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Interface shear damage of novel biocomposite materials based on polyhydroxyalkanoates biopolymer matrix

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ABSTRACT

Bioplastics reinforced with natural fibers are gaining attention for their potential in various emerging eco-friendly applications, resulting in new materials with exceptional mechanical and thermal properties. This study examines PHA biopolymer plastic as a polymer matrix for natural fibers, specifically date palm, coconut, and sisal. It focuses on calculating shear damage at the fiber-matrix interface for the biocomposites date palm/PHAs, coconut/PHAs, and sisal/PHAs. The results show that under mechanical stresses (σ = 40, 45, 50, and 55 MPa), damage levels increase due to the accumulation of stresses, with sisal/PHAs exhibiting more severe degradation at the interface compared to date palm/PHAs and coconut/PHAs. These findings highlight the significant role of natural fibers in enhancing the mechanical properties of composite materials.

KEYWORDS

bioploymers • PHA • natural fibers • date palm • coconut • sisal

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Introduction

Nowadays, the exploration of composite and biocomposite polymers has captivated researchers and manufacturers alike, motivating an in-depth investigation into this dynamic field [1–4]. The focal point of this pursuit extends beyond the enhancement of existing properties, aiming to innovate and introduce novel materials tailored for a diverse range of contemporary applications [5,6]. Composite materials, constructed by arranging various components such as fibers (synthetic or natural), matrices (thermoset or thermoplastic), and fillers, undergo a transformative process, resulting in the creation of materials with enhanced mechanical, thermal, and physicochemical properties [7,8].

These materials often exhibit superior strength [9], durability [10], and resistance to environmental factors compared to their individual constituents [11]. The emergence of biocomposite polymers, where natural fibers or fillers are incorporated into a polymer matrix, underscores a deliberate move towards sustainability and eco-friendliness in material design [12,13]. These materials find widespread application in diverse sectors, including biomedical, telecommunications, construction, aeronautics, and beyond [14–16]. In the biomedical field, the biocompatible nature of these polymers makes them invaluable for the development of implants, drug delivery systems, and other medical devices [17,18]. In telecommunications, their lightweight and durable properties make them ideal for components in electronic devices and sensing components [19,20]. Moreover, these materials play a pivotal role in eco-conscious building practices, offering sustainable alternatives for construction materials [21–23]. In aeronautics, where lightweight yet robust materials are essential, composite and biocomposite polymers have become integral to aircraft design, contributing to fuel efficiency and environmental sustainability [24–26]. In the pivotal area of food packaging, these materials are increasingly recognized for their eco-friendly nature and tailored properties, presenting a sustainable choice that aligns with environmental consciousness [27,28]. This expansive influence underscores the pivotal role of these materials in shaping a greener and more technologically advanced future across diverse industries. In the scope of this study, our attention is directed towards a meticulous investigation into the polyhydroxyalkanoates (PHA) biopolymer plastic as polymer matrix for natural fiber namely date palm, coconut, and sisal. The principal objective is to attain a thorough understanding of how the distinct reinforcing fibers impact the behavior of the PHA biopolymer matrix. In recent literature, several works have been dedicated to investigating the properties of PHA, positioning it as a leading candidate for emerging applications. The authors in [29] have conducted an extensive examination of the latest progress in PHA bioproduction, covering a spectrum from native to diversified cell factories. The exploration encompasses discussions on recovery and purification techniques, and the evaluation of the current status of industrial applications is positioned as a crucial milestone for startups. The culmination of the overview involves addressing contemporary challenges and outlining future prospects, providing valuable insights into a trajectory geared towards diminished carbon emissions and the realization of sustainable development goals. In [30], authors have presented an overview of the natural weathering behavior exhibited by representative completely biodegradable composites. The impacts of natural weathering on these composites closely align with those observed in natural fiber composites featuring conventional non-degradable matrices. Interestingly, no discernible influence of biodegradation from the biopolymer matrices manifested under natural weathering conditions for a minimum of 2 years. However, physical and mechanical deteriorations of the composites became evident after a few months of exposure, with the severity primarily dependent on the fiber content. In essence, the accessibility of the natural fibers and the rate of moisture ingress into the bulk matrix emerge as crucial factors governing the stability of biodegradable composites during natural weathering. Furthermore, a multitude of recent studies has underscored the profound impact of polyhydroxyalkanoates (PHA) biopolymers, shedding light on their fabrication processes and elucidating their influence across various contemporary industrial trends [31–34].

The present work delves into a comprehensive exploration of the mechanical properties of the polyhydroxyalkanoates (PHA) biopolymer, particularly when reinforced with natural fibers such as date palm, coconut, and sisal. The overarching goal is to develop novel materials boasting enhanced mechanical strength. By scrutinizing the distinctive characteristics of each fiber type, the study seeks to unveil their individual impacts and collective contributions to the overall attributes of the biocomposite. The investigation entails the calculation of shear damage at the fiber-matrix interface for three specific biocomposite materials date palm/PHAs, coconut/PHAs, and sisal/PHAs employing a genetic operator crossing approach based on the length of each fiber. This analysis is conducted under the influence of various applied mechanical stresses. The focused examination is intricately designed to provide valuable insights into the dynamic interplay between Date Palm, Coconut, Sisal, and the Polyhydroxyalkanoates (PHA) biopolymer matrix, thereby enriching our comprehension of the interface behavior in biocomposite materials such as bioplastic polymers.

Materials and Methods

Polyhydroxyalkanoates biopolymer matrix

Polyhydroxyalkanoates (PHAs) have very varied physicochemical properties depending on their chemical composition [35–38]. For example, the mechanical properties and crystallinity of the material vary depending on the presence of co-monomers. They can be implemented using techniques comparable to phase inversion and are resistant to most organic solvents. The PHA is non-toxic, biocompatible, biodegradable thermoplastics that can be produced from renewable resources. They have a high degree of polymerization, are highly crystalline, optically active and isotactic (stereochemical regularity in repeating units), piezoelectric and insoluble in water. These features make them highly competitive with polypropylene, the petrochemical derived plastic [39].

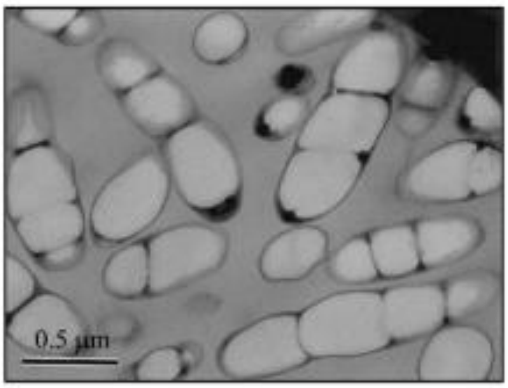

Fig. 1. Electron microscopy image of bacteria (Ralstoniaeutropha) containing polyhydroxyalkanoate granules

PHAs are a family of linear, biosourced and biodegradable polyesters, with varied structures and properties. PHAs are made of hydroxyalkanoate (HA) monomer units with different lengths and functionalities. The nomenclature of a monomer unit refers to the total number of carbons present and the location of the side chain if there is one. They have the particularity of being produced by bacteria. The first bacterial polyhydroxyalkanoates (PHAs) were discovered in 1925 by the French scientist Maurice Lemoigne [37,39,40]. Indeed, in the presence of a source of carbon, nitrogen and oxygen, certain Bacteria have the ability to generate PHA granules to use as an energy reserve. Figure 1 highlights in particular by transmission electron microscopy the presence of these PHA granules in bacteria of the Ralstoniaeutropha type [37,41]. The properties of PHA depend largely on the nature and content of the monomers which it contains [35–38]. In Table 1, the physical properties of the PHA biopolymer have been illustrated [42–45].

Table 1. Physical properties of the PHA biopolymer [42–45]

Matrix	Young's modulus, GPa	Tensile strength, MPa	Elongation at break, %
PHA	3.0	43.0	5.0

Fibers

Date palm fibers (DPFs) have very interesting properties that allow them to be exploited in different applications such as the production of biochar [46] and the manufacture of ropes and baskets [47]. In addition, they are biodegradable and they have a very low environmental impact and above all their technical-economic advantages, DPFs are widely used in industrial sectors and currently in automation, automobiles, aeronautics and construction in order to improve thermal insulation [48,49], The literature showed that date palm fibers can have high cellulose purity up to 70 %. The density of date palm is between 1.18 and 1.45 gm/cm^3 which are comparable to other widely used natural fibers. Date palm fibers can be used in automotive applications, in particleboard manufacturing and as reinforcement to panels to be used as thermal or sound insulation [50]. The major problem is that PDFs are hydrophilic in nature, which generates compatibility and interfacial bonding problems due to the hydrophobic nature of their matrix. To solve this problem, several physical and chemical treatment methods have been considered by many researchers and scientists [51,52]. One of the most effective is chemical treatment using the sodium hydroxide process. The principle of this treatment is to eliminate the existing amorphous polymers on the surface and to obtain cellulose-rich fibers which improve the mechanical properties of the pure matrix when used as reinforcement [53,54].

Coconut fibers. Coconut is the fruit of the coconut palm (Cocosnucifera), one of the representatives of the palm family or Arecaceae. The exterior of the fruit is smooth and light green or orange in color when the flower is not ripe (young coconut); brown in color and covered with a thick layer of brown woody fiber surrounding the mature nut (ripe coconut), and composed of a solid spherical shell which protects an edible whitish kernel. The coconut palm is found in tropical countries. The coir surrounds and protects the coconut, the fruit of the coconut tree. It is extracted manually or mechanically, then washed and dried. The fibers measure up to 35 cm in length with a diameter ranging from 12 to 25 μm. They have a high lignin content which makes them stronger than cotton.

They also have good resistance to microbial action and salt water. Traditionally, coconut fiber is used to make rope, fishing nets, canvas, carpets, brushes, brooms or mattress padding. More recently, geotextiles, made entirely from coir, are used to combat soil erosion. We also find insulation in the form of flexible panels and plywood (fibers and resins). Coconut fibers are classified as seed or fruit fibers. Coconut fiber is entirely biodegradable and transformed into substrate. It is suitable for mulching poor and sandy lands, hydroculture, massifs, annuals, flowering plants, trees, shrubs and rose bushes as well than the ground cover. These fibers are short and fine. In fact, plant fibers are classified into three categories: 1 - Seed or fruit fibers (e.g. cotton, coconut), 2 - Leaf fibers (e.g. sisal, abaca, esparto), 3 - Bast fibers or stem fibers (Ex: flax, jute, hemp, ramie) [55].

Sisal fibers. Sisal is a resistant plant in hot and arid climates, often unsuitable for other cultures. It only flowers once times, just before dying. Like all agaves, sisal forms a rosette of thick leaves that ends in a sharp point and has spiny edges. These leaves, although full of water, are very rigid thanks to the numerous fibers they contain. Fibers located inside the leaves lengthwise, are more abundant towards the surface. Defibration is carried out just after harvest. The pulp is scraped by hand or mechanically, releasing the fibers. They are several long tens of centimeters, thick and solid. A drying step is necessary before gathering the fibers ready for braiding of the plant. Sisal fiber is traditionally used to make ropes like that used, before plastic replaced it, to tighten straw bales. Traditional sisal clothing is found in China and Laos. These fibers also serve as flexible reinforcements to reinforce plaster or for composite materials. Sisal is increasingly used in high value-added finished products and for various other uses; it is the constant and superior quality of this sisal which allows its use in more profitable finished products. These product categories particularly plaster reinforcement in the construction sector and pulp production; have experienced constant growth for several years. The long fibers which are part of the hard fibers are generally used for the manufacture of baling twines, tying twines, ropes, maritime twines and polishing fabrics. Short fibers are used for the production of carpets and construction panels. Sisal fibers are also used in the paper industry to improve the performance of filter paper made from wood cellulose pulp. [55–58].

The different physical properties of fibers used in genetic modeling have been illustrated in Table 2.

Table 2. Physical properties of different fibers used in genetic modeling

Fiber	Density, g/cm^3	Deformation, %	Tensile strength, MPa	Young's modulus, GPa
Sisal [59–62]	1.50	5.12	228.00	9.40
Coconut [63,64]	1.17	13.70	107.00	5.00
Date palm [64]	1.08	17.40	290.00	5.25

Weibull formalism

The findings in our investigation were obtained using a genetic simulation primarily rooted in the Weibull formalism. This approach was adopted to assess the damage at the interface of the three materials, incorporating genetic operators like selection, crossing, and mutation. The Weibull formula (Eq. (1)), as outlined in [64–68], is employed to characterize the damage to the matrix under uniform stress conditions.

$$D_m = 1 - exp\left\{-\frac{V_{eff}}{V_0}\left(\frac{\sigma_f}{\sigma_0}\right)^m\right\}, \tag{1}$$

where σ_f represents an applied stress, V_{eff} denotes the matrix volume, m **and** σ_0 are the Weibull parameters, V_0 signifies the initial volume of the matrix.

The fiber is regarded as an element within an assembly of links, each having its distinct breaking strength. Fracture frequency arises when the weakest link in the assembly experiences a break. The failure of a fiber along its entire length is primarily influenced by repetitions. This rupture can be characterized by a law analogous to that of the matrix, as described in [65,69,70].

$$D_f = 1 - exp\left\{-A_f * L_{equi} * \left(\frac{\sigma_{max}^f}{\sigma_{of}}\right)^{m_f}\right\}, \tag{2}$$

where σ_{max}^f represents the maximum stress applied to the fiber, σ_{0f} is an initial stress applied to the fiber, m_f are Weibull parameters, $A_f = \pi^* a^2$, L_{equi} is the length of the fiber at equilibrium.

Fiber-matrix interface damage

The interface or interphase exhibits distinct properties compared to the constituents, specifically concerning mechanical properties. Despite these differences, it holds a crucial role in facilitating the transfer of forces between the matrix and the fiber. The quality of the interface is, therefore, a determining factor in shaping the ultimate performance of composites, as emphasized in references [71,72]. For this reason, the mechanical characterization of this pivotal area is imperative. The characterization of the interface involves determining its mechanical properties based on the materials used for the fiber and matrix. Additionally, it may be contingent on any modifications such as treatments that the materials undergo, including chemical formulations of the matrix, surface treatments of fibers, aging, etc. [71]. The composite test pieces undergo various types of testing, exemplified by the tensile test conducted at a specified angle relative to the fiber direction, the interlaminar shear test on a short further, and the three or four-point bending test. Additionally, monofilament test pieces are subjected to a range of tests, including the release test pull-out or micro-drop, the fragmentation test, and the indentation test, among others [71]. In our numerical simulation, we employed Eq. (3) derived from the pull-out heaving test. This test involves raising a fiber, either partially or entirely immersed in a micro test tube, by applying a pulling force [71,73]. The average shear stress τ_{app} at the interface is determined by:

$$\tau_{app} = \frac{F_{max}}{2\pi r l_e}, \tag{3}$$

where F_{max} is the maximum force measured;r :average radius of the fiber; l_e is the length of the fiber inserted into the matrix (length of the interface).

Results and Discussion

The composite materials with fiber reinforcements have optimized mechanical and physical properties, which depend not only on those of their constituents, their architecture but also on the bond between fiber and matrix. The quality of this fiber/matrix interface, which plays a primordial role in the mechanical resistance of composite materials, depends on several factors that can be identified using different tests carried out at the microscopic scale. Most of the experimental techniques described the fiber-matrix interface are often expensive and require significant preparation and implementation time, and that the geometry of plant fibers is highly variable. There are often surface defects [71]. For this reason, we have proposed and developed a simple numerical method based on genetic algorithms to characterize the mechanical properties of the fiber-matrix interface shear damage. The current study involved the computation of shear damage at the fiber-matrix interface for three distinct biocomposite materials: date palm/PHAs, coconut/PHAs, and sisal/PHAs, employing the genetic operator crossing approach. The damage to the fiber and matrix was calculated using Eqs. (1) and (2), respectively. The material-specific values presented in Tables 1 and 2 were applied in the computation of the objective function. Subsequently, the interface shear damage was determined based on the length of each fiber for the studied materials, subjected to various applied mechanical stresses (σ = 40, 45, 50, 55 MPa).

As per Cox's model [74], the transfer of load leads to the generation of interfacial shear stresses, reaching maximum amplitude at the ends of the fiber and exhibiting very small values in the middle. Cox observed that these stresses display symmetry at the midpoint of the fiber. To aid in this analysis, he presented the two tensile and shear stresses, as illustrated in Fig. 2. This figure served to rationalize our selection of specific points utilized in our genetic modeling, namely (-L, -L/2, 0, L/2, L).

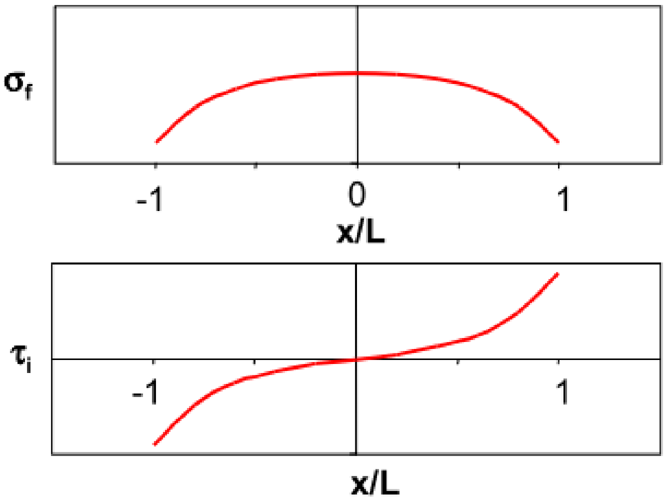

Fig. 2. Cox model; stress profiles in the fiber (σf) and at the interface (τi) [74]

Figures 3–5 visually represent the damage at the interface for the three distinct biocomposite materials. In particular, Fig. 3 depicts the initiation of shear damage at the interface (denoted as "D") for the date palm/PHAs biocomposite material. The damage initiates at a threshold of 0.089 when σ = 45 MPa and progressively increases to a maximum value of 0.165 at σ = 55 MPa. Furthermore, Fig. 2 illustrates the symmetry of weak damage in the middle of the fiber and strong damage at the ends of the fiber, as depicted by the black and green colors in Fig. 3. From the results, it can be inferred that an elevation in mechanical stress corresponds to an increase in the degree of damage. In the case of this material, the degradation of the interface is relatively modest.

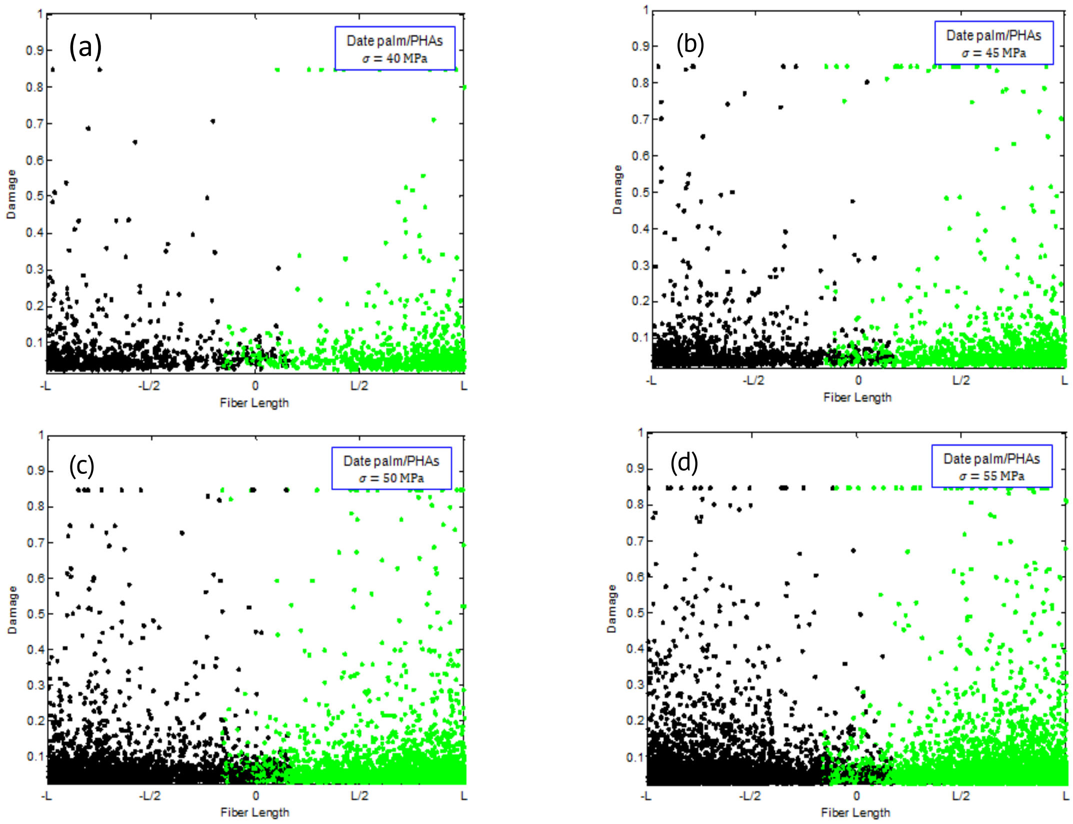

Fig. 3. Interface shear damage of date palm/PHAs biocomposite material:
(a) σ = 40 MPa, (b) σ = 45 MPa, (c) σ = 50 MPa, (d) σ = 55 MPa

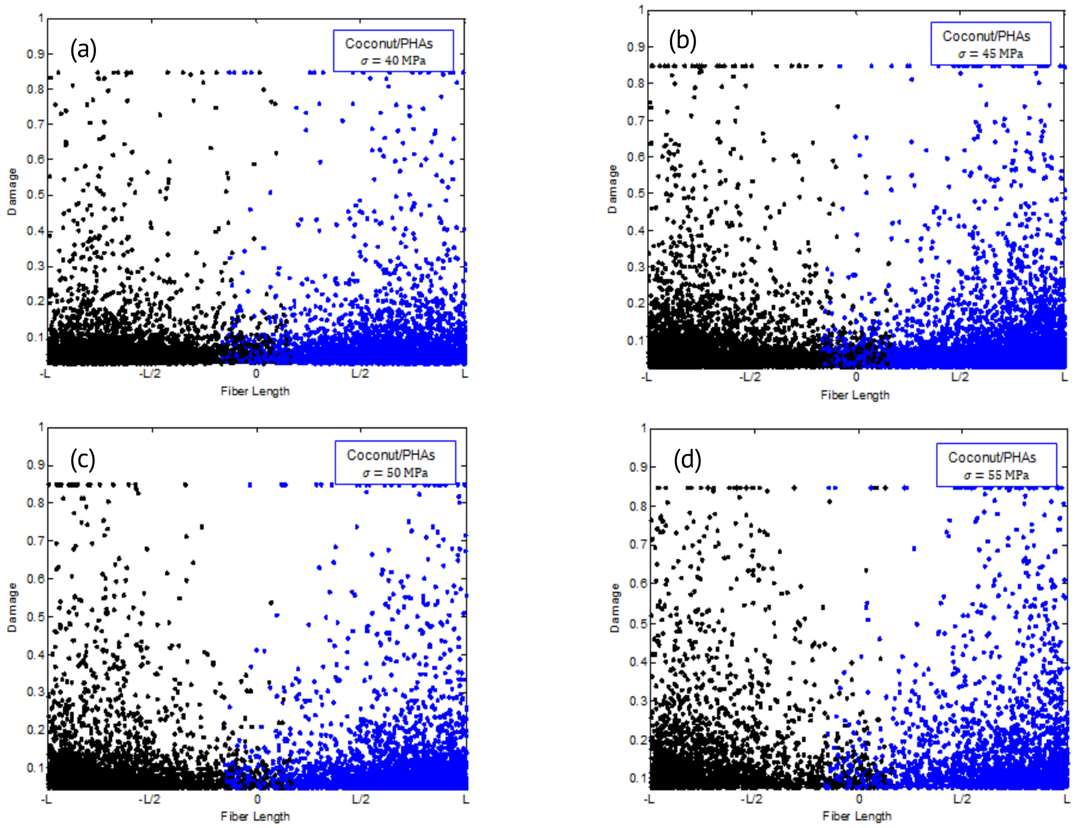

Fig. 4. Interface shear damage of coconut/PHAs biocomposite material:
(a) σ = 40 MPa, (b) σ = 45 MPa, (c) σ = 50 MPa, (d) σ = 55 MPa

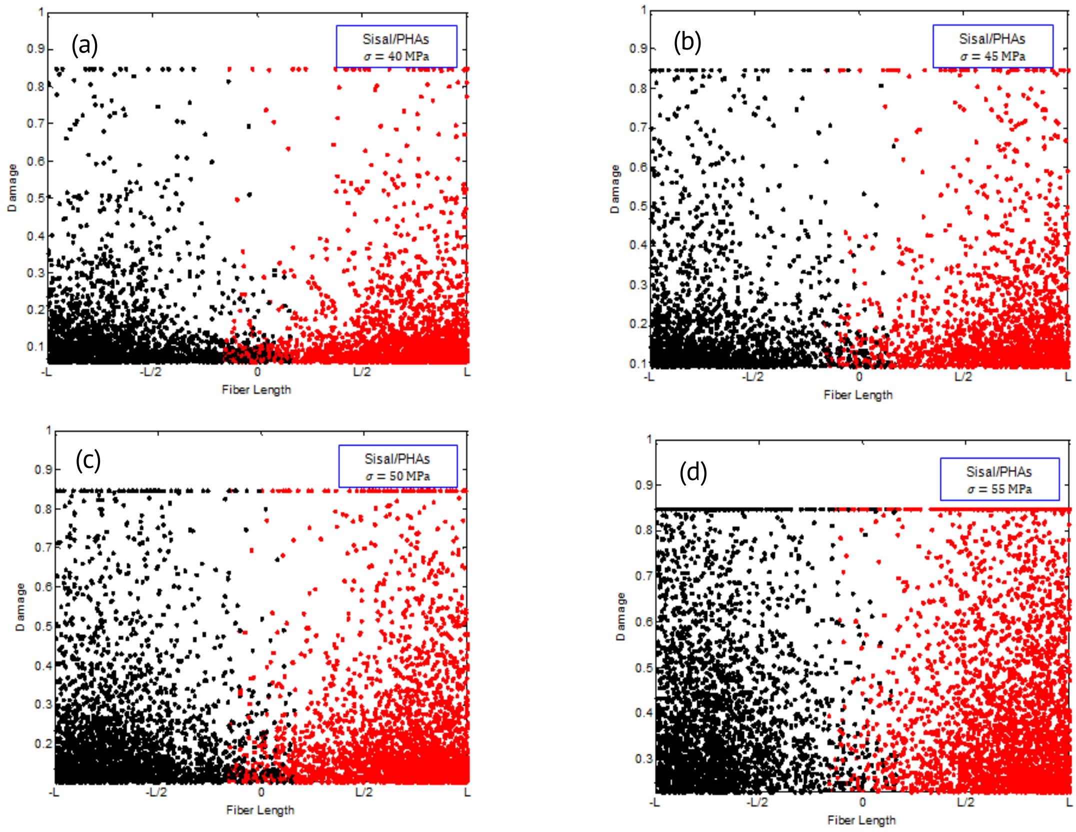

Fig. 5. Interface shear damage of sisal/PHAs biocomposite material: (a) *σ* = 40 MPa, (b) *σ* = 45 MPa, (c) *σ* = 50 MPa, (d) *σ* = 55 MPa

Figure 4 illustrates the extent of damage at the interface of the coconut/PHAs biocomposite material. The graph shows the initiation of shear damage at the interface, denoted as "D," at a value of 0.112 for a mechanical stress of σ = 45 MPa, escalating to a peak value of 0.2 at σ = 55 MPa. Similar to the preceding material, a symmetric pattern of weak damage in the middle and strong damage at the ends of the fiber is discernible for the coconut/PHAs biocomposite material. This is represented by distinct colors, black and blue, in Fig. 4. The elevation in damage level in the coconut/PHAs biocomposite material suggests that mechanical stresses concentrate at the interface, leading to a more pronounced degradation of the interface compared to the date palm/PHAs biocomposite material.

Figure 5 illustrates that the damage at the interface, labeled as "D," in the Sisal/PHAs biocomposite material commences at a value of 0.181 under a stress of 45 MPa and rises to a peak value of 0.389 when subjected to a stress of 55 MPa. Furthermore, the symmetry of the damage level is relatively low in the central region and significantly higher towards the ends of the fiber, as indicated by the contrasting colors of black and red in Fig. 5. The results suggest that the escalation in damage level is attributed to the accumulation of mechanical stresses, resulting in more severe degradation at the interface of the sisal/PHAs biocomposite material compared to the date palm/PHAs and coconut/PHAs biocomposite materials.

The aforementioned findings align with recent studies conducted by [50,75–78], which consistently affirm that the inclusion of natural fibers, especially date palm fiber, significantly enhances interfacial adhesion in composite materials. These studies highlight that natural fibers contribute substantially to the improved mechanical properties of composite materials, gradually supplanting synthetic fibers due to their specific strength, rigidity, lightness, and cost-effectiveness. Natural fiber-reinforced composites emerge as green, biodegradable, and sustainable materials, finding applications across various industries such as automotive, marine, aerospace, construction, wind energy, and consumer goods.

Conclusions

This study places a focused lens on the realm of bioplastics, particularly polyhydroxyalkanoates (PHA) reinforced with natural fibers (date palm, coconut, and sisal). The evaluation of shear damage at the fiber-matrix interface, facilitated through a genetic operator crossing approach based on the length of each fiber, sheds light on the pivotal role of mechanical stresses in influencing interface degradation. Findings indicate that varying mechanical stresses (σ = 40, 45, 50, and 55 MPa) correlate with an escalation in damage levels, underscoring the cumulative impact of these stresses. Noteworthy is the distinct behavior observed in the Sisal/PHAs biocomposite material, revealing more pronounced degradation compared to its counterparts. This observation accentuates the intricate interplay between natural fibers and the PHA biopolymer matrix. Importantly, these outcomes align with recent studies conducted by the authors in [50,75–78] highlighting the broader transition toward eco-friendly materials, emphasizing the substantial contribution of bioplastics reinforced with natural fibers in elevating mechanical properties.

In practical terms, the enhanced mechanical properties of these biocomposites make them suitable for a variety of eco-friendly applications. For example, packaging solutions, in lightweight automotive components, in durable construction materials. The specific degradation behavior of sisal/PHAs could also be advantageous in applications requiring controlled material breakdown, such as in agricultural or disposable products.

Future research should aim to optimize the fiber-matrix interface to reduce the degradation observed, especially in sisal/PHAs composites. Additionally, exploring other natural fibers or hybrid fiber combinations could enhance the performance of these bioplastics. Additionally, a key direction for future work involves incorporating advanced learning techniques, such as machine learning algorithms, to accurately model and predict the behavior of these biocomposites under various environmental conditions.

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