibre loading. In case of sisal-glass composites, the addition of glass in sisal glass composites displayed higher impact strength then PALF- glass due to the relatively higher modulus of glass fibres. The total fibre fraction in sisal-glass hybrids biocomposites was maintained at 30% of hybrid biocomposite. It was further concluded that the chemical treatment of PALF or sisal fibres by alkali or cyanoethylation resulted in higher impact strength than untreated control samples of hybrid composites of glass with both these fibres. On the basis of results, alkali treatment was recommended for better tensile and impact properties, whereas cynoethylation was found to be better suited for flexural properties in both the hybrid composite samples[52]. For better impact properties, alkali treatment with 5% NaOH was also recommended over bleaching or acrylonitrile grafting treatment of coir fibres in polyester matrix based coir-glass hybridcomposites[53]. A study of sisal banana hybrid composites showed improvement in impact strength when the fraction of sisal fibres was increased, while keeping total fibre loading as constant. It was concludedthat the hollow sisal fibres provided better impact properties and their higher fibrillar angle of 20^o resulted in improvement of impact strength when sisal content increases in sisal-banana hybrid biocomposites[7].

Different types of impact testing techniques and standard test methods used by researchers for determining the impact properties of hybrid biocomposites are mentioned in Table 4. It should be noted that test method ASTM D6110 is the dedicated standard for charpy impact testing and is evolved from ASTM D256-B. The previous versions of ASTM D256 allowed specimens with Appendix X2 calibration procedure, eligible to be tested for charpy impact testing also**.**

Overall, it can be concluded that the incorporation of high strength, tough fibres at optimum volume fraction and moderate interfacial addition with, matrixas well as layering pattern of laminates and mixing of reinforcementplay a key role in improving the impact properties of the hybrid biocomposites. The placement of laminates and the type of testing plays a significant role in determining the impact properties of hybrid biocomposites.

Table 4 — Different types of testing techniques to determine impact properties of hybrid biocomposites

Impact testing	Standard	References
Izod	ASTM D256	1,4,16,17,22–25,30,35,37,38, 49,52,53,58,61,62,67,90,125
	ISO 180	55
Charpy	ASTM D6110	122
	ASTM D256	30,47,65,91,107
	ISO 179	3,63,78,99,101
Falling weight	ASTM D7136	27,100
	ASTM D3763	27,41
	Self-designed setup	100,126

4.4 Other Properties

Apart from mechanical properties, such as tensile, impact and flexural as well as other properties, such as dynamic mechanical properties, swelling, water absorption, thermal or sound barrier, have also been discussed by various researchers. However, the studies on these properties in the case of hybrid biocomposites are relatively less. A study investigated the storage modulus, loss modulus and damping behavior of coir-bagasse reinforced epoxy novolac hybrid biocomposites[113]. These properties were discussed as a function of frequency, temperature and pattern of placement of fibres in the composites. The dynamic mechanical behaviour was investigated at regions below and above glass transition temperature. Storage modulus of bilayered composites increased with the rise in frequency from 1Hz to 20Hz. The damping behaviour of trilayer arrangement in the same frequency displayed lesser energy dissipation as the frequency increased. It was also discussed that dynamic mechanical properties are in close relation to static mechanical properties. The homogeneity of composite along with efficient interface between fibre and matrix was ascribed as the reason for better results in static mechanical and dynamic behaviour[113]. Similar trend in storage modulus and damping behaviour was observed in coir and glass hybrid biocomposites when the frequency was varied from 1Hz to 10Hz. It was observed that introduction of stiffer glass fibres in the hybrid biocomposites led to better performance in storage modulus even in the rubbery temperature region[63].

Glass and oil palm hybrid biocomposites were investigated at eight different volume ratios within fixed total fibre loading of 40%. The damping value of hybrid biocomposite was more than neat oil palm biocomposites because introduction of less hydrophilic glass fibres led to a weaker interface with phenol formaldehyde resin. An effective immobilized region of matrix could not be formed at the interface, ultimately resulting in higher damping values. The storage and loss modulus of hybrid biocomposites increased as the content of oil palm fibres reduced due to less efficient packing of glass and oil palm fibres in hybrid composites, which hinders their efficient wetting by the matrix[56]. The loss and storage modulus of sisal-oil palm hybrid biocomposites in natural rubber matrix was investigated by Jacob *et al.*[89]. It was observed that the damping values were higher for neat rubber matrix as it has highly mobile polymeric chains. The incorporation of fibres led to stiffening of matrix which reduced its damping behaviour. The effect of chemical surface treatment of sisal and oil palm fibres NaOH resulted in further reduction in damping behaviour of hybrid biocomposites. The storage and loss modulus also improved with treated fibres. It was attributed to a stronger interfacial addition of fibres and matrix that led to larger degree of stiffening in matrix. In all the above discussed hybrid biocomposites it was a common trend that the storage modulus increased when total volume fraction was increased up to a certain optimum fibre loading, due to strengthening of the matrix. The incorporation of fibres led to restriction in the movement of matrix polymeric chains and increasing the storage modulus.

Interfacial adhesion of sisal-coir hybrid biocomposites by restricted equilibrium swelling technique was studied by Haseena *et al.*[11]. Three aromatic solvents were used for swelling the hybrids and their effect with respect to orientation, fibre loading and binding agent was investigated. The machinability of glass square hybrid biocomposites with polyester matrix was optimized using factorial design methodology. Drilling related parameters along with feeding rate was optimised for these hybrids[62]. Noise reduction coefficient and sound absorption coefficient of coir board debris have been investigated by a research group. It was concluded that the porous nature of coir fibres and larger lumen present in it makes it a better choice for use in sound barrier applications[46]. A setup of bamboo- glass fabric and aluminum hybrid composites was discussed to measure the shear strength[59]. Effect of fibre loading, frequency and chemical surface treatment of sisal and oil palm fibres in sisal- oil palm hybrid biocomposites using natural rubber as matrix, on dielectric properties was investigated by Jacob *et al.*[127]. In this study, the addition of hydrophilic lignocellulosic fibres led to a decrease in volume resistivity of hybrid biocomposites. The value of dielectric constants increased due to the polar nature of lignocellulosic fibres. It was noticed that the values of dielectric constant reduced after chemical surface treatment of sisal and oil palm fibres by alkali and silane treatment. Chemical treatments reduced the polar nature of the cellulosic reinforcements. Overall, it was observed that the incorporation of lignocellulosic fibres increased conductivity of hybrid biocomposites because of the presence of polar groups in lignocellulosic fibres which assisted the flow of charge through the composites. Thermal properties such as thermal conductivity, thermal diffusibility and specific heat of cotton based hybrid biocomposites were investigated by Alsina *et al.*[110]. Various combinations of fibres were taken for reinforcement such as jute-cotton, sisal-cotton and ramie-cotton. In this study, the experimental and theoretical values were compared for the series and parallel combinations.

The stress relaxation behaviour of sisal-oil palm fibre reinforced natural rubber hybrid biocomposites, and the effect of untreated and chemically surface treated fibres wereinvestigated by Jacob *et al.*[40]. It was claimed that the chemically treated fibres resulted in better adhesion of fibres with the matrix and resulted in a reduced rate of stress relaxation than untreated fibres. Thermal stability also improved with chemical treatment and amount of loading of both the fibres in hybrid biocomposites.

Water absorption behaviour was studied for glass-silk hybrid biocomposites with epoxy resin. It was reported that the addition of glass fibres in silk biocomposites resulted in reduction of water uptake of composites. This was due to the fact that glass fibres are inherently water impermeable and restricted contact of water with the silk laminates[54]. Similar studies on water uptake with glass fibre based hybrid biocomposites were reported by another research group where glass-kenaf hybrid biocomposites were used[68].

Another investigation on water uptake behaviour of hybrid biocomposites of sisal and oil palm fibres with natural rubber matrix was carried out by Jacob *et al.*[123]. It was observed that water uptake is Fickian in nature when the pure matrix is present and when fibres are introduced, the diffusion of water in composites becomes non-Fickian. The change in the water diffusion characteristic is due to non-homogenous nature of composites which also contain microcracks as well as hydrophilic fibres. Alkalization and silane treatment of sisal and oil palm fibres resulted in reduced water uptake, although the silane treatment displayed better results by uptake of a lesser amount of absorbed water. At higher concentration of alkali, the adhesion of fibres with matrix increased, thereby reducing the velocity and mean free path of diffusing water molecules. Silane coupling agent treated samples displayed more resistance towards water molecules than alkali treated samples due to the presence of strong hydrogen bonding between hydroxyl groups of fibres and highly electronegative fluorine atoms. A different study on flax-basalt reinforced vinyl ester hybrid biocomposites determined the effect of moisture on Mode-I interlaminar fracture toughness by Almansour *et al.*[94]. It was found that hybridisation by basalt fibres diminished the initiation of crack by shielding the swollen flax fibres. External layers of basalt resulted in a reduced detrimental effect of moisture on mechanical properties of flax-vinyl ester composites. Similar observations were made with glass-flax hybrid composites where the moisture uptake was reduced when glass layers were used on external layers[98]. Water uptake behaviour of polyethylene terephthalate-hemp biocomposite was analyzedand it was observed that increasing the content of hydrophobic polyethylene terephthalate reduced water uptake[102].

It can finally be summarized that there are various properties of hybrid biocomposites that have been extensively studied by many researchers. Mechanical properties, such as tensile, flexural and impact, are reported in majority of reports. Three point bending is the most popular testing method for determining the flexural properties and izod impact testing method for the impact properties for hybrid biocomposites. Other properties, such as water uptake behaviour, dynamic mechanical behaviour, dielectric properties and noise barrier properties, have also been reported by few researchers.

A summary of mechanical properties investigated by various researchers for different hybrid biocomposites is mentioned in Table 5. It is to be noted that the table contains only the maximum values of mechanical properties attained during the experiments.

Table 5 — Summary of mechanical properties of hybrid biocomposites

Fibre-1	Fibre-2	Matrix	Tensile strength MPa	Tensile modulus GPa	Flexural strength MPa	Flexural modulus GPa	Impact property	Remarks	Ref.
Flax	Glass	Soyabean oil based	123.3	3.5	130	6.9	2.7 J	Symmetric and asymmetric laminates of glass and flax were used at different volume fractions. Glass to flax ratio of 1:0, 4:1, 3:2, 2:3 and 0:1 was studied at total fibre loading between 31%and 40%	41
Flax	Basalt	Epoxy	-	-	107.4	4.03	-	Role of placement of laminates was studied in detail. Density of composites was measured between 1.11 g/cm^3.and 1.32 g/cm^3. Total fibre fraction was 20- 24%	36
Flax	Glass	Phenolic	450	40.8	-	-	-	Unidirectional composites were fabricated at total fibre loading of 67%. Flax fibres were 32% and glass fibres were 35% of total composite volume.	66
Flax	Basalt	Epoxy	153.16	8.11	137.95	8.02	-	Total fibre loading was maintained at 21.18-24.82% and laminated composites were prepared using resin infusion technique. Density ranged from 1.27 g/cm^3 to 1.31 g/cm^3.	27
Banana	Sisal	Epoxy	31.56	0.39	-	-	-	Hand layup technique was used while maintaining total fibre loading at 40% and 3 different fibre ratios.	9
Banana	Coir	Polypropylene	31.33	0.76	31.33	0.76	73.17 J/m	Fibres were intimately mixed with matrix using screw extruder and injection moulding technique was used to produce 4mm thick composite samples. Banana to coir ratio was 1:3, 1:1 and 3:1	105
Banana	Jute	Epoxy	85.91	-	151.3	1.23	484.54 J/m	Epoxy matrix was readily mixed with fine glass particles and banana to jute fibre ratio was 7:3	91

(*Contd.*)

Table 5 — Summary of mechanical properties of hybrid biocomposites — Contd.

Fibre-1	Fibre-2	Matrix	Tensile strength MPa	Tensile modulus GPa	Flexural strength MPa	Flexural modulus GPa	Impact property	Remarks	Ref.
Banana	Kenaf	Epoxy	58	0.28	24	-	15.81 J	Study primarily targets effects of orientation and stacking sequence on mechanical properties. Composite samples with thickness of 5-9 mm were prepared at fibre loadings of 38.73% to 40.5 %. Banana to kenaf fibre ratio was 40:60, 45:55, 50:50, 55:45 and 60:40	107
Banana /Jute	Glass	Epoxy	57	0.29	12.1	1.45	12 J	Effect of horizontal and vertical orientation of laminates was determined on mechanical properties at fibre loading of 40%.	65
Sisal	Jute	Epoxy	102.08	2.03	361.9	17.5	30.1 kJ/m^2	Results for 3mm thick composites samples reinforced by differently treated fibres at fibre loading of 30% and jute to sisal fibre ratio of 1:0, 1:3, 1:1, 3:1 and 0:1were compared	16
Sisal	Glass	Polyethyene	41.95	1.776.18	-	-	-	Sisal to glass ratio was at 3:7, 1:1 and 7:3. Fibres were intimately mixed prior to making the composites	21
sisal	Banana	Polyester	58	1.60	62	2.95	38 kJ/m^2	Banana to sisal fibre ratio was maintained at 1:1 and total fibre loading ranged between 20% to 50%. For composites with 40% fibre loading, fibre ratio was 1:3, 1:1 and 3:1	103
Sisal	Glass Carbon	Polypropylene	22.4	3.65	52.6	4.51	-	Tribological and mechanical properties of intimately mixed short fibre hybrid composites for sisal/glass and sisal/carbon fibre at 1:3, 1:1 and 3:1 fibre ratio were investigated. Density of composites ranged from 0.907 g/cm^3 to 1.172 g/cm^3	85
Coir	Glass	Polyester	45.8	-	110.9	-	687.8 J/m	The study specifically targets chemical treatment of fibres and the corresponding mechanical properties. Glass to coir fibre ratio was 7:13 and total fibre loading was 20%	53
Coir	Sisal	Natural rubber	15.46	0.0012 to-0.0038	-	-	-	Fibre orientation was found to be dependent on total amount of fibre loading. Samples were 2 mm thick and density ranged from 0.96 g/cm^3 to 1.02 g/cm^3	10
Coir	Coconut shell	Polypropylene	8% inc	50% inc	-	-	-	The reinforcing ingredients were obtained from waste of coir industry. Reinforcement loading was maintained at 20% by weight. Thickness of samples was 2 mm and ratio of fibre to filler was 1:0, 3:1, 1:1, 3:1 and 0:1.	12
Kapok	Cotton	Polyester	52.87	1.64	55.34	0.7	119.2 kJ/m^2	Cotton to kapok ratio was maintained at 2:3 by weight and composite samples were prepared at 5 mm thickness. Total fibre loading was 58- 65% and density ranged between 1.14 g/cm^3 and 1.23 g/cm^3	30

(*Contd.*)

Table 5 — Summary of mechanical properties of hybrid biocomposites — Contd.

Fibre-1	Fibre-2	Matrix	Tensile strength MPa	Tensile modulus GPa	Flexural strength MPa	Flexural modulus GPa	Impact property	Remarks	Ref.
Kenaf	Kevlar	Epoxy	145	3.37	100.3	-	51.41 kJ/m^2	Study exclusively targets the role of fibre orientation and its effects on mechanical properties. Density of composite samples ranged between 0.87 g/cm^3 and 1.1 g/cm^3	47
Kenaf	Kevlar	Epoxy	64.7	5.29	51.28	2.74	50.1 kJ/m^2	Woven laminates of Kevlar and kenaf fibres were fabricated at 30% fibre loading using hand layup followed by compression	115
Kenaf	Glass	Epoxy	-	-	68.1	-	13.1 kJ/m^2	Fibres were intimately mixed and total fibre loading was fixed at 9% by weight and fibre ratio was 1:1	67
Kenaf	Glass	Polyester	194.6	-	291.6	-	-	Study targeted effect of fibre orientation on ageing at total fibre loading of 35% and kenaf to glass fibre ratio of 3:7. Density ranged between 1.31 g/cm^3 and 1.66 g/cm^3	72
Jute	Banana	Epoxy	17.89	0.72	59.84	9.17	18.23 kJ/m^2	Laminates of fibres were cross plied using hand layup technique at 1:3, 1:1 and 3:1 fibre ratio. Composite samples were 2 mm thick. No chemical treatment of fibres was carried out	90
Jute	Glass	epoxy	56.68	-	28.81	1.83	5.49 J	Primarily different fibre ratio at 31:0, 25:7 and 18:19 were studied. Density of composites was 0.95- 1.14 g/cm^3	4
Jute	Cotton	Novolac	59.4	7.1	165.6	11.30	13 kJ/m^2	Layered and preferentially oriented laminated were prepared at fibre loadings of 17% to 40%	1
Jute	Hemp	Polyester	35% inc.	26% inc.	-	-	86% inc.	Sheet moulding compounding was used for fabrication of composites[#]	25
Oil palm EFB	Glass	Epoxy-vinyl ester	29.02	0.061	-	-	11.56 kJ/m^2	Results are based on statistical modelling. Density of samples lies between 1.08 g/cm^3 and 1.24 g/cm^3	58
Oil palm EFB	Jute	Epoxy	37.9	3.31	-	-	-	Fibre ratios of 4:1, 1:1 and 1:4 were studied maintaining total fibre loading of 40%. Thickness of composite samples was 4 -_5 mm	6
Oil palm EFB	Kenaf	Polyhydroxy butyrate	53.30	5.4	77.9	7.3	42.2 J/m	Laminated green composites using compression moulding were fabricated and the effect of stacking sequence on mechanical properties was determined	108
Oil palm	Clay	Polyethylene	11% inc.	49%inc.	-	-	-	Homogenous dispersion was achieved at 1:1 ratio of fibres by weight, at total loading of 25%. [#]	31
Hemp	Glass	Polypropylene	41.5	4130	73.3	4.6	32 J/m	Results were observed for total fibre loading of 40% wt.	23
Silk	Glass	Epoxy	-	-	114	5.44	-	3 pt. bending procedure was usedandglass to silk ratio was maintained at 1:1	54
Flax	Glass	Epoxy	10% dec.	-	-	-	25% dec.	10mm thick composite samples with total fibre loading of 31-56% were produced andfalling weight impact was studied[#]	57

(*Contd.*)

Table 5 — Summary of mechanical properties of hybrid biocomposites — Contd.

Fibre-1	Fibre-2	Matrix	Tensile strength MPa	Tensile modulus GPa	Flexural strength MPa	Flexural modulus GPa	Impact property	Remarks	Ref.
Sugar palm	Seaweed	Starch	17.2% inc.	9.3% inc.	-	-	8.4% dec.	Matrix contained Starch:agar:glycerol at 70:30:30. The resulting starch was thermoplastic in nature. Seaweed to SPF ratio was 1:3, 1:1 and 3:1 and density of samples ranged between 1.34 g/cm^3 and 1.36 g/cm^3. #	37

(-) Indicates that the corresponding mechanical tests were not conducted in the study.

Author did not quote absolute values but only the percentage increase compared to reference samples. Increase in values is denoted by "inc", and decrease is denoted by "dec".

From the data available in Table 5, we can conclude that the mechanical properties for hybrid biocomposites having glass fibres as one reinforcement, report relatively higher values than hybrid composites having both reinforcements as natural fibres/fillers. Most cases reported positive hybrid effect in mechanical properties after hybridisation.

5 Factors Influencing Properties of Hybrid Biocomposites

There are various parameters in hybrid biocomposites which have been investigated by the research community. The effect of these parameters on various properties of hybrid biocomposites, as discussed in the previous section, is determined and efforts are made to understand the mechanism and improve them. The effect of fibre loading, fibre length, orientation of fibres, layering pattern of laminates, degree of mixing of fibres and dispersion of fibres in the matrix are few of the common parameters whose effect on mechanical and other properties has been discussed by researchers. In this section, the studies using these parameters and their general findings will be discussed.

Fibre loading of composites is one of the most importantfactors which is primarily investigated for the targeted properties. In the case of hybrid composites,not only the total fibre volume fractionare discussed but also the ratio or content of two or more reinforcements present within the hybrid system discussed. The loading of various fibres has been mentioned in following two ways:

(i) In terms of total fibre loading along with the ratio of enforcements within it.

(ii) Fibre loading of each individual enforcement in terms oftotal composite fraction.

The former (i) way of mentioning the fibre loading has been widely used in studies. The effect of fibre loading has been discussed on mechanical properties, such as tensile, flexural, impact, dynamic mechanical properties, water absorption, thermal and sound absorption, and many more. It has been observed that tensile properties improve fibre loading subject to dispersion always follow a similar trend as tensile properties when the volume fraction of fibres is considered. of reinforcements and arrangement of laminates. Flexural and impacts properties do not Higher fibre loading leads to too many fibre ends that may initiate cracks[55].

However, it has to be noted that most of the studies are carried out with a fixed total fibre fraction and the observation in the end properties is made by varying the ratio of reinforcements within the fixed total fibre content. On several occasions, the reason for sticking to a particular loading has not been made explicit. It is possible that the generalisations made at one total volume fraction may not be true at another total fibre volume fraction. Only few elaborate studies are reported in the hybrid biocomposites, where effect of varying total fibre volume fraction as well as the varying ratio of reinforcements within them, are investigated.

Besides the volume fraction or fibre loading, there are other factors which are discussed in lesser detail. One of these factors is the effect of variation in fibre length on the properties of hybrid biocomposites. There are relatively fewer studies targeting the effect of fibre length or aspect ratio on properties of hybrid biocomposites. A study used sisal and glass of length varying from 1mm to 10mm. At length about 8mm, the mechanical properties started to decrease due to curling of sisal fibres and increased breakage of glass fibres. Tensile properties were found to be best at 6mm length of sisal and glass fibres[21]. Based on the results of tensile properties, the lengths of sea shell and oil palm fibres were optimized at 10 mm and 6 mm respectively in the sisal-oil palm fibre based natural rubber biocomposites[20].

The intermingling of fibres, degree of dispersion of fibres and the orientation of fibres within composites are other important factors which directly affect the end properties of composites. The layering pattern is linked with the degree of dispersion. The stress concentrations, transfer of load between fibres and deformation of structure all depend on the placement of fibres in the hybrid composites. There are broadly three configurations, and the literature available for hybrid biocomposites use any one of these three configurations[42]. The schematic representation is given in Fig 6. The simplest and most used configuration reported for hybrid biocomposites has been an interlayer structure which is denoted in Fig. 6(a). This configuration has been preferred by researchers, possibly due to the ease of processing different fibres during the composite fabrication process. Fig. 6(b) denotes the intralayer configuration where yarns or rovings may be present within the same layer but the fibres are still in their own bundles. The intimate mixing of fibres as denoted in Fig. 6(c) is rarely reported for hybrid biocomposites.

Effect of layering pattern of laminates on properties of hybrid biocomposites has been reported in few studies[7,41,80,99,104,115]. Hybrid biocomposites from coir - glass with different stacking sequence were studied. Trilayer composites with two adjacent layers of glass fibres were reported to have most superior properties in terms of tensile strength and tensile modulus. Bilayered composites displayed better flexural properties and maximum damping. Intimately mixed composition is not considered best for any static or dynamic mechanical property[113]. The layering pattern of two fibres as mentioned in this study is shown in Fig. 7.

Two different hybrid composites were prepared from hemp with glass and hemp with jute fibres. The layering pattern was the same in both the samples. Hemp was placed in the middle layer, whereas other fibre layers were placed on the top and bottom. Impact and tensile properties were highly dependent on the material which was placed at top and bottom layers of hybrid composites. The presence of stiffer fibres on top and bottom layers ensured protection of inner layers comprising of less strong fibres during regimes of impact loading and three point bending tests[24].

A study on glass-flax hybrid biocomposites investigated the effect of layering pattern on impact properties, and the mechanism of impact energy absorption was also discussed. It was mentioned that the symmetry of laminates was crucial in determining the best impact properties and the delamination contributed to absorption of impact energy. It is to be noted that impact properties are highly dependent on face or back stacking sequence when the drop weight impact tests are carried out instead of izod or charpy testing methods. Banana - sisal hybrid biocomposites displayed higher impact properties when bilayer arrangement of fibre layers was tested because sisal fibres have higher fracture toughness than banana fibres. Intimately mixed arrangement has better stress transfer than any of the laminated arrangement and thus displayed lowest impact strength. Flexural properties also displayed the same trend for laminated samples as impact properties, due to the presence of only one interlaminar plane present in bilayer arrangement as compared to two interlaminar planes in trilayer arrangement. The shearing during the flexural testing delaminates the structure and thus laminated structures with higher number of interlaminar planes are susceptible for failure during

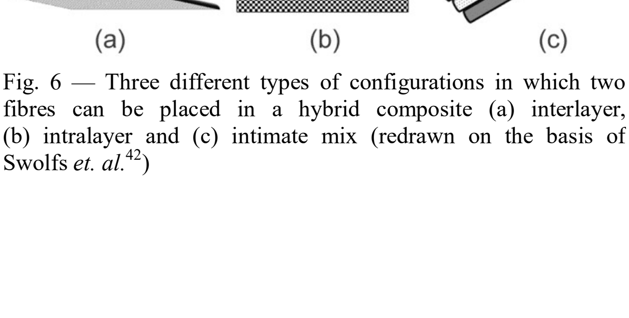

Fig. 6 — Three different types of configurations in which two fibres can be placed in a hybrid composite (a) interlayer, (b) intralayer and (c) intimate mix (redrawn on the basis of Swolfs *et. al.*[42])

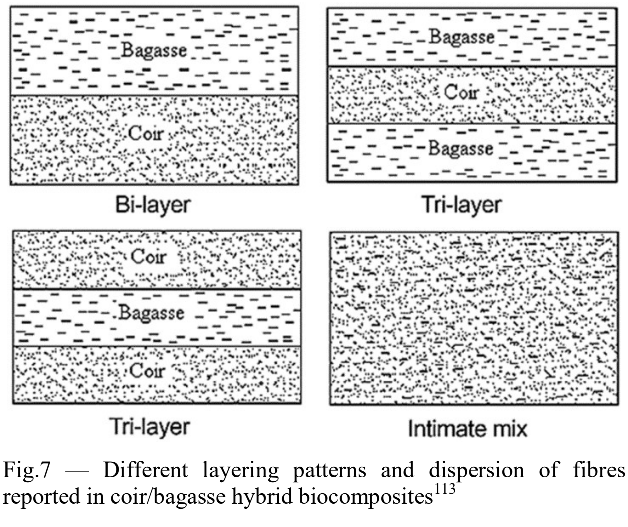

Fig.7 — Different layering patterns and dispersion of fibres reported in coir/bagasse hybrid biocomposites[113]

loading[7]. A study on laminated hybrid bilayer biocomposites investigated the effect of laminated arrangement on the hybrid effect in tensile strength and elongation at break. The effect of placement of laminates on the striking side of drop weight type impact tester and the mechanism of failure has also been discussed in detail[50].

It is observed that most of these parameters are not clearly mentioned in research papers for hybrid biocomposites, as they are in the case of hybrid composites. There are various important parameters such as intimate mix or layered one, orientation of fibres in 2D or 3D, the layering pattern, total fibre loading and the ratio of reinforcements within the total fibre loading etc. that affect hybrid effect. Therefore, these parameters should be clearly mentioned by authors, so that the studies can be understood in a better way. Laminates or layers used in hybrid biocomposites are usually mentioned as woven or nonwoven in most studies. Specifications such as weaving pattern, end/pick density, areal density, count of warp and weft, type of manufacturing techniques, and thickness for nonwovens have not been reported in most studies. It would be better if authors clearly mention details of parameters because the end properties are highly dependent on these factors. A summary of the stacking sequence, dispersion of fibres, fibre loading and orientation of fibres within the hybrid biocomposites is mentioned in Table 6.

It can be concluded from Table 6 that the dispersion of fibres varies widely within the structure of hybrid biocomposites. Both woven and nonwoven laminates have been reported and the effect of total fibre loading has been studied. The dispersion and placement of fibres within the structure finally affects the end properties of composite.

6 Hybrid Effect

The deviation of properties from expected behaviuor,onaddition of other reinforcement is termed as hybrid effect. It can be higher than expected, i.e. positive hybrid effect or negative hybrid effect which means lower than the expected theoretical values of these properties.

Tensile strength and tensile modulus both displayed negative hybrid effect at low as well as high volume fraction of OPEFB fibres in OPEFB -glass hybrid biocomposites. Only 0.74 volume fraction of OPEFB fibres displayed the tensile properties close to theoretical values predicted by parallel and Hirsh models. Positive hybrid effect in elongation was observed, whereas negative hybrid effect was observed in tensile properties. Lack of perfect intermingling of glass and OPEFB fibres which led to sandwich type structures was found responsible for diminished hybrid effect[49].

In another study, at total fibre loading of 40% and banana to sisal fibre ratio to be 4:1, it was found that tensile and flexural properties displayed positive hybrid effect, whereas impact strength showed negative hybrid effect[7]. Sisal-coirfibre reinforced natural rubber hybrid biocomposites displayed no negative or positive hybrid effect at lower total fibre loadings but at higher total fibre loadings, the positive hybrid effect was observed in tensile strength and Young's modulus of composites. It was mentioned that a better and uniform distribution of fibres at value of total fibre loading resulted in an almost ideal situation where fibres were uniformly distributedand the experimental values were close to the theoretical values. Further introduction of fibres led to closeness in the fibresand their accumulation in certain regions led to deviation from theoretical values[10]. Hybrid bilayer laminate biocomposites from glass and oil palm fibre reinforced epoxy resin displayed negative hybrid effect in tensile strength and Young's modulus[50]. It was stated that the laminate nature of composite led to negative deviation from the rule of hybrid mixture. The positive deviation in case of elongation at break was observed because oil palm fibre laminates underwent load bearing after the catastrophic failure of stronger and less extensible glass fibre laminates. Drop weight type impact tests were carried out and an increase in impact strength was noticed when the glass laminates were on the striking side. The impact strength was considerably lower when the oil palm fibre laminates were placed on the striking side. However, this observation was termed as positive hybrid effect in impact strength but there was no reference to the theoretically expected values. In a study, hybrid biocomposites consisting of laminates of carbon and flax displayed no hybrid effect in tensile strength while negative hybrid effect was observed for flexural strength. It was attributed to vast difference between the mechanical properties of constituting flax and carbon fibres[117].

All the above mentioned studies compared their experimental results with theoretically expected values, i.e. values expected from ROHM (Rule of

Hybrid Mixture) or any other theoretical model and the synergistic effect was discussed. One such result has been shown in Fig. 8, where effect of relative volume fraction of fibres on tensile modulus of hybrid composites has been reported. The experimental values of the composites are compared with theoretical values predicted by parallel and Hirsh model. The experimental values are lesser in comparison to the theoretical values showing a negative hybrid effect in tensile modulus.

However, there are few reports where researchers observed increment or decrement in properties when

Table 6 — Stacking sequence and dispersion of fibres within the hybrid biocomposites along with other parameters

Dispersion	Nature of reinforcement	Total fibre loading , %	Remarks	Ref.
Laminate	Woven	-	Effect of stacking sequence of laminates on mechanical properties was determined	61
Both intimate and layered	-	30	Dynamic mechanical properties with varying degree of fibre dispersion were studied	113
Laminate	Nonwoven	-	Machinability of hybrid biocomposites was studied using statistical modelling	62
Laminate	Nonwoven	-	A prepreg processwas proposed for the fabrication of composites	63
-	Nonwoven, Randomly oriented	-	The potential of coir fibres as sound insulation material in hybrid particle boards was investigated	46
Laminate	-	10 to 30	Effect of fibre ratio and total loading on mechanical properties was determined	8
Laminate	Woven	17	Effect of fibre and particulate loading on tensile and erosion behaviour was reported	45
Laminate	Nonwoven, Randomly oriented	20, 30, 40	The study was primarily based on fibre chemical treatmentsand itseffecton mechanical and dynamic mechanical properties	22
Laminate	Randomly oriented	20, 30, 40	Experimental results were compared to theoretical models and hybrid effect was discussed in detail	49
Laminate	-	20	Study was primarily based on chemical treatment and their effect on mechanical properties	53
Laminate	Random(Flax), Woven (glass)	-	Effect of stacking sequence of laminates on flexural properties was investigated in detail	41
Sandwich	-	25	Silk-glass based hybrid composites were studied and effect of glass fibres loading in water absorption of composites was reported	54
-	Randomly oriented	40	Effect of oil palm fibre loading on damping and storage modulus was studied	56
-	Randomly oriented	8 to 16	Laminated structures were studied for feasibility in housing panel applications	125
Intimate and laminate	Randomly oriented	40	The placement of laminates, dispersion of fibresand fibre loading positively affected the hybrid effect	7
Intimate	Randomly oriented	40	Intimately mixed fibres and their effect dynamic mechanical properties was reported	103
Intimate	Randomly oriented	-	Sheet molding compounds for housing application with various fibre combinations and fibrevolume fractionwas investigated	25
Intimate	Aligned	-	Effect of transverse and longitudinal placement of composite specimen on water uptake and mechanical properties was reported	20
Intimate	Aligned	-	Short and intimately mixed fibre reinforced hybrid composites were studied	109
Intimate	Randomly oriented	25	Hybrid biocomposites were analysed for mechanical and dynamic mechanical properties	34
Intimate	Randomly oriented	20	Different fibre loadings and mixing ratios were used as a parameter to observe tensile and impact properties	51
Partial mixing	Woven (Ramie-warp, Cotton-weft)	-	Fibre content in warp and weft directions of reinforcing fabrics and their layering patternswere investigated	119

(-)Indicates that the data is not reported in the study.

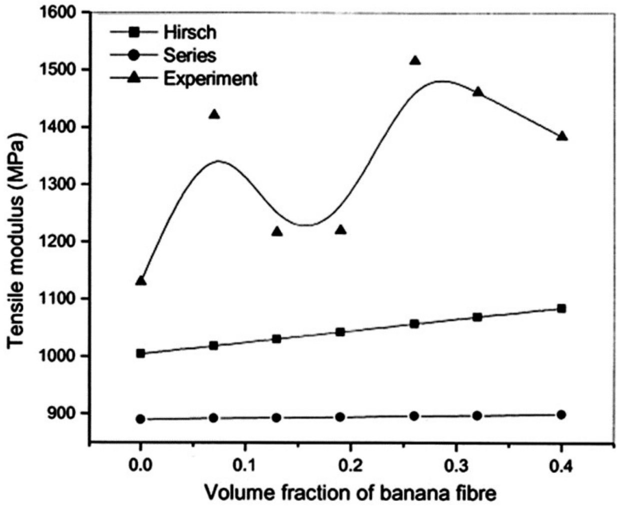

Fig. 8 — Experimental values of tensile modulus of sisal-banana fibre hybrid biocomposites alongwith theoretical models[7]

another fibreis introduced and have quoted this increase or decrease in performance as positive or negative hybrid effect. According to the authors, tensile, flexural and impact properties displayed positive hybrid effect on addition of glass fibres to the PALF-glass hybrid biocomposites[52]. Similarly, an improvement in tensile and flexural properties was observed after addition of glass fibres in glass-silk hybrid biocomposites and it was claimed that a positive hybrid effect was observed in these properties[54]. Alkali treatment of kenaf fibres in kenaf-glass-polyester resin hybrid biocomposites by 6% NaOH solution for three hours resulted in improved tensile modulus, flexural strength, flexural modulus and impact strength as compared to hybrids with untreated kenaf fibres. Thiswas termed as positive hybrid effect by the authors[38]. Another group reported that trilayer hybrid biocomposites having laminates of coir and bagasse displayed positive hybrid effect in tensile and flexural properties[113]. 5% NaOH was used to treat sisal and jute fibres for 30 min, and the resulting unidirectional bilayered hybrid biocomposite displayed an enhancement in tensile strength by 4.5%, flexural modulus by 2%, flexural strength by 16% and impact strength by 2% as compared with composites made of untreated fibres. Thiswas regarded as positive hybrid effect in mechanical properties arising due to alkali treatment of sisal and jute fibres[16]. Glass-kenaf fibre based hybrid biocomposites having totalfibre content of 40% and fibre ratio of 70:30 was claimed to have minor negative hybrid effect in compressive strength when compared to other samples with different fibre ratio[69]. The phenomenon of hybridisation has been discussed by Kalaprasad *et. al.*[21] with the layering pattern and intermingling of fibres. Hybrid biocomposites consisting of intimately mixed fibres displayed positive hybrid effect on tensile properties, while negative hybrid effect on flexural properties. On the other hand, flexural properties in laminated fibre arrangement displayed positive hybrid effect.

It is to be noted that in these studies, the improvement or degradation in mechanical properties on the addition of a particular fibre is mentioned as positive or negative hybrid effect in the reported property by the authors. Such enhancement or decrement in property may simply be mentioned as hybrid effect instead of positive or negative hybrid effect, unless the synergism in the experimental results is compared with theoretically expected behaviour.

7 Biodegradability

Biodegradability is an event/process in which chemical bonds are cleaved by the action of enzymes or living organisms, such as bacteria, fungi and their bio-secretions. There has been no worldwide accepted definition of biodegradable polymers. Even the standard test methods based on ASTM, ISO, etc. determine degradation in a specific environment at a given time, and classify samples accordingly [128].

There are few studies on use of activated sludge [128], compost material [129], standard test method ASTM D 5988-96 [130], ISO 14855:2017 [131], ISO 15985:2004 [131] and exposure to bacteria [132] to determine biodegradability of biocomposites. Apart from these, almost all the studies report soil burial methods to determine biodegradability of hybrid and non-hybrid biocomposites.

Several reports on biodegradability for jute[133], banana[134,135], kenaf[129,131,136], oil palm[137], rice husk[138], bamboo[130], and pineapple[130,139] fibre reinforced non-hybrid composites have reported using soil burial studies under different conditions of temperature, humidity and duration. Biodegradable polymers such as polylactic acid[129,132,134,138,140], starch[131], polybutylene succinate[133], polycaprolactone[134,140], and polyhydroxybutyrate[128,141] have been reported as matrix in these reports.

Degradation of biocomposites results in erosion of the surface and internal structure, leading to weight loss and deterioration in mechanical properties with duration of testing. Degradation in mechanical properties[139] and amount of CO_2 evolved[130] as a measure of biodegradability has been reported, but weight loss has been most widely studied[131–134,138,140,142,143] as a parameter to determine the biodegradability of biocomposites. Weight loss due to soil burial is determined using he following equation:

$$\text{Weight loss}\left(\%\right)=\left(\frac{Wi-Wd}{Wi}\right)\times 100 \quad \ldots (1)$$

where Wi is the weight of specimen before soil burial; and Wd, the weight of specimen after soil burial.

A study on rice husk and montmorillonite reinforced polyethylene films by Majeed *et. al.*[146] stated that increase in the rice husk content in the filler composition results in increased rate of degradation, as they are hydrophilic and more prone to microbial degradation than other filler. Instead of determining weight loss or deterioration in mechanical properties, surface inspection by electron microscopy and presence of holes in composite film were used as a measure to evaluate the degradation. Weight loss as well as deterioration in mechanical properties as a measure of degradation in filler reinforced hybrid composites has been reported[147,149]. A study on banana-kenaf reinforced hybrid biocomposites reports the degradation of composite samples in garbage, instead of soil [135].

Studies on biodegradability of non-hybrid biocomposites are limited but there are only a few studies conducted on biodegradability of biocomposites having two or more reinforcing phases in fibre or filler form. Some studies on hybrid biocomposites reinforced by kenaf[135,138], oil palm[144], sisal[144], flax[145], rice husk[146,147] and wood flour[147] have been reported and all of these report soil burial tests under different conditions. Chemical treatment of fibres which improve adhesion to matrix has been reported to slow down the process of degradation[144,148].

8 Conclusion

It can be concluded that the literature available on natural fibre or filler based hybrid composites, i.e. hybrid biocomposites, is very limited and this review discusses the studies on various aspects of composites in context of hybrid biocomposites. Hybrid biocomposites have been relatively less explored by the research community as compared to hybrid composites with no biodegradable component.

Natural lignocellulosic fibres are predominantly used as biodegradable reinforcement, although particulate fillers are also reported by few researchers. Most of the published work in the hybridisation of biocomposites used glass fibres as one of the reinforcements. It is observed that the incorporation of glass fibres improves mechanical properties of the final hybrid biocomposites. However, the use of glass fibres in biocomposites is in contradiction to the primary motive of using natural fibres in composites. It is well established that the natural fibresare used as reinforcement in composites because of biodegradability, less abrasion during processing, lightweight, etc. and are considered upon as a possible replacement for glass fibre based composites.

The incorporation of stronger fibre or filler in composite usually improves the overall mechanical properties, which is quite obvious. This strengthening effect, in case of hybrid biocomposites, is many times referred to as positive hybrid effect. Similarly, any deteriorating effect, due to addition of weak fibre or filler, is termed as negative hybrid effect for a particular property. It is quite obvious that incorporation of a stronger fibre may lead to enhancement of some mechanical properties. This enhancement should not be quoted as positive hybrid effect. The deviation of experimental results from the expected behaviour, as predicted by ROHM, is many times not referred to, in such studies. Further, there are less studies discussing the phenomena of hybridising effect for hybrid biocomposites. The focus of majority of studies is primarily on improving the mechanical property of biocomposites, by incorporating other fibres or fillers. The mechanism and effect of interactions between different fibres, on flexural and impact properties of hybrid biocomposites, has not yet been reported in detail, as in the case of tensile properties.

It is also observed that majority of studies have so far explored the effect of varying content of two or more fibres or fillers at a fixed total fibre loading, without justifying the reason for sticking to that particular total fibre loading. Hybrid biocomposites mostly use at least one cellulosic reinforcement which has polar surface and poor interface with non-polar matrix. Therefore, surface modification of cellulosic fibres improves interface by various chemical treatments. So far, only a few scientists working in field of hybrid biocomposites, have reported chemical treatment of natural fibres and fillers. Majority of the studies do not report surface treatment process for natural fibres or fillers in hybrid biocomposites. Mechanical properties, such as tensile, impact and flexural properties, are extensively explored, whereas reports on other properties like dynamic mechanical properties, water absorption behaviour, thermal insulation, etc. are reported only by few researchers. In hybrid biocomposites, izod impact testing and three-point bending are most popular techniques for investigation of impact and flexural properties, respectively.

Future Scope

Research in biocomposites is gaining thrust because of environmental concerns and single reinforcement based biocomposites currently face many challenges, due to inherent limitations of natural fibre/filler based reinforcements. Therefore, hybrid biocomposites are looked upon as a possible solution to overcome these challenges, by incorporating another reinforcement to overcome the limitations of single reinforcement based biocomposites.

The potential of hybrid biocomposites is yet to be fully explored. Many mechanisms and phenomena, which are extensively studied for hybrid composites, are relatively less explored in hybrid biocomposites. Pseudoductility, failure development, biodegradation behaviour and fatigue resistance, and the effect of parameters such as aspect ratio and degree of dispersion of fibres or particulate fillers, total fibre loadings, stacking sequence of laminates, manufacturing techniques, etc. may possibly be explored for natural fibre based hybrid composites.

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