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# Environmental degradation in biocomposites



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## s0010 7.1 Introduction

### s0015 7.1.1 Biocomposites

p0010 The emergence of biocomposites as an alternative to petroleum-based composites has largely been to superior technical properties along with problems associated with depleting petroleum resources and increasing environmental legislations. The biocomposite industry has found acceptance (mainly as nonload bearing structures) in the construction and automotive sector and reports suggest a projected growth rate of 22% per year [1]. Biocomposites—mainly containing natural reinforcements in a petroleum/biopolymer-based matrix possess many appealing characteristics such as high specific properties, lightweight, positive environmental impact, and biodegradable nature. However, the problems associated with biocomposites/bioplastics and in particular natural fibres are its degradation behaviour in the presence of moisture, temperature, sunlight, and microbial attack which restricts its increased use in industrial applications. Research on studies related to degradation behaviour of biocomposites in different environmental conditions is increasing indicating that it is currently a very relevant research topic.

p0015 Natural fibre reinforced composites are currently being used in the furniture, construction, and transport industry mainly in the interior and as nonstructural applications. Recent studies report on the development of natural fibre-based sandwich panels for use as side panels in aircrafts [2]. It is apparent that the use of NFC for exterior applications is not prevalent as they face a high risk of degradation when they are exposed to outdoor elements. Another issue that is limiting the use of biocomposites in industrial applications is that there are currently no international testing standards available for assessing the durability in biocomposites. In most cases, standards for wood and synthetic fibre reinforced composites are used for biocomposites which may not be technically appropriate. Moreover ageing studies for wood-based panels for use as interior panels does not exist and most accelerated ageing techniques make use of Arrhenius equation which only takes into account temperature variations and not humidity parameters and moisture content of natural fibres [3]. As a result most of the studies related to ageing of natural fibre composites use standards for wood-based materials with simplification of test methods. The ASTM D 1037

test on accelerated ageing consists of cycles of six treatment steps, i.e., immersion in water at 49°C for 1 h, steaming at 93°C for 3 h, freezing at -12°C for 20 h, drying at 99°C for 3 h, steaming at 93°C for 3 h, and drying at 99°C for 18 h. Several studies attempted the ageing by shortening the number of cycles [4] and eliminating certain ageing conditions [5].

p0020 The degradation behaviour is attributed to the inherent characteristics and chemical composition of natural fibres [6]. The degradation behaviour of natural fibres in composites can be related to structure and composition of natural fibres. Natural fibres comprise of cellulose, hemicellulose, lignin, and pectins. Cellulose is formed of cellulose microfibrils aligned along the length of the plant fibre and is hydrophilic. Hemicellulose comprises of a mixture of sugars and functions as a cementing material. Being hydrophilic hemicellulose can be easily hydrolysed by acids and bases.

p0025 Lignin is a mixture of complex aromatic and aliphatic hydrocarbons and gives rigidity to the plant structure. It is hydrophobic and is resistant to the attack of microorganisms. Pectins are mixture of complex polysaccharides that are present in the cell walls and nonwood parts of terrestrial plants. The function of pectin is to aid in cell wall extension and plant growth. In natural fibres, the responsible constituent for water uptake is hemicellulose. The presence of voids within the natural fibre structure also increases water absorption. As the content of hemicellulose increases, water uptake and rate of biodegradation in natural fibres increase. The component in fibres responsible for photodegradation is lignin. When biocomposites are exposed to sunlight, the ultraviolet radiation causes changes on the surfaces leading to colour fading and yellowing followed by deterioration in mechanical properties. It is the lignin present in natural fibres that mainly absorbs the UV light resulting in the formation of quinones and coloured chromophoric groups.

p0030 This chapter aims to review the different types of environmental degradation conditions that biocomposites can be exposed to. Recent studies dealing with mechanical and morphological data before and after ageing have been highlighted.

## s0020 **7.2 Types of environmental degradation**

p0035 The response of composite materials towards different environmental factors is different. The effect of environmental factors on the performance of composite materials must be well thought in the initial stages of design and tailoring of polymer composites [7]. The short-term and long-term response of composites towards environmental factors such as temperature, moisture, and biological attack can limit the usefulness of composites by retarding their mechanical properties during service. The main types of environmental degradation that will be reviewed in this chapter are:

u0010 • Temperature and moisture

p0045 Amongst all the different environmental factors, temperature and moisture are the most important and studies have revealed that when polymer composites are subjected to a combination of both temperature and moisture it results in a more aggressive and adverse effect

on the properties of composites. Composite materials can be exposed to cryogenic temperatures, elevated temperatures, and thermal cycling between these extremes. Natural fibres in particular are susceptible to both high temperature and moisture and are the primary points of attack followed by the interface and the matrix which exhibits transitions from glassy stage to a rubbery stage at its glass transition temperature.

u0015 • Weathering

p0055 Composites commonly used in automotive and aerospace applications are subjected to varying conditions of sunlight, rain, moisture, and humidity during their service life. The effect of weathering on properties of composites depends upon what type of materials has been used and on the presence of surface coating agents. As the effect of weathering needs to be assessed over a long period, test methods have been developed to simulate natural weathering conditions at an accelerated rate so that long-term weathering effects can be estimated in a shorter time. However, these types of accelerated weathering test results do not reflect accurate response as that of materials in actual use.

u0020 • Biological attack

p0065 The biological attack on polymer composite materials consists of fungal growth or marine fouling. Fungal growth is only possible in moist conditions and mechanical properties are affected after long-term exposure to micro-organisms. The degradation of polymer composites due to biological factors depends on the chemistry of the composites and the presence of acidic or basic entities. Natural fibres are prone to attack by microbes and promote the rate of degradation in composites. The effect of biological attack on biocomposites is usually analysed through soil burial tests and testing in a bioreactor.

p0070 The common methods to protect biocomposites from water uptake and weathering include the use of chemical treatments on natural fibres and/or resin and bio-based coatings on composite surfaces. A review of hygroscopic ageing on cellulosic fibres and biocomposites elaborates on the different techniques for providing moisture and weathering resistance for natural fibre composites [8]. Chemical treatments on natural fibres deal with acetylation and mercerization treatments where decline in water absorption is related to changes in physical and chemical morphologies of natural fibres. Bio-based coatings on composite panels comprise of silica and furanic coatings where the hydrophobicity of the coatings play an important role in imparting water resistant characteristics to the composites. A recent study on the use of furanic-based coating on flax fabric reinforced phenolic panels was undertaken by Mokhothu and John. Composite samples with and without coating were conditioned at 90°C and 90% R.H for 3 days and water sorption and strength properties were analysed. Samples coated with furans exhibited lower water uptake and improved strength retention properties after ageing [9]. In a similar study, the researchers focused on the use of external coatings on the biocomposite panels prepared from woven flax fabric and furan based resin. The coatings were found to have good adhesion with the substrate and exhibited good properties when compared to uncoated samples [10]. Another approach is the development of superhydrophobic surfaces using nanoparticle coatings. Researchers have explored the use of nanosilica-based coatings produced by sol-gel technique for cotton fabrics. Hydrophobic properties are induced on the textile substrates by reducing surface energy and formation of covalent bonds between hydroxyl groups of nanosilica and hydroxyl groups of natural fibres.

s0025 **7.3 Case studies**

s0030 **7.3.1 Effect of temperature and moisture**

p0075 Natural fibre composites are prone to moisture uptake when exposed to humid and wet conditions. This is mainly due to the hydrophilic nature of natural fibres imparted by the cellulose and hemicellulosic groups present in natural fibres. Water attaches onto the hydroxyl groups present in cellulose/hemicellulose and forms intermolecular hydrogen bonding with the fibres. This in turn reduces the interfacial adhesion between fibres and matrix resulting in debonding between the fibre and matrix and leads to impaired mechanical properties. The water absorption pattern in natural fibres has been reported to be 0.7%–2% after 24 h, 1.5% after a week, and upto 22% upon prolonged exposure (several months) [11]. The nature of the fibre, matrix, processing technique, and exposed surface area determine the water absorption capacity of composites.

p0080 The transport of water in polymer composites occurs via three main mechanisms:

p0085 Diffusion within the matrix

p0090 Diffusion through pores or cracks in the matrix

p0095 Capillary flow along the interface between fibre and matrix

p0100 In a study dealing with lignocellulosic fibres, flax fibres were exposed to a range of various temperatures from –40 to 140°C and relative humidities ranging between 25% and 85% in an environmental chamber. It was observed that the mechanical properties drastically decreased with hygrothermal ageing. Scanning electron micrographs revealed changes in flax fibre microstructures due to water uptake [12].

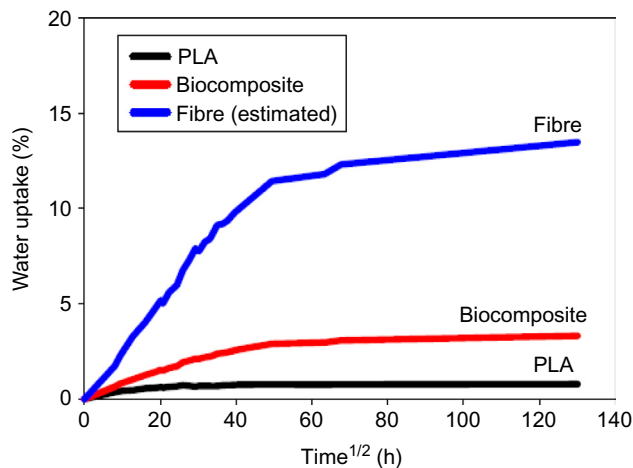
p0105 The behaviour of flax fibres and flax fibre reinforced polypropylene composites when exposed to moisture were investigated by Stamboulis et al. [13]. The authors chose two types of flax fibres: untreated green flax fibres and duralin flax fibres where flax fibres were subjected to steam or water-heating at temperatures above 160°C for 30 min in an autoclave. The fibres were placed in dessicators at different relative humidities followed by weighing the fibres at regular intervals. The authors placed the flax-PP composites in water for 60 days at room temperature and measured the water uptake. The moisture uptake and swelling of the duralin treated fibres was found to be 30% lower than that of untreated fibres. In the case of the composites, improved performance was observed for the composites containing duralin treated fibres. This was attributed to the formation of a water resistant system from the depolymerization of hemicellulose and lignin which cements the cellulose microfibrils.

p0110 Anuar et al. [14] investigated the degradability of kenaf fibre reinforced PLA biocomposites immersed in river water, tap water, and sea water. The water uptake of the samples was measured every day for 3 days. The authors made the general observation that water absorption generally depended on the amount of fibre content and water uptake increased with immersion time. In the present study, it was observed that the PLA biocomposite samples exhibited a higher water uptake when immersed in tap water.

p0115 In a similar study, the effects of long-term immersion of flax-PLA biocomposites in sea water were investigated by Le Dugiou et al. [15]. The injection moulded samples were immersed in sea water at a depth of 5 m for 2 years where temperatures ranged from 8°C to 19°C. The weight gain of the samples was periodically measured. Fig. 7.1



f0010 **Fig. 7.1** Biological development on the surface of Flax/PLA biocomposite.



f0015 **Fig. 7.2** Water uptake in PLA, flax fibre, and the biocomposite.

presents the biological development on the surface of the samples. The water uptake in the samples is shown in Fig. 7.2. It was observed that water uptake in biocomposites reaches plateau values of 3.3% within 2 months. The water uptake in fibres was theoretically calculated and the saturation weight value was found to be 12% which is in accordance with values obtained from dynamic vapour sorption [16] and gravimetric methods [8].

p0120 In a similar study, Krasowska et al. [17] investigated the degradation behaviour of two types of biocomposites in Baltic sea water. The two systems chosen were: ramie reinforced Ecoflex biocomposites and ramie fibre reinforced corn starch resin which contained cellulose nanofibers. The samples were immersed in a perforated basket and placed at a depth of 2 m in the Baltic sea. The authors observed that ramie reinforced

Ecoflex biocomposites degraded at a faster rate than the biocomposites containing cellulose nanofibres. The authors also carried out experiments in compost and observed that degradation was faster in compost when compared to sea water as the former provided an ideal environment for the growth of fungi.

p0125 Dixit et al. [18] investigated the influence of water sorption on coir fibre-epoxy composites containing fillers of cow dung, wheat husk, and rice husk. The authors immersed samples in water at 100°C for 24 h. Samples were also placed in water at room temperature for a week and the resulting changes in tensile properties were monitored. The first observation was that reduction in mechanical properties (tensile, flexural, and impact) was higher when samples were submerged in water at 100°C. Amongst all the fillers, it was observed that tensile strength decreased the maximum (60%–90%) in the case of composites containing cow dung while a reduction in the range of 30%–70% was observed for wheat husk and rice husk containing composites. The same trend was observed for impact strength with cow dung containing composites registering the maximum decrease (65%) compared to wheat husk and rice husk samples. Immersion experiments at 100°C resulted in a maximum decrease of 94% for cow dung containing composites. This was attributed to the presence of nitrocellulose in cow-dung which being hydrophilic promoted water uptake.

p0130 The effect of immersion of kenaf fibre reinforced polyester composites in different types of water solutions for 260 days was investigated by Nosbi et al. [19]. In this particular case, samples were prepared by pultrusion technique and solutions chosen were distilled water, sea water, and acidic solution at ambient temperature. The weight change of the samples was regularly monitored. The authors observed that irrespective of the type of solution, all the samples exhibited a Fickian water absorption pattern. The samples immersed in distilled water recorded the maximum moisture uptake as there was no interference to the diffusion path followed by water molecules while in acidic and sea water, the presence of acidic ions blocked the diffusion of water into the biocomposites. Samples immersed in distilled water registered the minimum decrease in compressive strength.

p0135 Polypropylene composites containing wheat flour and saw dust were immersed in brine solution and water for a period of 15 weeks and the resulting surface and mechanical properties were evaluated [20]. The authors observed that samples immersed in the brine solution exhibited maximum crazing, discolouration, and reduction in mechanical properties. This was attributed to the presence of chloride ions affecting the interfacial strength in composites. Another observation was that composites containing sawdust exhibited greater reduction in mechanical properties and higher moisture uptake. The authors attributed this to greater hydrophilic character of sawdust, however the particle size of the fillers would also have played a role.

p0140 The effect of chemical treatment of natural fibres and fillers on the water sorption has been investigated extensively. It has been noted that several chemical modification techniques such as—alkali treatment, acetylation, and cyanoethylation—have proved to reduce the water uptake in biocomposites during short-term immersion experiments. A detailed review on the hygroscopic ageing in lignocellulosic fibres and its composites has been documented by Mokhothu and John [21]. The authors are of the view that a higher level of hydrophobicity needs to be introduced to natural fibres so



that they can withstand long-term environmental ageing. These include the use of bio-based coatings and development of superhydrophobic surfaces.

s0035 **7.3.2 Effect of weathering**

p0145 An in-depth review on the degradation behaviour of natural fibre composites under various conditions was documented by Azwa et al. [5]. The exposure of biocomposites to sunlight results in colour and weight loss and deterioration of mechanical properties which occurs due to degradation of fibres and matrix. In the case of biocomposites, both natural fibres and the polymer matrix absorb the ultraviolet rays from the sunlight. This leads to changes in the chemical structure of the polymers via molecular chain scission, surface oxidation, and breakdown of molecules to form active radicals. The main degradation processes occurring by weathering includes photoradiation, thermal degradation, photooxidation, and hydrolysis [22,23]. The effect of sunlight and oxygen on the polymers causes density gradients leading to stresses and in combination with macromolecular chain scission results in propagation of cracks. These cracks cause light diffusion leading to a whitening effect in appearance and an adverse effect on mechanical properties [24].

p0150 The lignin present in natural fibres absorbs ultraviolet radiation resulting in the formation of chromophoric groups, quinones, and hydro-peroxy radicals that cause the yellow colour associated with ageing of polymers. In the case of wood fibre reinforced polymer composites, it was observed that upon UV exposure, two competing reactions occurred namely, formation of paraquinone chromophoric structures generated by oxidation of lignin leading to yellowing and reduction of paraquinone structures to hydroquinones leading to photobleaching [25].

p0155 The photodegradation of natural fibre reinforced polymer composites depends on various factors such as amount of fibre, presence of compatibilizers, processing techniques, and weathering conditions. Generally it has been observed that oxidation rate of the composites increases with fibre content and decreases with the presence of compatibilizers. The presence of moisture along with ultraviolet radiation results in greater loss of mechanical properties and colour change. Processing techniques also affect the rate of degradation and it has been seen that biocomposites manufactured by extrusion were also observed to degrade more than injection moulded composites due to the presence of a surface layer of the polymer matrix.

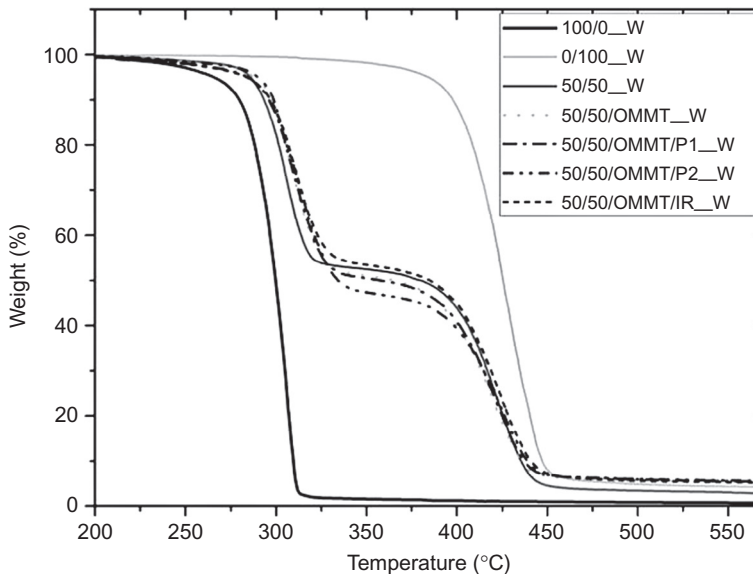
p0160 Natural fibre composites can be subjected to natural weathering conditions which relates to the action of the environment in which the material is subjected to during its service life. The behaviour of materials subjected to environmental exposure is evaluated over a duration of several years. Ageing of composites can also be studied by accelerated weathering studies where samples are placed in an ageing chamber that simulates a natural environment condition and exposes the samples to ultraviolet radiation, heat, and moisture in a controlled manner.

p0165 Accelerated weathering studies were conducted on banana fibre reinforced PLA biocomposites in an Xenon Arc Weatherometer [26]. The specimens were subjected to specified weathering cycles of light and darkness [3.8 h light and 50% relative humidity, 1 h dark and 95% RH at 38 black panel temperature]. The samples were taken out

at intervals and tested for tensile and impact properties. The main observations were that after weathering the tensile strength of PLA and PLA biocomposites decreased by 45% and 53%, respectively. However it was observed that composites containing nanoclay exhibited lower reduction rate in tensile strength. This was attributed to the fact that nanoclay penetrates into the microvoids at the PLA-banana fibre interface and prevents the weakening of the interface. A similar trend was observed in the case of impact strength. The surface of the biocomposites also exhibited blisters after weathering while the composite containing nanoclay displayed microcracks.

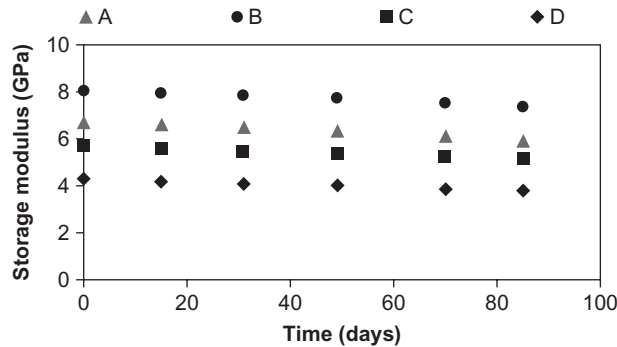
p0170

In another study [27], poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate)/poly(butylene adipate-*co*-3-terephthalate) (PHBV/PBAT) biocomposites containing nanoclay were subjected to alternating cycles of UV light, rain, and dryness according to ISO standards. After weathering the samples were tested for thermal stability and changes in colour. The authors observed that all the samples [100/0: PHBV/0, 0/100:0/PBAT, 50/50: 50PHBV/50PBAT, 50/50/OMMT: 50PHBV/50PBAT/3Organommodified montmorillonite, 50/50/OMMT/P1: 50PHBV/50PBAT/3Organommodified montmorillonite/Propolis siccum extract, 50/50/OMMT/P2: 50PHBV/50PBAT/3Organommodified montmorillonite/Propolis powder with SiO<sub>2</sub>, 50/50/OMMT/IR: 50PHBV/50PBAT/3Organommodified montmorillonite/Irgaguard B1315 & F3000] registered a decrease in thermal stability after weathering (Fig. 7.3). Certain samples containing antimicrobial agents (propolis) showed a reduced degradation during weathering. Regarding colour change, most of the samples exhibited a slight decline in brightness and colour, however samples containing propolis changed from brown colour to a neutral shade.



f0020

Fig. 7.3 TGA results of specimens after weathering test.



f0025 **Fig. 7.4** Change in storage modulus over time for biocomposites (A) untreated hemp mat-unsaturated polyester resin (UPE), (B) acrylonitrile treated hemp mat-UPE, (C) silane-treated bi blue steam grass-CaCO<sub>3</sub>, and (D) silan -treated BBSG and green flax core-CaCO<sub>3</sub>-UPE.

p0175 Information on durability behaviour is very important for use in certain applications such as the building sector. A number of studies on weathering behaviour of wood-based composite samples have been documented in literature [28,29], Mehta et al. [30] investigated the long-term effects of weathering on biocomposites (hemp fibre reinforced unsaturated polyester) used as building components. The samples were exposed to ultraviolet radiation for 2016 h. The authors observed that as the exposure time of the samples to weathering conditions increased, storage modulus decreased (Fig. 7.4) and the surface roughness of the samples also increased.

p0180 Acrylonitrile butadiene styrene (ABS) samples containing four different types of fillers [carbon-black-filled, sunflower hull (SFH) and distillers' dried grains with solubles (DDGS)] were subjected to weathering under ultraviolet (UV)/condensation conditioning [31]. The authors observed that impact properties of the ABS samples containing SFH and DDGS exhibited higher property retention in comparison to neat ABS.

p0185 In a study focusing on protein-based composites, silk fibre reinforced gelatin biocomposites were subjected to alternating cycles of sunshine and condensation for a period of 60 h; the authors observed a loss of tensile strength of about 70% for composites containing 30% silk fibre [32].

p0190 Researchers suggest that it is difficult to correlate data acquired from accelerated weathering experiments with natural weathering process. Though an understanding of the basic degradation mechanisms of composites is obtained, it is crucial to run natural weathering tests to obtain accurate data.

### s0040 7.3.3 Effect of biological attack

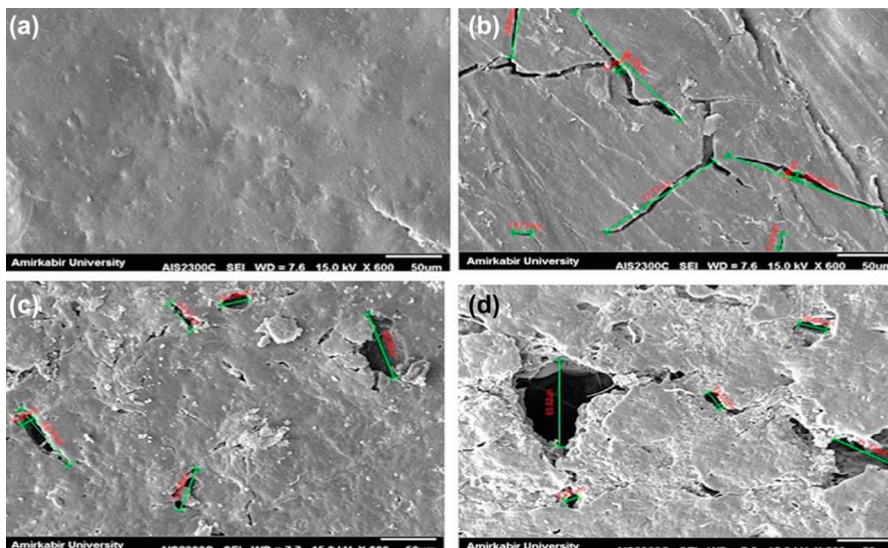
p0195 Several studies measure the biodegradation in biocomposites by means of soil burial test and testing in a bioreactor. Soil burial test comprises of placing samples in soil/compost for long durations and testing the mechanical properties/dimensional changes/morphology before and after soil burial. In a bioreactor, samples are placed in a composting vessel containing a mixture of compost and the percentage of biodegradation

is theoretically calculated by measuring the amount of CO<sub>2</sub> evolved from the composting vessel for a period of 45 days.

p0200 Silk fibre reinforced gelatine composites were subjected to soil burial for days and mechanical properties determined before and after burial. It was found that the tensile properties underwent a drastic reduction in properties with time. Another observation was that the gelatin matrix degraded from the composites within 24h. This was attributed to the attack of microorganisms present in soil on gelatine and silk which are both natural polymers.

p0205 In another study [33], PLA composites containing oil palm empty fruit bunch fibre was compounded with a slow releasing fertilizer and was subjected to soil burial tests at a temperature of 30°C and relative humidity of 80%. The samples were recovered at different stages of degradation and weighed to ascertain the mass loss during soil burial. The surfaces of the samples were also analysed using scanning electron microscopy. The biodegradation rate of the samples containing fibres and fertilizer was found to be lower than that of neat PLA. The scanning electron micrographs depicted the changes that occurred during the degradation period. The surface of the composite samples exhibited traces of shrinkage and roughness and exposed the natural fibre bundles. The scanning electron micrographs also revealed the presence of cracks and holes which were produced by the degradation of oil palm fibres (Fig. 7.5).

p0210 Soil burial test of biocomposites from wheat gluten and rubber wood sawdust were carried out by Bootklad et al. [34]. Compression moulded samples were buried in soil for 15 and 30 days and the subsequent weight loss was measured. The authors observed that this type of green biocomposites could be degraded within 15 days. During



f0030 **Fig. 7.5** Scanning electron micrographs, (A) PLA10/NPK15/EFB15 (0 weeks), (B) PLA10/NPK15/EFB15 (2 weeks), (C) PLA10/NPK15/EFB15 (4 weeks), and (D) PLA10/NPK15/EFB15 (8 weeks).



the first 15 days, the weight loss was attributed to the leaching of glycerol which was used as a plasticizer in the system. The authors also observed that the biodegradation rate of composites containing 20 weight percent of rubber-wood waste was slower than that of wheat gluten biocomposites.

p0215 In another study, Pantyukhov [35] investigated the biodegradation behaviour of a range of lignocellulosic filler reinforced low-density polyethylene composites. The lignocellulosic fillers included flax shives, sunflower husk, hay, birch leaves, and banana skin. Soil mixture comprising of sand, garden soil, and horse manure were prepared and samples were placed in the soil for a period of 1 year. The authors observed the greatest weight loss was in the case of hay filled composites followed by lignosulfonate, husk, banana, leaves, and shives. This was attributed to the chemical, fractional, and particle size composition of the fillers. It is generally known that fibrous particles have a larger surface area than spherical particles and in that respect, hay particles having a fibrous structure would undergo biodegradation at a faster rate and hence register the higher mass loss.

p0220 In an interesting study, samples of okra fibre reinforced corn starch composites were immersed in soil containing a variety of microbial species. The weights of the samples were monitored every week. The authors observed that increasing the weight of the okra fibres decreased the rate of biodegradation; this was attributed to the slower degradation of okra fibres when compared to corn starch matrix. Changes in morphology were also observed and a highly rough and heterogeneous surface for the corn starch matrix was observed [36].

p0225 The effect of different additives on the biodegradation of flax fibre reinforced polylactic acid composites was investigated Kumar et al. [37]. The samples were subjected to soil burial test for long periods of time. The authors observed that biodegradation occurred at a faster rate in presence of mandelic acid (which was used as a compatibilizer) while degradation slowed down in presence of dicumyl peroxide. This was attributed to the amphiphilic characteristics of the mandelic acid. Another interesting observation was that flax composites containing nonwoven flax biodegraded at a faster rate than those samples containing woven flax.




p0230 In a study involving the use of bioreactor, the biodegradation in polycaprolactone (PCL)/eggshell (ES) biocomposite (50/50, w/w) was studied by Gonzalez Petit et al. [38] for 8 weeks, where several parameters such as temperature, moisture, soil pH, light, and anaerobic condition were monitored during composting. The morphological changes observed in scanning electron micrographs showed that the degradation process occurred mainly on the surfaces of biocomposite samples' surface. A higher weight loss and degradation was registered for the PCL/ES samples than for pure PCL. It was observed that soil pH, heat, and an aerobic environment accelerated the degradation process, while a photo-controlled, anaerobic, and moisture-saturated environment reduced microbial activity and delayed the biodegradation process.






p0235 Similar studies were performed by Muniyasamy et al. [39] on green composites from poly(butylene-co-adipate terephthalate) and distillers dry grains with solubles (DDGS). The authors observed that the presence of DDGS increased the rate of degradation of biocomposites as DDGS was the preferential point of attack by microorganisms.

s0045 **7.4 Conclusions**

p0240 This chapter analyses the performance characteristics of natural fibre reinforced composites under various environmental conditions. The different environment factors covered are exposure to moisture, temperature, and humidity along with weathering elements. The response of natural fibres to various weathering elements depends on the chemical composition of the fibres. Of all the environmental elements, combined exposure to moisture and temperature were found to have a drastic decline in the performance of biocomposites. This is attributed to the absorption of moisture by hydrophilic natural fibres resulting in fibre debonding from the matrix leading to a weak interface. The long-term exposure of biocomposites to sunlight results in absorption of ultraviolet radiation leading to colour change, surface roughening, and mechanical property deterioration which is further aggravated in the presence of moisture. The presence of natural fibres increases chances of biological attack (bacterial and fungal attack) and accelerates the degradation of biocomposite materials.

bi0010 **References**

- bb0010 [1] Satyanarayana KG, Arizaga GGC, Wypuch F. Biodegradable composites based on lignocellulosic fibers – an overview. *Prog Polym Sci* 2009;34:982–1021.
- bb0015 [2] Anandjiwala R, Chapple S, John M, Schelling H-J, Doecker M, Schoke B. A flame proofed artefact and a method of manufacture thereof; 2013. WO2013/084023.
- bb0020 [3] Zandvliet C, Bandyopadhyay NR, Ray D. Proposition of an Accelerated Ageing Method for Natural Fibre/Polylactic acid composite. *J Inst Eng India Ser* 2015;96:151–8.
- bb0025 [4] Okkonen A, River BH. Outdoor aging of wood-based panels and correlation with laboratory aging: part 2, forest products research society. *For Prod J* 1996;46:68–74.
- bb0030 [5] McNatt JD, Forest CL. Analysis of ASTM D1037 accelerated-aging test products research society. *For Prod J* 1989;39:51–7.
- bb0035 [6] John MJ, Thomas S. Biofibres and Biocomposites. *Carbohydr Polym* 2008;71(3):343–64.
- bb0040 [7] Jayaram SH. Impingement of environmental factors that defines a system on composites performance, <http://www.engineeringcivil.com/impingement-of-environmental-factors-that-defines-a-system-on-composites-performance.html>. 
- bb0045 [8] Celinot A, Freour S, Jacquemin F, Casari P. Characterization and modelling of moisture diffusion behaviour of natural fibres. *J Appl Polym Sci*, doi:10.1002/APP.39148. 
- bb0050 [9] Mokhothu TH, John MJ. Bio-based coatings for reducing water sorption on natural fibre reinforced composites. In: Proc. Of 13th annual UNESCO/IUPAC workshop and conference on functional polymeric materials & composites, 7-10 September, South Africa, 2015; 2015.
- bb0055 [10] <http://osirysproject.eu/biocomposites-with-good-outdoor-durability/>.
- bb0060 [11] Azwa ZN, Yousif BF, Manalo AC, Karunasena W. A review on the degradability of polymeric composites based on natural fibres. *Mater Des* 2013;47:424–42.
- bb0065 [12] Thuault A, Eve S, Blond D, Breard J, Gomina M. Effects of the hygrothermal environment on the mechanical properties of flax fibres. *J Compos Mater* 2014;48(14):1699–707.
- bb0070 [13] Stamboulis A, Baillie CA, Garkhail S, Van Melick HGH, Peijs T. Environmental Durability of Flax Fibres and their Composites based on Polypropylene Matrix. *Appl Compos Mater* 2000;7:264–73. 

- bb0075 [14] Anouar H, Zuraida A, Fuad F. Biodegradable PLA-Kenaf fibre biocomposite for cleaner environment, [http://irep.iium.edu.my/845/1/Manuscript\\_Hazleen.pdf](http://irep.iium.edu.my/845/1/Manuscript_Hazleen.pdf). 
- bb0080 [15] Le Duigou A, Bourmaud A, Davies P, Baley C. Long term immersion in natural seawater of Flax/PLA biocomposite. *Ocean Eng* 2014;90:140–8.
- bb0085 [16] Hill C, Norton G, Newman G. The water vapor sorption behavior of natural fibers. *J Appl Polym Sci* 2009;112:1524–37.
- bb0090 [17] Krasowska K, Brzeska J, Rutkowska M, Janik H, Sreekala MS, Goda K, et al.  Environmental degradation of ramie fibre reinforced biocomposites. *Pol J Environ Stud* 2010;19:937–45.
- bb0095 [18] Dixit S. Degradation analysis of lignocellulosic fillers infused coir epoxy composites in different environmental conditions. *International Journal of Lignocellulosic Products* 2014;1:160–79.
- bb0100 [19] Nosbi N, Akil HM, Ishak ZAM, Bakar AA. Degradation of compressive properties of pultruded kenaf fibre reinforced composites after immersion in various solutions. *Mater Des* 2010;31:4960–4.
- bb0105 [20] Fakhru T, Islam MA. Degradation behaviour of natural fibre reinforced polymer composites. *Procedia Engineering* 2013;56:795–800.
- bb0110 [21] Mokothu T, John MJ. Hygroscopic aging in cellulosic fibres and biocomposites. *Carbohydr Polym* 2015;131:337–54.
- bb0115 [22] Beg MDH, Pickering AL. Accelerated weathering of unbleached and bleached kraft wood fibre reinforced polypropylene composites. *Polym Degrad Stab* 2011;96:97–106.
- bb0120 [23] Wang W, Sain M, Cooper PA. Hygrothermal weathering of rice hull/HDPE composites under extreme climatic conditions. *Polym Degrad Stab* 2005;90:540–5.
- bb0125 [24] Fabiyi JS, McDonald AG, Wolcott AP, Griffiths PR. Wood plastic composites weathering: visual appearance and chemical changes. *Polym Degrad Stab* 2008;93:1405–14. 
- bb0130 [25] Matuana LM, Jin S, Stark NM. Ultraviolet weathering of HDPE/wood flour composites coextruded with a clear HDPE cpa layer. *Polym Degrad Stab* 2011;96:97–106. 
- bb0135 [26] Sajna VP, Nayak SK, Mohanty S. Weathering and Biodegradation Study on Graft Copolymer Compatibilized Hybrid Bionanocomposites of Poly (lactic acid). *J MATER ENG PERFORM* 2016;25:2895 2016–.
- bb0140 [27] Bittmann B, Bouza R, Barral L, Bellas R, Cid A. Effect of Environmental Factors on Poly(3-hydroxybutyrate-co-3-hydroxyvalerate)/Poly(butylene adipate-co-terephthalate)/Montmorillonite Nanocomposites with Antimicrobial Agents. *Polymer Composites*, 2016.
- bb0145 [28] Podgorski L, Arnold M, Hora G. *Coatings World*, February 39-49; 2003.
- bb0150 [29] Nzokou P, Kamdem DP. *J Top Fpr Prod* 2002;2:200–9.
- bb0155 [30] Mehta G, Mohanty AK, Drzal LT, Kamdem DP, Misra M. Effect of Accelerated Weathering on Biocomposites Processed by SMC and Compression Molding. *J Polym Environ* 2006;14:359–68.
- bb0160 [31] Chevali Venkata S, Nerenz Brent A, Ulven Chad A. Acrylonitrile Butadiene Styrene (ABS)/Lignocellulosic Fiber Biocomposite: Effect of Artificial Weathering on Impact Properties. *J BIOBASED MATER BIO* 2012;4:42–50.
- bb0165 [32] Shubra QTH, Alam AKMM, Saha M, Saha D, Khan JA, Quaiyyum MA. The Preparation and Characterization of Silk/Gelatin Biocomposites. *Polym-Plast Technol Eng* 2010;49:983–90.
- bb0170 [33] Harmaen AS, Khalina A, Azowa I, Hassan MA, Tarmian A, Jawaid M. *Polymer Composites* 576-583; 2015.
- bb0175 [34] Bootklad M, Chantarak S, Kaewtatip K, Novel biocomposites based on wheat gluten and rubber wood sawdust. *J Appl Polym Sci*, doi:10.1002/APP.437052016. 

- bb0180 [35] Pantyukhov P, Kolesnikova N, Popov A. Preparation. Polymer Composites: Structure and Properties of Biocomposites Based on Low-Density Polyethylene and Lignocellulosic Fillers; 2016.
- bb0185 [36] Ghulera A, Singha AS, Rana RK. Mechanical, Thermal, Morphological, and Biodegradable Studies of Okra Cellulosic Fiber Reinforced Starch-Based Biocomposites. *Adv Polym Technol* 2016.
- bb0190 [37] Kumar R, Yakabu MK, Anandjiwala RD. Biodegradation of flax fibre reinforced polylactic acid. *Express Polym Lett* 2010;4:423–30.
- bb0195 [38] González Petit M, Correa Z, Sabino MA. Degradation of a Polycaprolactone/Eggshell Biocomposite in a Bioreactor. *J Polym Environ* 2015;23:11–20. <http://dx.doi.org/10.1007/s10924-014-0655-x>.
- bb0200 [39] Muniyasamy S, Reddy MR, Misra M, Mohanty A. Biodegradable green composites from bioethanol co-product and poly(butylene adipate-co- terephthalate). *Ind Crop Prod* 2013;43:12–819.



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## **Non-Print Items**

### **Abstract**

The aim of this chapter is to discuss the environmental degradation behaviour in biocomposites. As the economic advantages of weight reduction have become mandatory for many advanced industries, bio-based composites have emerged as preferred candidates as they possess interesting properties like high mechanical properties along with low weight. This chapter presents the background on biocomposites, highlighting the important advantages and problems associated with natural fibre reinforced composites. The different types of environmental degradation conditions ranging from exposure to moisture, temperature, and humidity along with weathering elements are presented. Case studies dealing with the investigation of the effect of environmental degradation on the properties of biocomposites have been discussed.

**Keywords:** Environmental degradation, Weathering, Natural fibres, Biocomposites.

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