Aging Effects on Mechanical Properties of Biocomposites with Recycled Polymers

Md. Sarif Sakaeyt Hosen, Md Enamul Hoque, Md. Zillur Rahman, and Suresh Sagadevan

Abstract This chapter discusses the aging effects (e.g., photochemical, hydrothermal, natural, thermal, freeze–thaw cycling, and xenon-arc light) on various mechanical characteristics of biocomposites with the recycled polymer. In general, biocomposites show a decrease in tensile strength after photochemical aging, but tensile modulus and strain to failure do not vary significantly with increasing aging time. In contrast, tensile modulus and ultimate tensile strength of biocomposites reduce significantly with hydrothermal aging, but the strain to failure increases. Also, flexural modulus, tensile modulus, tensile strength at break, and elongation at break increase after natural aging. Furthermore, there is an increased tensile modulus and strength of biocomposite under accelerated thermal aging. The process and result of aging depend on the type of biocomposites. As the use of biocomposites is widely increasing, this chapter can be useful for getting insight into the durability performance of the biocomposite materials.

Keywords Aging effect · Biocomposites · Hydrothermal aging · Mechanical properties · Natural aging · Recycled polymer · Thermal aging

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1 Introduction

1.1 Biocomposites with Recycled Polymers

A biocomposite with recycled polymer is a composite material made by a matrix (polymer) and a reinforcement of natural fibers. Polymers are derived from renewable and nonrenewable resources. Polymers are two types: thermoplastic and thermosetting polymers. Thermoplastic polymers can be easily recycled, whereas thermosetting polymers are much more difficult to recycle. Thermoplastic polymers are polyethylene, cellulose acetate, polyvinylchloride, polystyrene, polypropylene, and polycarbonate. Natural fibers are derived from different plant and animal sources (AL-Oqla et al. 2015; Arifur Rahman et al. 2015). Some examples of natural fibers are cellulose-based cotton, jute, and flax. The animal fibers are protein-based, which include wool and silk. In this chapter, the focus is on the recycled polymer matrix (Salit et al. 2015). Natural fiber-reinforced polymer matrix composites can be cheaper, tougher, and environmentally friendly (Asim et al. 2015).

Like woods (Hoque et al 2016), when polymer matrix composites (PMC) are exposed for a long time to the environment their characteristics change irreversibly from the materials' original characteristics. The process in which characteristics change over time in polymer matrix composites can be termed as 'aging'. Composites' stiffness, strength, and fatigue life are time-dependent changes. These can all be related to changes in the mechanical characteristics of the polymer matrix. Aging processes can be categorized into three type's chemical, physical, and mechanical. All three aging processes depend on material type, the environment, and the mechanical loads (Enamul Hoque et al. 2014). However, the aging of biocomposites with recycled polymers makes uncertainties about the long-term performance of composites under conditions encountered in service (Martin 2008).

1.2 Significance of Aging

A matrix material experiences environmental condition which alters the characteristics of the matrix or mechanical loads that act over a lengthy time. Composite performance depends mainly on the viscoelastic nature of the matrix. As stiffness, strength, and fatigue life of composites vary over time, the mechanical characteristics of the polymer matrix also change. An aging study is generally conducted to know the mechanical, physical, and morphological changes of the composites exposed to the application environment. This study also helps to estimate the durability of the composites, thereby choosing the optimum material for a particular application (Stark 2006). The durability of a material can be known fully by long-term (real-time) testing. It also filters admissible postulant materials by aging then the long-term performance test can run. As a result, the expense may reduce involvement in confining the field of admissible postulant substances. In addition to materials

filtering, rapid testing provides information about the remaining service life of an existing structure and provide a suggestion for betterment (Brebu 2020). The decision for methods of material selection can be made by aging. This also gives a fundamental principle for manufacturers to adopt new and advanced specific methods.

2 Aging of Biocomposites with Recycled Polymer

2.1 Important Terms of Aging

Environmental deterioration factor is the general term for specific application conditions, which affects every material. Some examples of environmental deterioration factors are mechanical loads, moisture, heat, etc. Environmental deterioration of polymeric composites can occur by chemical, physical, and biological processes or a combination of them under different factors such as mechanical stress, temperature, moisture, light, air, high-energy radiation, and chemical agents. Variations in the material occur by these factors in two steps. Firstly, deterioration of appearance, physical and morphological characteristics or mechanical characteristics happen, and then the changes are converted in the form of H_2O, C_2O, and other simple inorganic compounds (mineralization).

Polymer materials are affected by environmental deterioration factors, including chemical, physical, and mechanical processes. When one or more physical characteristics of a material system change due to the environmental factors attack, then the critical deterioration process occurs. However, environmental deterioration factors should be inside the boundaries of the service environment to occur a critical deterioration process (Martin 2008).

The accelerated aging process of polymer matrix materials was performed simulating outdoor conditions (González-López et al. 2020; Seldén et al. 2004). The water spraying cycle and UV light incidence were carried out under temperature and emission. The degree of deterioration can be calculated considering the decreased molecular weight and mechanical characteristics or surface deterioration such as discoloration or erosion. The outcomes of this study can be useful to determine the stability of material for the different environmental conditions. Notably, accelerated aging and outdoor condition relations are not straightforward because the environmental condition can vary depending on the location of the applied material. The shift or acceleration factor put in the correlation between simulated and outdoor conditions.

2.2 Environmental Conditions and Variables

During the aging process of the polymer matrix, some environmental deterioration factors are always active, their work being conducted through a molecular process. that works on the network: these changes, severally, calculate remarkable variation at the stage of bulk mechanical characteristics. Therefore, a well-designed accelerating protocol should swift the work of deterioration factors without outgoing from the fundamental molecular processes. The aging processes on polymer matrix biocomposites may lead to unchangeable (damage accumulation) and changeable effects. Several parameters such as temperature and temperature changes of polymer matrix and polymer morphology, fiber structure, relative humidity, degree of damage, oxidative attack, mechanical loads and cyclic loads, internal moisture concentration, fiber volume fraction, air pressure, and related applied stress level, and aging time induce and drive the damage accumulation continuously. Different portions of the structure will not expand similar amounts if the temperature throughout the structure is different. As a result, it increases the thermal stresses, which are combined with various destined stresses. Consequently, the modification happening in real-time circumstances for long period solicitations, regenerating the deterioration processes copied by the swift testing. The deterioration process, kinetics, and their influence all need to understand to determine the deterioration, which is relatively difficult. However, assessing the initial deterioration can be an easy way to comprehend the deterioration. It is also significant to use physical characteristics (crack density, glass transition temperature, and weight) to evaluate the condition of a material exposed to long-term aging. This condition can be interrelated through a formative way to alter bulk characteristics connected for the material engagement. Overall, the accelerated aging and real-time aging have a significant impact on the mechanical characteristics of materials to evaluate the accelerating factor for every deterioration process.

3 Aging Processes for Mechanical Characteristics

3.1 Mechanical Deterioration Process

The mechanical deterioration process directly affects engineering characteristics such as strength because deterioration causes delamination, matrix cracking, fiber breaks, interface damage, and inelastic deformation (Ahmad et al. 2018). During high-temperature operation of polymer composites, lateral multiple cracks and micro-cracks (which are situated in a plane) in the polymeric matrix of multi-dimensional composites are the most critical damage process. Mechanical static and fatigue loadings are the main reason for lateral multiple cracks and micro-cracks. In contrast, mechanical and environmental factors, and hydrothermal exposure cause residual stresses. Lateral matrix cracks of composites laminates are produced due to hydrothermal cycling that begins within the surface and develops deeply in the

laminate with cycles. Lateral matrix cracking affects composite stiffness, strength, and thermal characteristics. Lateral matrix cracking can also increase the absorption of moisture in the laminate.

3.2 Physical Deterioration Process

Polymers below glass transition temperature are in a non-equilibrium condition and want to reach the equilibrium condition. In this process, volume, enthalpy, and entropy decrease with time. This process increases stiffness and brittleness, which helps to the rapid advancement of several damage situations. As a result, thermal behavior, associated reduction of free volume, and segmental dynamics are the factors on which the mechanical characteristics of glassy polymers and their composites depend. Viscoelastic relaxation times of polymers increase with increasing aging time because of the time aging and time–temperature factors (Babaghayou et al. 2016). This makes the exploration of polymer matrix composites difficult because physical aging is related to the time-dependent viscoelastic behavior of polymers at a higher or lower temperature than glass transition temperature under load.

3.3 Thermo-Oxidative Deterioration Process

Thermo-oxidative deterioration is an important deterioration process of polymeric matrices because of thermal instability and oxidative attack, thermo-oxidative reactions, chain scission, and crosslinking (Lila et al. 2019). In an inert atmosphere, thermal deterioration of PMCs is a thermolysis phenomenon, but in the air, it is subjugated by oxidation (Fong et al. 2015). Thermolysis is defined as the breaking of a covalent bond in the polymer network. Normally, the thermal stability of high-performance polymers is very good in an inert condition. In polymer matrix, chemical and mass variations occur from the surface, and these changes significantly happen when exposing to high temperature and long-term exposure (Callister and Rethwisch 2018). Even slight mass variations can be an indication of surface variations. Oxidation reactions depend on the circulation of O_2 inside and circulation of oxidized products outside mainly the mass transport process. The resin fracture toughness has a major effect on thermo-oxidative resistance. Large fracture toughness increases the thermo-oxidative resistance due to reduced weight loss during aging. The toughened structure also provides better resistance to crack formation, which could supply a trail way for O_2 circulation. The nature of oxidation reactions and their related chemical changes have a significant influence on the accelerated aging (Martin 2008). These figures on temperature determining changes in mechanical characteristics. The nature of the reaction can vary remarkably with the variation of the polymer matrix. The

presence of unreacted and defective groups is also the reason for the topical problems. For the desired application, if we can identify correct deterioration processes that will help significantly to conduct an appropriate test program.

3.4 Hygrothermal Deterioration Process

The polymeric matrix of composites can absorb and circulate the low molecular mass element such as H_2O. The equilibrium absorbed amount and mass transport kinetics vary according to the chemical structure and morphology of a polymer. The arrangement and quantity of the fibers and fiber-matrix interfacial behavior are related to these characteristics in the case of composites. A simple approach based on Fick's law can be used for determining the time evolution penetrant concentration profile of a low molecular mass element when the absorbed amount is vanishingly small. The combination of moisture and heat influences reinforced plastics in many ways. The high influence of moisture is reflected by changes in matrix-dominated mechanical characteristics that are closely associated with the glass transition temperature. Humidity and high-temperature plasticize the matrix and accelerate the polymers' sub-glass transition temperature relaxation process in the direction of thermodynamic equilibrium (Panthapulakkal et al. 2006). Moreover, the residual stresses within the composites are changed by the hydrothermal swelling that can lead to micro-crack formation. These micro-cracks provide fast circulation paths and then alter the moisture absorption characteristics of the laminate. Surface blistering and matrix cracking are the short-term hydrothermal effects on composites under micro void generation, hydrothermal cycling conditions, and no applied loads.

4 Effect of Aging on Mechanical Properties

4.1 Accelerated Photochemical Aging

This section discusses the mechanical characteristics of biocomposites before and after accelerated photochemical aging. Moreno et al. (2018) investigated the biocomposites consisted of recycled low-density polyethylene waste (LDPEW), pinewood waste (PWW), and maleic anhydride grafted polyethylene (coupling agent). The LDPEW/PWW composite has a tensile strength of 12 MPa before photochemical aging, as shown in Fig. 1. The tensile strength decreases with aging up to 48 h, but it restores after 672 h of aging in the case of LDPEW/PWW composite. A rise in tensile strength occurs up to 168 h of aging because of residual stress relaxation that is stored in the material during their thermomechanical molding, and then it remains the same up to 1344 h of aging before reducing slightly up to 4032 h of aging. In another study, Stark and Matuana reported that polyethylene experiences crosslinking at the

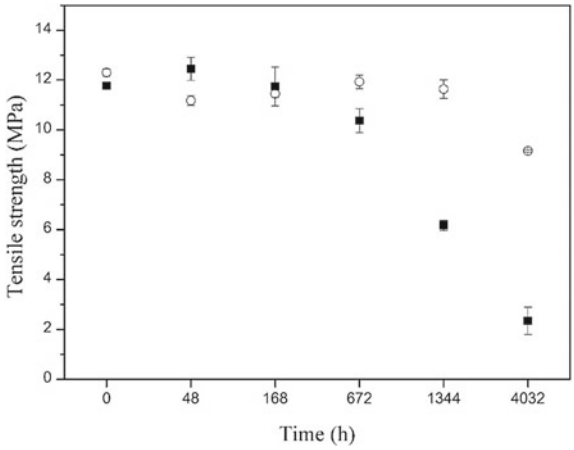

Fig. 1 Tensile strength of LDPEW (■) and LDPEW/PWW (○) composite as a function of time of aging (Moreno et al. 2018). Adopted with permission from copyright (2018), Elsevier

preliminary stage of accelerated aging, and the ability of polyethylene to crosslink is then hindered by wood flour (Stark and Matuana 2004). As a result, there is a potential for polyethylene chain scission to dominate in the primary aging stage.

The influence of accelerated weathering on the elongation at break for LDPEW and LDPEW/PWW is presented in Fig. 2. The LDPEW/PWW composite has an elongation of around 25% before aging. Elongation is slight because of the low deformation of the fiber. Elongation at break decreases substantially in LDPEW than LDPEW/PWW composite. Interestingly, after 4032 h of aging, elongation at break is the same for both neat LDPEW and LDPEW/PWW composite.

Figure 3 shows Young's modulus for LDPEW and LDPEW/PWW composite during accelerated aging. The increase in Young's modulus of LDPEW/PWW composite is 300% as compared to the LDPEW. However, there are no significant changes in Young's modulus of composites with aging time.

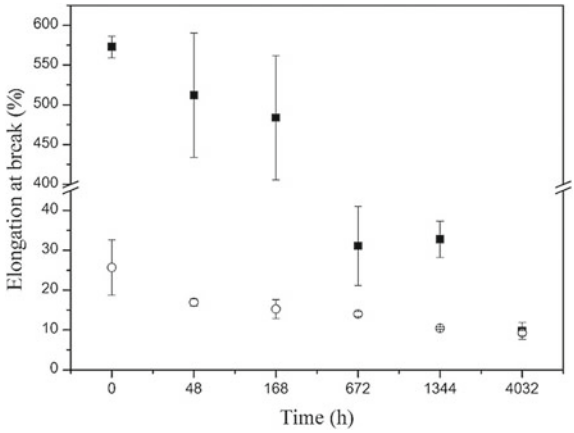

Fig. 2 Variation in elongation at breaks with weathering time for LDPEW (■) and LDPEW/PWW composite (○) (Moreno et al. 2018). Adopted with permission from copyright (2018), Elsevier

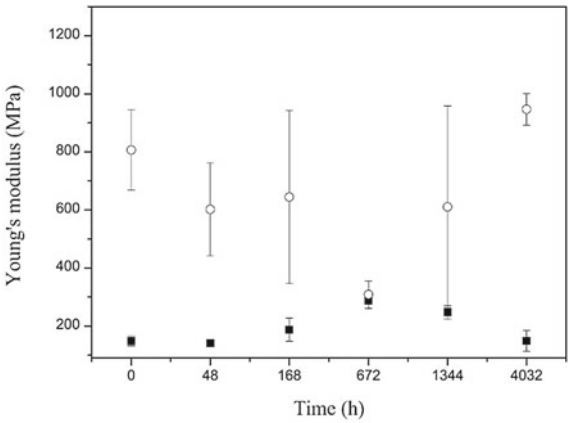

Fig. 3 Young's modulus of LDPEW (■) and LDPEW/PWW (○) as a function of weathering time (Moreno et al. 2018). Adopted with permission from copyright (2018), Elsevier

4.2 Hydrothermal Aging

The mechanical characteristics of the biocomposites during hydrothermal aging are discussed in this section. Kollia et al. examined the influence of the hydrothermal aging of flax/epoxy composites (Kollia et al. 2020). They used three different types of resins, such as greenpoxy56 (GP56) with hardener SD8822, greenpoxy56 (GZ56) with hardener SZ8525, RSF816RG (RSF816) with hardener RSF816, super sap INR (SP) with hardener super sap INS.

The stress–strain curves significantly change when specimens are exposed to distilled water. Stress–strain curves of the specimens at pristine state, and dry and wet conditions after saturation are illustrated in Fig. 4. Strain to failure affected by hydrothermal aging. A significant increase in strain to failure is observed due to water absorption, which even becomes higher with an increase in temperature. Strain to failure of the samples tends to decrease after drying.

Figures 5 and 6 show the tensile modulus and strength of GP56, GZ56, RSF816, and SP biocomposites, respectively, at both dry and wet conditions. During hydrothermal aging, the modulus of elasticity decreases for all samples, especially at 60 °C due to higher water uptake. Maximum reduction in stiffness happens for the GZ specimen and minimum for the SP specimen because of the variations in the production processes. Improvement in stiffness happens after drying.

The tensile strength of specimens reduces when exposing to water and high temperature. Water molecules are absorbed by the fiber due to its hydrophilic nature, which affects the fiber-matrix interfaces and minimizes interfacial bonding strength. Therefore, there is a reduction in tensile properties (stiffness and strength) after water aging.

Figures 7 and 8 show the flexural modulus and strength of biocomposites, respectively, during hydrothermal aging. The flexural modulus of GZ56 and SP816 specimens decreases by 55 and 26%, respectively, after hydrothermal weathering at 40 °C. Flexural modulus decreased further when water aging was conducted at 60 °C. The

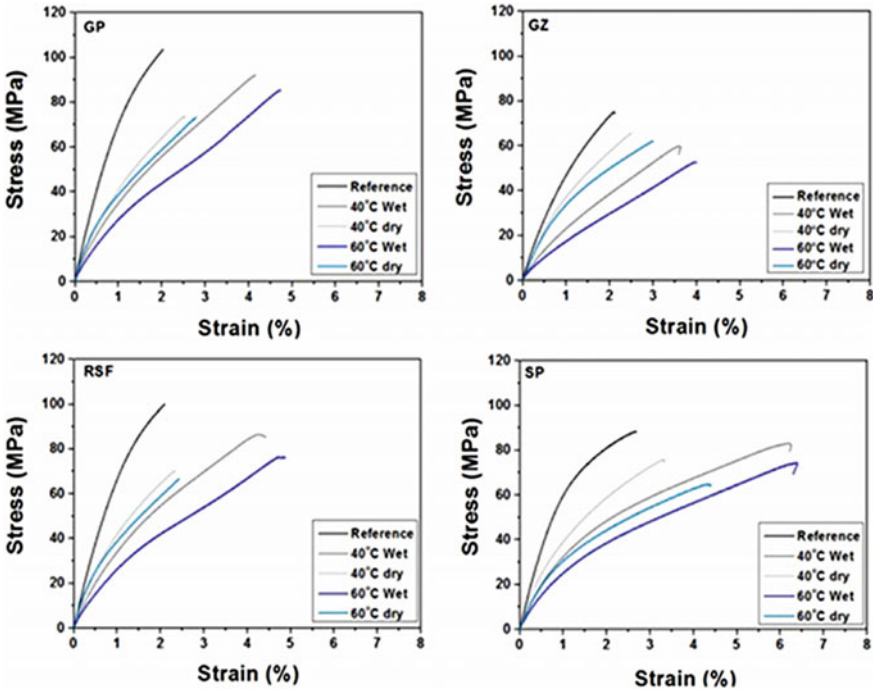

Fig. 4 Stress–strain curves of GP56, RSF816, GZ56, and SP biocomposites at the pristine state and wet and dry conditions for 40 and 60 °C (Kollia et al. 2020)

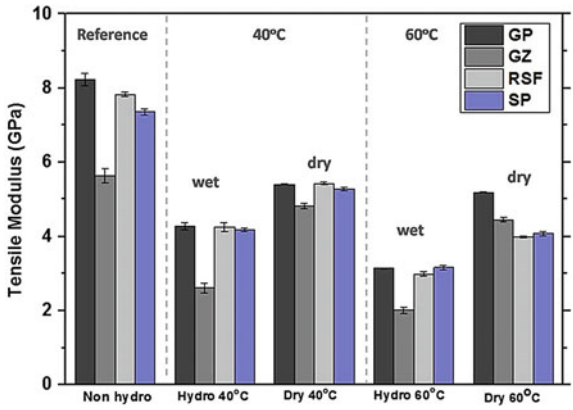

Fig. 5 Tensile modulus of GP56, RSF816, GZ56, and SP biocomposites before and after hydrothermal weathering at 40 and 60 °C (wet and dry conditions) (Kollia et al. 2020)

flexural modulus of the samples recovers slightly after drying. For all aged samples, the strain to failure is 5% higher than the dried samples. Strain to failure is increased in wet condition because water molecules act as a plasticizer.

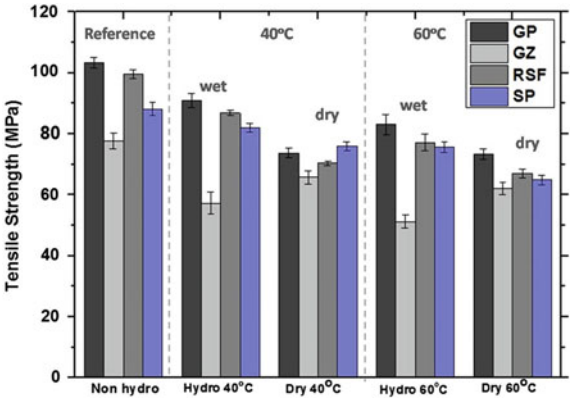

Fig. 6 Tensile strength of GP56, RSF816, GZ56, and SP biocomposites before and after hydrothermal weathering at 40 and 60 °C (wet and dry conditions) (Kollia et al. 2020)

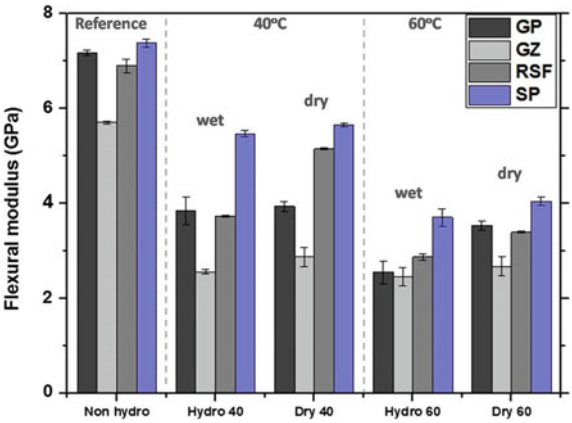

Fig. 7 Flexural modulus of GP56, RSF816, GZ56, and SP biocomposites before and after hydrothermal weathering at 40 and 60 °C (wet and dry conditions) (Kollia et al. 2020)

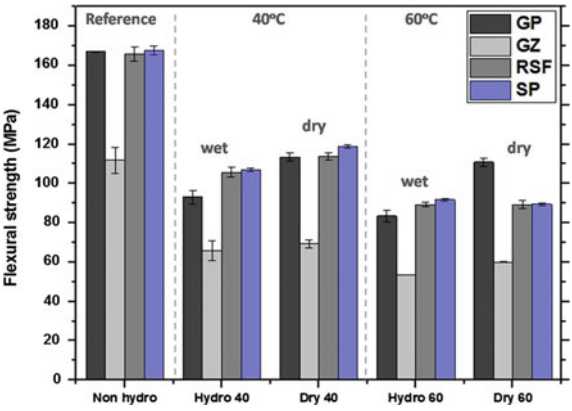

Fig. 8 Flexural strength of GP56, RSF816, GZ56, and SP56 biocomposites before and after hydrothermal weathering at 40 and 60 °C (wet and dry conditions) (Kollia et al. 2020)

The ultimate flexural strength is significantly affected by the aging process. For example, flexural strength decreases around 50% for all specimens at 60 °C after aging. This is again because of the decline in fiber-matrix adhesion after water absorption by the plant fiber. However, the flexural properties (stiffness and strength) of the samples restore slightly after drying.

4.3 Natural Aging

Natural aging affects the mechanical characteristics of recycled polypropylene, ethylene vinyl acetate, and wood flour composites (PP, EVA, and ITA), as discussed in this section. Natural aging was conducted for 4, 8, and 12 months for these materials by Silva et al. (2017). They found that different environmental conditions (rain, humidity, and solar radiation) affect the samples during natural aging. The effects can even be severe on the mechanical characteristics of the sample when the coupling effect of environmental conditions occurs (Homkhiew et al. 2014). Elastic modulus PP/EVA and PP/EVA/ITA specimens increase with the exposure time (see Fig. 9). The rise in tensile modulus happens from 483 to 1095 MPa under natural aging. This is because the chain scission phenomenon occurs during aging, which results in shorter macromolecular chains and promotes PP crystallization.

Figure 10 demonstrates the tensile strength of PP/EVA and PP/EVA/ITA at natural aging. The tensile strength at break decreases significantly for both composites because of the chain scission reaction induced by irradiation. Elongation at break of the composites also decreases with increasing natural aging.

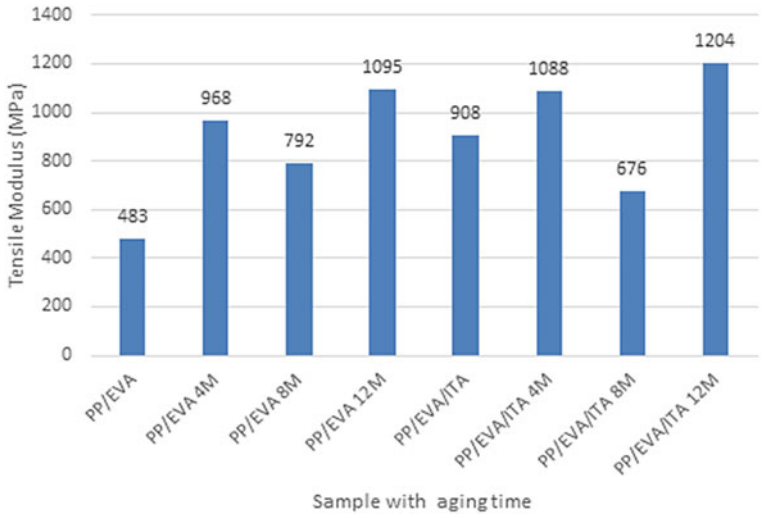

Fig. 9 Changes in tensile modulus after natural aging (Silva et al. 2017)

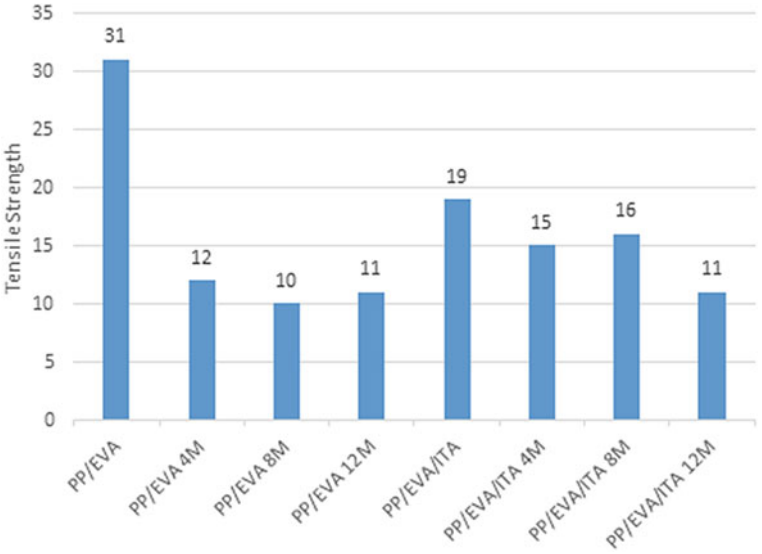

Fig. 10 Changes in tensile strength after natural aging (Silva et al. 2017)

Figures 11 and 12 show the flexural modulus and strength of PP/EVA and PP/EVA/ITA specimens, respectively, under natural aging. Flexural modulus and strength increase by 13.45 and 5.16%, respectively, after four weeks of exposure time in the case of PP/EVA/ITA specimens. All properties (tensile and flexural) reduces substantially after eight weeks of aging time. The decrease in flexural strength is

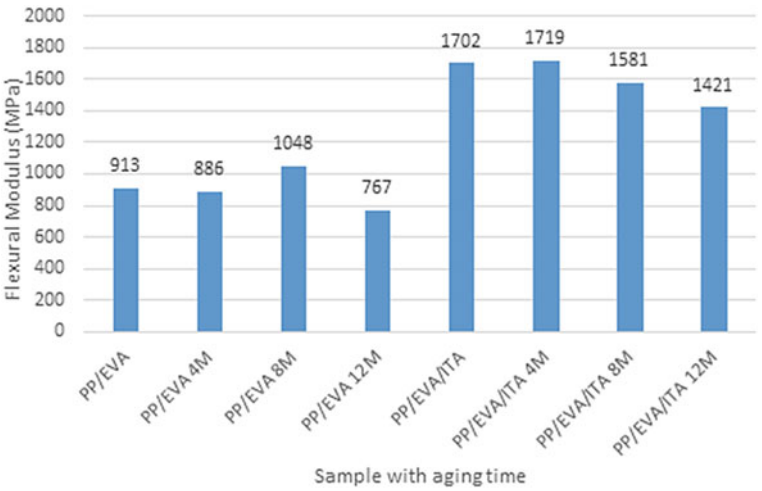

Fig. 11 Changes in flexural modulus after natural aging (Silva et al. 2017)

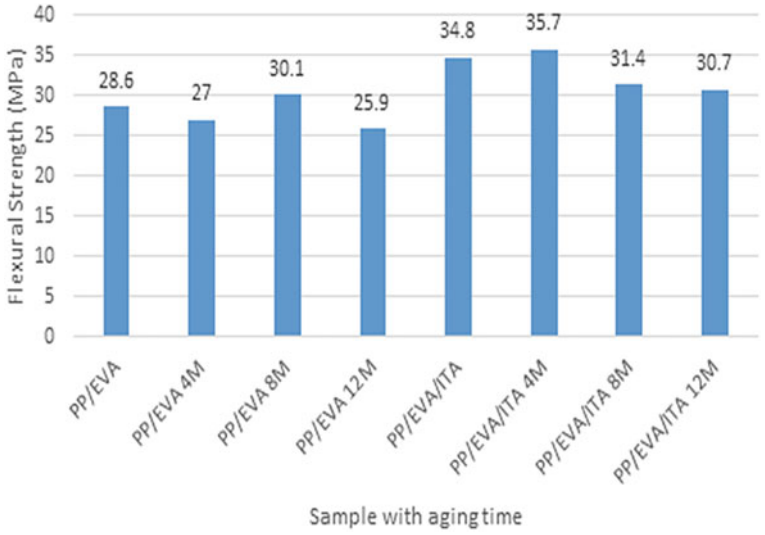

Fig. 12 Changes in flexural strength after natural aging (Silva et al. 2017)

because of the inability to transfer stress from wood flour to the plastic matrix and micro-cracks formation happened by wood flour swelling under the rain.

Elongation and contraction of materials happen due to the heating and cooling cycles. The thermal coefficients of natural fiber and polymer matrix are different, resulting in residual stresses generated within the structure after 8 weeks of exposure time. This also leads to structural deformation, thereby reducing the mechanical properties after eight weeks.

4.4 Thermal Aging

The mechanical characteristics of bagasse fiber/polylactic acid (PLA) composite under accelerated thermal aging are discussed in this section, based on the study made by Lila and co-workers (Lila et al. 2019). A temperature cycle of −20 to 65 °C was applied to the biocomposites for twelve hours a day at each temperature for 12 weeks. The tensile strength and tensile modulus increase up to 8 weeks exposure time due to multiple annealing of biocomposites, as shown in Figs. 13 and 14, respectively. Multiple annealing also increases the stability of PLA. There is a reduction in tensile properties after 8 weeks of exposure time due to decreased interfacial adhesion between the fiber and matrix. The weak interfacial bonding is owing to the residual thermal stresses derived from different thermal expansion coefficients of fiber and matrix material under continuous heating and cooling cycles.

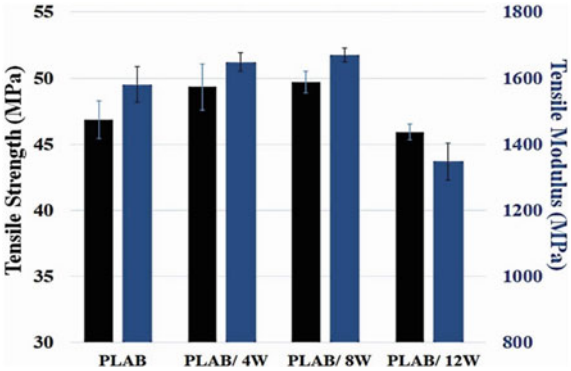

Fig. 13 Variation in tensile modulus of bagasse fibers reinforced PLA biocomposites under accelerated thermal weathering (Lila et al. 2019)

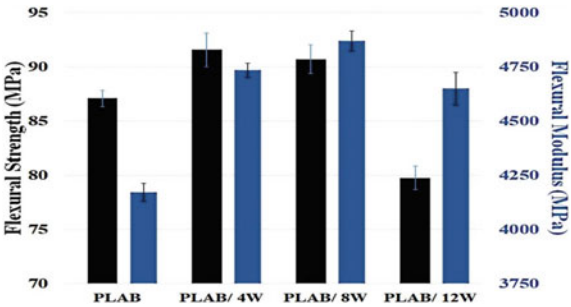

Fig. 14 Variation in flexural strength of Bagasse fibers reinforced PLA biocomposites during accelerated thermal aging (Lila et al. 2019)

4.5 Water Immersion-Freeze–Thaw Cycling Accelerated Aging and Xenon-Arc Light Accelerated Aging

The stability of composites largely depends on moisture. Lignocellulose fibers absorb moisture, and fiber swelling happens. As a result, cracks are formed in composites by destroying interfacial interaction. The stability of composites decreases with increasing the wood content (Turku et al. 2018). Changes in flexural and tensile properties due to freeze–thaw cycling are demonstrated in Fig. 15. The stability of composites can be increased using less hydrophilic filler material. The decrease in composite stability can happen because of removing lignin, washing of wood fibers, and reducing the amount of hemicellulose (Pilarski and Matuana 2005).

The mechanical properties (tensile and flexural) of wood-plastic composites decrease by 2–30% under accelerated freeze–thaw cycling and xenon-arc light conditions. Mechanical properties loss after full freeze–thaw cycling is similar to the properties loss after only water immersion. Mechanical properties reduce due to the decreased interfacial interaction by absorbed moisture in the composite (Matuana and Kamdem 2002). The swelling of wood particles also happens due to the existence of water. As a result, stress is generated into the matrix, and crack formation

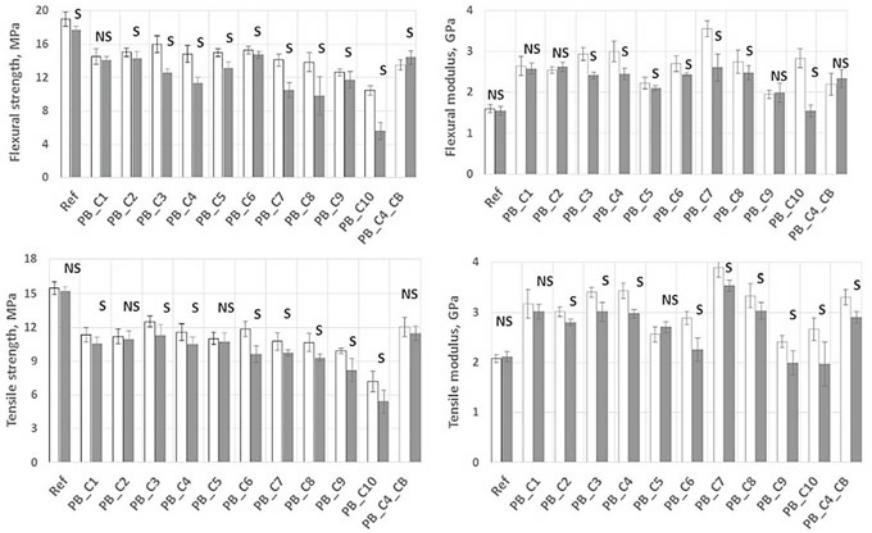

Fig. 15 Changes in flexural and tensile characteristics due to freeze–thaw cycling and xenon arc accelerated aging (Turku et al. 2018)

occurs. However, the mechanical properties of the composites can be restored by drying.

5 Conclusions

This chapter discussed the effect of aging on the mechanical properties of biocomposites with recycled polymers. Mechanical characteristics of biocomposites during aging depends on the aging process (Natural aging, Thermal aging… etc.). The mechanical properties (tensile and flexural) of wood-plastic composites decrease by 2–30% under accelerated freeze–thaw cycling and xenon-arc light conditions, bagasse fiber/polylactic acid (PLA) composites under accelerated thermal aging reduced due to weak interfacial adhesion between the fiber and matrix, recycled PP/EVA and PP/EVA/ITA composites under natural aging showed a positive effect. Generally, biocomposites have better resistivity against environmental factors. Mechanical properties (tensile modulus and strength, and flexural modulus and strength) reduce after aging, and they can be recovered after drying.. From the aging result, it is possible to obtain information about the suitability and durability of the composite materials for a specific application. Before application of any product if we conduct a suitable aging process that will be beneficial. Bio-composites with recycled polymers are not limited number. Everyday people think about it and find something new application. However, there is still an enormous research opportunity

for evaluating the aging effects on mechanical characteristics of biocomposites with recycled polymers.

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