

# IMPROVING THE PERFORMANCE OF HYBRID BIOCOMPOSITE MATERIALS USING EPOXY, VINYL ESTER AND POLYIMIDE POLYMER MATRIX: COMPARATIVE STUDY

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Original scientific paper

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## Abstract:

The present study aims to assess the impact of alfa and date palm natural fibers as reinforcement for epoxy, vinyl ester, and polyimide polymer (Alfa-Date palm/epoxy, Alfa-Date palm/vinyl ester, and Alfa-date palm/polyimide) through a genetic approach. The study employs a genetic simulation rooted in the Weibull formalism, employing genetic operators such as selection, crossing, and mutation for a nuanced evaluation of the damage at the fiber-matrix interface across the studied materials. The genetic algorithm results authentically capture the intrinsic behaviour of these materials, emphasising key mechanical properties like Young's Modulus, deformation, and stress at break. The obtained results highlight that the matrix showing the highest Young's modulus value at its fiber-matrix interface exhibits superior strength. These conclusions align with contemporary advancements in natural fiber-reinforced composite materials tailored for diverse industries and eco-friendly applications.

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## 1. INTRODUCTION

Natural fibers are gaining increased attention for their pivotal role in elevating the properties of composite materials across various applications. Seamlessly woven into our daily lives, these fibers, often unnoticed, bring about a transformative impact [1-3]. The remarkable mechanical strength [4-5], resistance to thermal stress [6], and inherent flexibility [7] of natural fibers contribute significantly to enhancing the overall characteristics of composite materials [8-10]. In addition, as sustainability becomes a focal point in material science, researchers are actively exploring these fibers as alternatives to synthetic

counterparts, aiming for composite materials that not only match but surpass existing performance standards while aligned with our green ecosystems [11-12]. Notably, alfa and date palm fibers, abundant and possessing exceptional properties, emerge as promising contenders in this pursuit [13-15]. Alfa and date palm fibers find diverse applications across various industries due to their unique properties and versatility. Their exceptional mechanical strength makes them suitable for reinforcing composites used in construction [16,17], automotive sector [18], textile industry [19], food packaging [20], agriculture sector [21,22], and various industrial purposes [23-26]. In this context, the current study delves into

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evaluating the mechanical stress response of alfa and date palm fibers as reinforcements in well-established composite materials such as epoxy resin, vinyl ester, and polyimide. Understanding how these natural fibers impact the mechanical stress characteristics of these composites is crucial for advancing their utilisation in various applications. In recent literature, several studies have investigated the incorporation of natural fibers to enhance the mechanical properties of composite materials. Authors in the paper [27] highlighted whether the inclusion of CTS (chitosan) particles in a DP (date palm) / bio-epoxy composite could result in improved mechanical properties. Experimentations revealed that among the various CTS fractions studied, DP/CTS20 exhibited the highest tensile strength (24.04 MPa) and tensile modulus (4.93 GPa), flexural strength (45.11 MPa), and impact strength (2.70 J/m<sup>2</sup>) [28]. In the paper [29], authors investigated the influence of date palm fiber loading and orientation on the tensile properties of Date Palm Fiber Reinforced Polyester Composite (DPFRPC). The aim was to identify the optimal combinations of fiber loading and orientations that result in the highest tensile strength for DPFRPC. Their findings revealed that the tensile strength of DPFRPC increases with fiber loading, reaching an optimal level at 30%, beyond which it starts to decline. Conversely, as the fiber orientation angle increases, the tensile strength of DPFRPC decreases. Another study [28] where the main purpose was to study the impact of alfa fiber content, alkali treatment, maleic anhydride grafted polypropylene (MAPP) on the tensile and flexural behaviour of alfa fiber-reinforced polypropylene (PP) composites. Their outcomes revealed a substantial improvement in the tensile modulus and flexural strength of composites containing 30% untreated Alfa fiber in polypropylene (PP) compared to pure PP. However, there is no noteworthy increase in terms of tensile strength and flexural modulus. Additionally, composites subjected to a combined treatment involving both alkali and maleic anhydride grafted polypropylene (MAPP) exhibit a significant enhancement in both tensile and flexural properties compared to both untreated and individually treated composites. Another recent paper [30], where authors performed a numerical study utilising genetic algorithms and Weibull probabilistic approach to investigate the impact of date palm, doum palm, and alfafibers on fiber-matrix interface damage in biocomposite materials. Results align with recent studies, indicating that date palm fibers enhance

epoxy ductility and strength, while improving adhesion between fiber and polymer matrix, suggesting potential for ecological product development. In the paper [31], authors evaluated the tensile strength of epoxy bio-composites reinforced with palm fibers, treated with sodium carbonate NaHCO<sub>3</sub>, and varying fiber weight percentages. Two predictive methods, artificial neural network (ANN) and response surface methodology (RSM), were employed. Both methods demonstrated high accuracy in predicting the mechanical performance of the composites, with correlation coefficients exceeding 0.97 for stress and Young's modulus. Comparisons with experimental results revealed minimal errors for both ANN and RSM approaches [31]. Furthermore, several reviews, such as those by [32-37], have highlighted the significant impact of natural fibers on the mechanical properties and performance of composites. These studies underscore the growing interest in exploring sustainable alternatives for composite reinforcement and provide valuable insights into the potential applications and advantages of natural fiber-reinforced composites. The main objective of this study is to assess the influence of Alfa and date palm natural fibers on the fiber-matrix interface damage in hybrid biocomposite materials (Alfa-Date palm/epoxy, Alfa-Date palm/vinyl ester, and Alfa-date palm/polyimide) using a genetic approach. The results obtained from the genetic algorithm accurately represent the behaviour of the three materials, particularly regarding mechanical properties such as Young's Modulus, deformation, and stress at break. This investigation contributes significantly to the field of reinforced hybrid composite polymers by evaluating interface damage in biocomposites reinforced with different types of palm fibers, rather than focusing solely on individual fibers or specific biocomposite formulations. Additionally, the utilisation of advanced numerical techniques, including genetic algorithms, enhances the accuracy and reliability of the findings. Moreover, the study offers valuable insights into the mechanical behaviour of these biocomposites and their potential applications across various sectors. The study concludes that the matrix with the highest Young's modulus value at its fiber-matrix interface demonstrates the greatest strength. These findings align with recent results in the field of natural fiber-reinforced composite materials designed for diverse industries and eco-friendly applications.

## 2. METHODS AND NUMERICAL MODELS

### 2.1 Characteristics of the Materials Used Natural Fibers

Alfa fibers are extracted from raw alfa stems, by different processes such as mechanical, chemical or enzymatic extraction. They are used, in their raw state, for artisanal manufacturing of objects and in the extracted state in the paper industry. The esparto fibers are circular in sections with a central hollow area. Their density is low ( $1400 \text{ kg/m}^3$ ), they are biodegradable, and they come from a renewable source. The structure of esparto fiber is heterogeneous, consisting mainly of cellulose (40%–50%), lignin (17.71%–24%), hemicellulose (22.15% – 28%) and 5% wax [38-43].

The author in paper [39] presented a scanning electron microscope (SEM) images depicting an alfa fiber surface and its corresponding enlargement where they made an examination of a fiber resulting from the first mechanical extraction, they showed that the surface of this is no different from the gross surface area, this is unsurprising since this extraction simply consisted of reducing the section.

The date palm is also the symbol of resistance through its structure and unique characteristics. It is considered a thermophilic species. Its vegetation stops at  $10^\circ\text{C}$ . Maximum vegetation intensity is reached at temperatures of  $30\text{--}40^\circ\text{C}$ . The fruit ripening period corresponds to the hottest months of the year [44,45]. Date palm fiber is made up of 35% cellulose, 28% hemicellulose, 27% lignin and 7% of fat [46].

Agoudjil et al. [47] presented SEM micrographs showing the shape of a single fiber from the petiole of the DN date palm. They observed that the surface of the petiole fiber is cylindrical and irregular in shape with numerous filaments and cells, allowing adhesion between the fiber and probably a polymer matrix. A cross-section of a petiole fiber reveals a large number of simple hollow fibers collected and bonded by a layer.

The length, width and thickness of the Alfa and DPFs used are 20 mm,  $15 \mu\text{m}$  and  $10 \mu\text{m}$  respectively with a volume fraction of 14%. These fibers have undergone alkaline treatments (treatment with sodium hydroxide solution NaOH) also called mercerization of plant fibers, improves fiber-matrix adhesion by removing natural and artificial impurities which are accumulated on the surface of the fiber [38,39].

Table 1 presents the properties of Alfa and Date palm fibers that were used in the genetic simulation.

**Table 1.** Mechanical characteristics of the fibers used

Fibers treated	Young's modulus (GPa)	Deformation at break (%)	Stress at break (MPa)
Alfa [38]	28.43	2.43	474
Date palm [39,48]	7.5	5	203

### 2.2 Matrix Polymers

The epoxy resin family represents one of the highest-performing resin categories on the market. Epoxy resins generally outperform other resins in terms of mechanical properties and resistance to environmental aggression. The Epoxy resins polymerized at room temperature are generally used with hardening agents such as polyamines or polyamides, polymerization at room temperature leads to polymers having a lower glass transition temperature ( $T_g$ ), greater flexibility, and finally lower resistance to impact and thermal and electrical shock [49-51]. Epoxy resins can also be polymerized at high temperatures with hardening agents such as aromatic polyamines or anhydrides. Epoxy systems polymerized under these conditions generally demonstrate strong resistance to thermal degradation, high  $T_g$  and good mechanical properties. The most common cross-linkers for epoxy resins are amines, where the non-binding doublets of the amine function react with electrophilic oxirane rings and produce hydroxyl groups. This reaction is a poly addition, and therefore no reaction by-products are observed [51-53].

Vinyl ester resins are similar in their molecular structure to polyester resins, but they differ in the position of the reactive sites, which are located at the end of the chain. This gives vinyl ester resins greater ductility compared to polyester resins. Vinylesters also contain fewer ester groups, which lead to better water and chemical resistance in general. The vinyl ester resin used has the trade name Derakane 411-350. It is accelerated with a cobalt octoate dosage of 0.1% by the weight of the resin. It is catalysed with peroxide, whose trade name is Butanox M50, with a dosage of 1 to 1.75% by weight of resin. It contains 45% styrene. Their properties have been presented in Table 2 [54-56].

Polyimide (PI) is manufactured by a process called sintering into bars, plates and tubes. Polyimide is a polymer that does not melt at high temperatures. With excellent mechanical resistance, it stands out for its great dimensional stability and resistance to creep at temperatures above 260°C. Featuring good wear resistance, even without lubrication, coupled with high PV values, this material is ideal in friction and wear conditions, extending service life and reducing maintenance costs. With high purity and a low out gassing rate, it is ideal for aerospace applications in the vacuum and semiconductor industries. Thermoset polyimide resins are reactive prepolyimide resins with controlled molecular weights, prepared from the condensation of diamine aromatic dianhydrides in the presence of reactive final capping agents. Reactive caps are generally mono-anhydride compounds books with a thermally sensitive functional group capable of polymerization, copolymerization, or cross-linking. Examples of reactive end-capping agents include maleic anhydride (MA), 5-norbornene-2,3-dicarboxylic acid (NA), 4-Phenylethynylphthalic anhydride (PEPA) [57,58].

The main mechanical properties of these matrices are presented in Table 2.

**Table 2.** Mechanical characteristics of the matrix used

Matrix type	Young's modulus (GPa)	Deformation at break (%)	Stress at break (MPa)
Epoxy [54-56]	4.1	1.5	130
Vinyl ester [54-56]	3.2	4	81
Polyimide [58]	1.7	4.3	60

### 2.3 Weibull Probabilistic Formalism

The results of the study were obtained through a genetic simulation primarily based on the Weibull formalism. This approach was employed to calculate the damage at the interface of the three materials, utilizing genetic operators such as selection, crossing, and mutation. The Equation (1) Weibull, as presented in references [59-63], describes the damage to the matrix ( $D_m$ ) under uniform stress conditions:

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

where are:

$\sigma_f$  - applied stress;

$V_{eff}$  - the volume of the matrix;

$mand\sigma_0$  - Weibull parameters;

$V_0$  - the initial volume of the matrix.

The fiber is considered a constituent within an assembly of links, each possessing its individual breaking strength. Fracture frequency occurs when the weakest link in the assembly breaks. The failure of a fiber along its entire length is primarily attributed to its repetitions. This damage of fiber ( $D_f$ ) can be characterised by a law analogous to that of the matrix presented by Equation 2 [62-65]:

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

where are:

$\sigma_{max}^f$  - the maximum stress applied to the fiber;

$\sigma_{of}^f$  - The initial stress applied to the fiber;

$m_f$  - Weibull parameters;

$A_f = \pi \cdot a^2$  - Surface of the fiber circle;

$L_{equi}$  - The length of the fiber at equilibrium.

### 2.4 Volume Fraction

The volume fraction plays a pivotal role in determining the mechanical properties of a composite material. This ratio, expressed as the volume of a particular component relative to the total volume of the composite, directly influences theoretical models [66].

$$V_f = \frac{\text{Reinforcement volume}}{\text{Composite volume}} \% = \frac{v_f}{v_c} \quad (3)$$

$$V_m = \frac{\text{Matrix volume}}{\text{Composite volume}} \% = \frac{v_m}{v_c} \quad (4)$$

where are:

$V_f$  - Fiber volume fraction (volume content or rate),

$V_m$  - Matrix volume fraction (volume content)

$v_c$  - Composite volume,

$v_m$  - Matrix volume,

$v_f$  - Fiber volume.

$$V_m + V_f + V_v = 1 \quad (5)$$

By neglecting the vacuum, which generally does not exceed 3%, otherwise, the composite is unsuccessful, the Equation (5) will be resumed to:

$$V_m + V_f = 1 \quad (6)$$

## 3. RESULTS AND DISCUSSION

A genetic simulation was used to apply tensile stress to a representative elementary volume consisting of two fibers (Alfa and Date palm)

embedded in three types of matrices (Epoxy, Vinyl ester, and Polyimide). The application of tensile stress generates shear stress at the interfaces between the fibers and matrices.

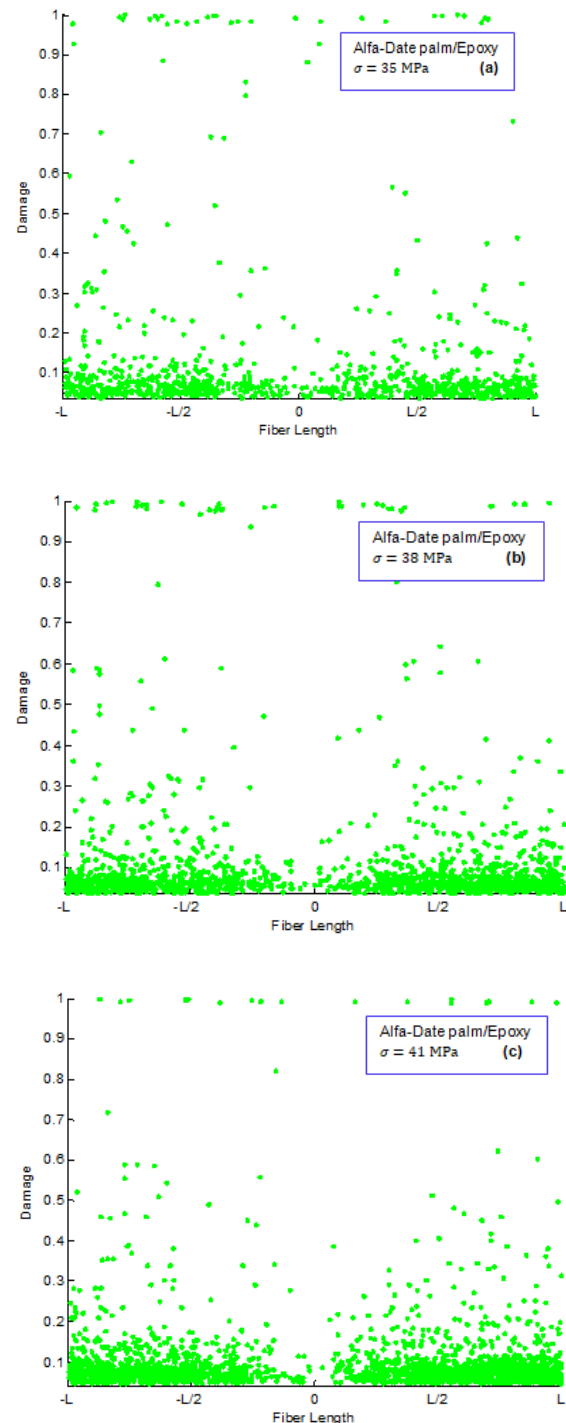
The effect of epoxy, vinyl ester, and polyimide polymers on fiber-matrix interface damage in hybrid biocomposite materials (Alfa-Date palm/epoxy, Alfa-Date palm/vinyl ester, and Alfa-Date palm/polyimide) was studied. The genetic approach is primarily based on the Weibull probabilistic model (Equations 1 and 2), using the genetic operator crossing to calculate the interface damages of three materials. The algorithm randomly generates an initial population of 9,600 individuals [62-64,67], which is subsequently enhanced through a set of genetic operators, including selection, crossing, and mutation. In this genetic modeling, an initial population of 9,600 individuals is generated, with each individual representing a random variable. This choice is primarily based on the theory of genetic algorithms and aims to achieve convergence towards the stopping criteria, using a mutation probability of 0.19. These optimal values are applied to the selection and mutation genetic operators. In each case; Young modulus of each fiber and others parameters are used (Tables 1 and 2). The population consists of chromosomal genes representing the following variables: the shear stress (35, 38, 41, 44 and 47 MPa), the Young's modulus, the modulus of the matrix shear, the fiber diameter and the half distance R. The selection used is of roulette type and with a selected mutation value equal to 0.19. The calculations were performed using the optimal iteration values of the damage at the interface, enabling the optimization of the results of the genetic model.

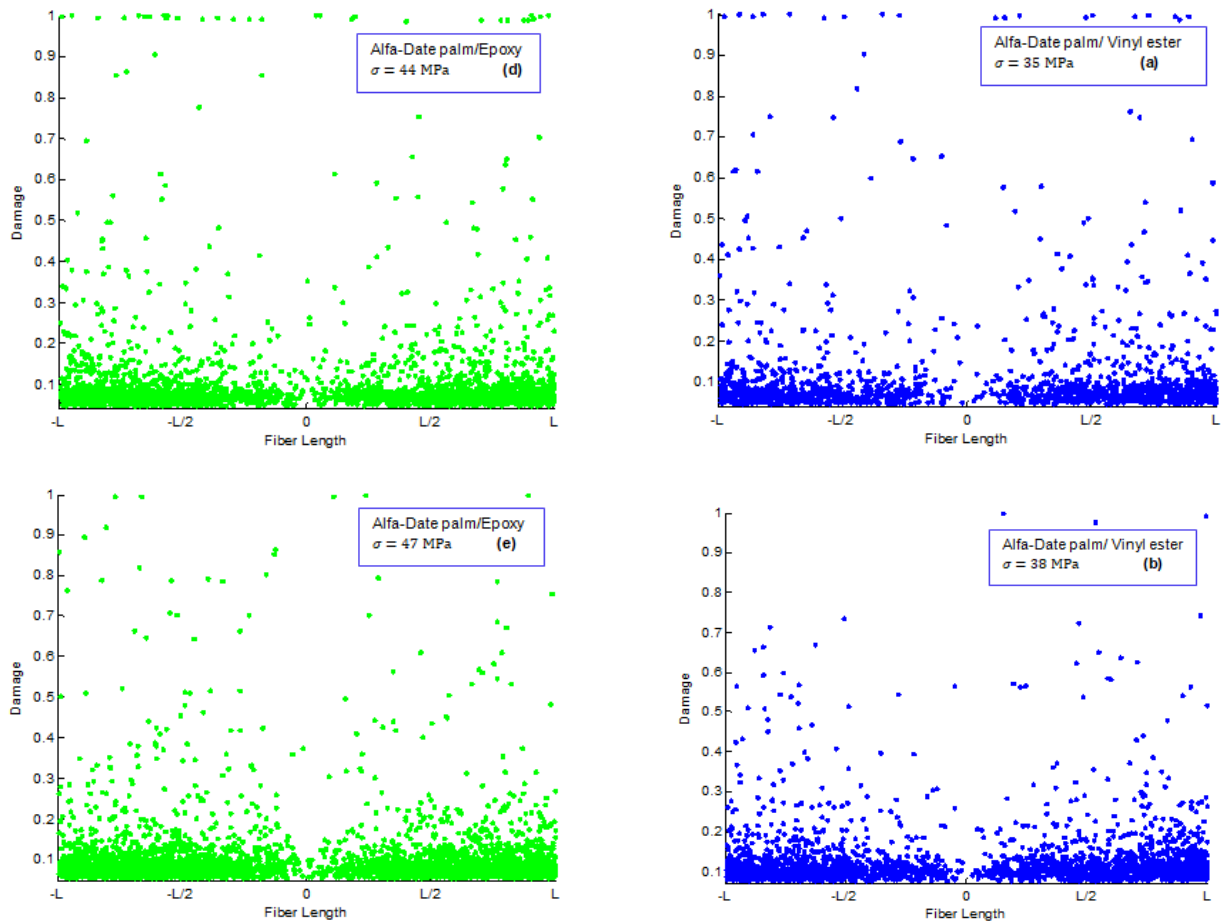
Tensile stress was applied to a representative elementary volume consisting of two fibers, Alfa and Date palm, embedded in one of the three matrices: Epoxy, Vinyl ester, or Polyimide. This stress generated shear stress at the fiber-matrix interface, which is the subject of this study.

### 3.1 Alfa-Date Palm/Epoxy Hybrid Biocomposite Material

The behaviour of the interface under increasing mechanical stress values (35, 38, 41, 44, and 47 MPa) is depicted in Fig. 1, which comprises five subfigures. In Fig. 1(a), it is observed that damage to the interface initiates at a value of 0.086 for a mechanical shear stress of 35 MPa and increases

linearly with the stress. Subsequent Figs. 1(b) through (e) show the progression of damage as the stress increases, reaching a maximum value of 0.112 for a stress of 47 MPa, as depicted in Fig. 1(e). The presence of symmetry in the damage at the middle of the fiber is noted. The random variables, depicted graphically by the green dots and the green cloud, indicating that the damage is concentrated at the ends of the fiber, as previously demonstrated by Cox's micromechanical model, in comparison to the middle of the fiber [68].

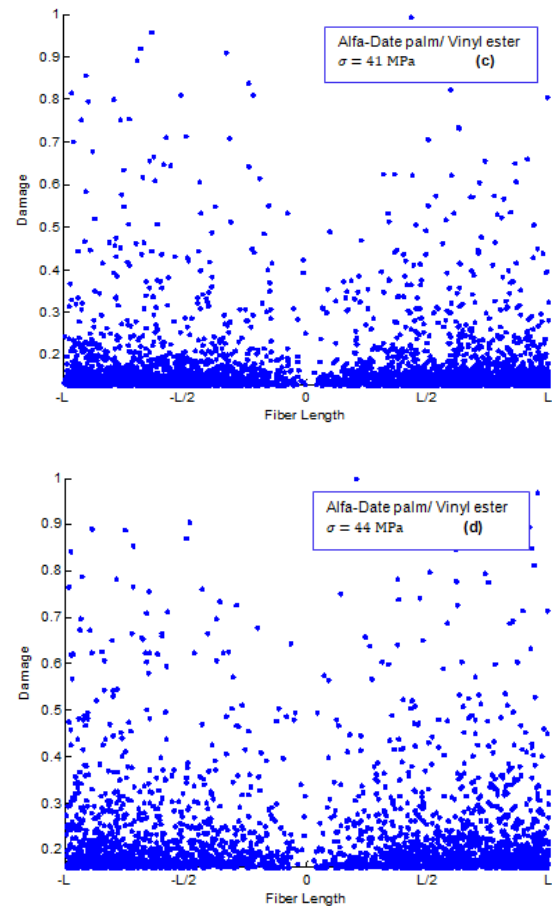


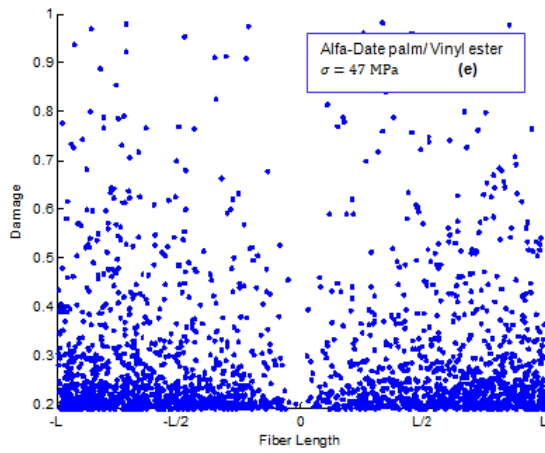


**Fig. 1.** Interface fiber-matrix damage level of Alfa-Date palm/Epoxy: (a)  $\sigma=35$  MPa, (b)  $\sigma=38$  MPa, (c)  $\sigma=41$  MPa, (d)  $\sigma=44$  MPa, (e)  $\sigma=47$  MPa

### 3.2 Alfa-Date Palm/Vinyl Ester/ Hybrid Biocomposite Material

Fig. 2 presents the shear damage levels under varying mechanical stress conditions, with each subfigure depicting different stress values (35, 38, 41, 44, and 47 MPa). In Fig. 2(a), the onset of shear damage is observed at a damage value of  $D = 0.094$  when  $\sigma = 35$  MPa, and this damage level increases gradually as the stress rises. Subsequent Fig. 2 (b) through (e) show the progression of damage with increasing stress, reaching a maximum value of  $D = 0.246$  for a stress of  $\sigma = 47$  MPa, as illustrated in Fig. 2(e). Symmetry in the distribution of damage along the fiber's length is evident. The random variables, depicted graphically by the blue dots and the blue cloud, indicate that damage tends to concentrate at the fiber ends, consistent with Cox's micromechanical model.

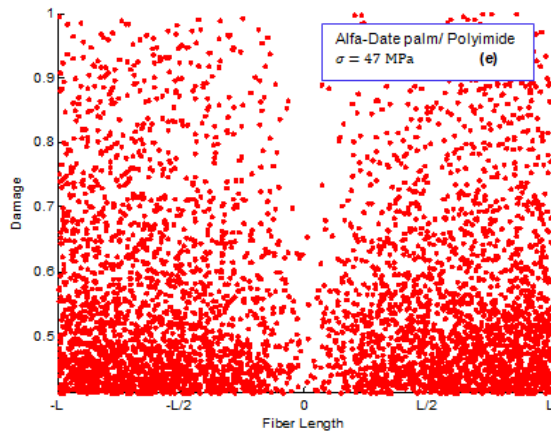
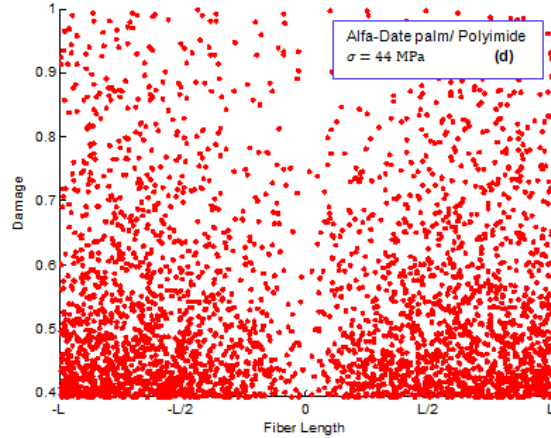
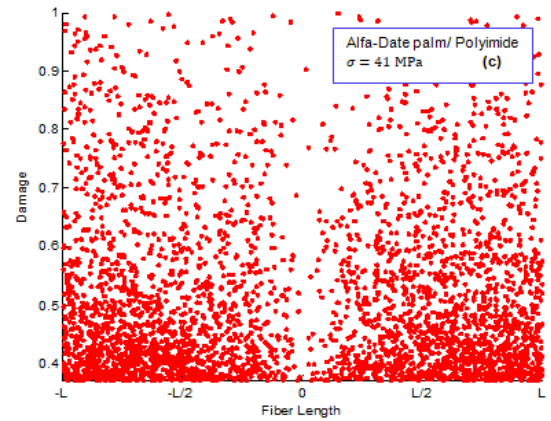
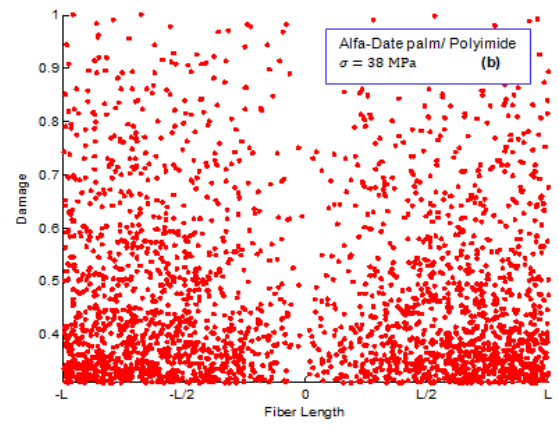




**Fig. 2.** Interface fiber-matrix damage level of Alfa-Date/Vinyl ester: (a)  $\sigma=35$  MPa, (b)  $\sigma=38$  MPa, (c)  $\sigma=41$  MPa, (d)  $\sigma=44$  MPa, (e)  $\sigma=47$  MPa

### 3.3 Alfa-Date Palm/Polyimide Hybrid Biocomposite Material

Fig. 3 illustrates the shear damage levels under varying mechanical stress conditions, with each subfigure depicting different stress values (35, 38, 41, 44, and 47 MPa). In Fig. 3(a), the initiation of shear damage is observed at a damage value of  $D = 0.282$  when  $\sigma = 35$  MPa, and this damage level increases progressively as the stress increases. Subsequent Fig. 3 (b) through (e) demonstrate the progression of damage with increasing stress, culminating in a maximum value of  $D = 0.489$  for a stress of  $\sigma = 47$  MPa, as depicted in Fig. 3(e). Symmetry in the distribution of damage along the fiber's length is apparent. The random variables, depicted graphically by the red dots and the red cloud, suggest that damage tends to concentrate at the fiber ends, consistent with Cox's micromechanical model, as opposed to the middle of the fiber.



**Fig. 3.** Interface fiber-matrix damage level of Alfa-Date palm/Polyimide: (a)  $\sigma=35$  MPa, (b)  $\sigma=38$  MPa, (c)  $\sigma=41$  MPa, (d)  $\sigma=44$  MPa, (e)  $\sigma=47$  MPa



The interface has properties different from those of the constituents (the fiber and the matrix), particularly with regard to mechanical properties. However, it plays a very important role in the transfer of forces between the matrix and the fiber. The quality of the interface, therefore, determines the final performance of the composites. For this reason, the mechanical characterization of this strategic area is essential. The characterization of the interface consists of determining its mechanical properties as a function of the fiber/matrix materials used and possibly, as a function of the modifications (treatment) that they undergo (chemical matrix formulation or fiber surface treatment, ageing, etc.) [69]. The genetic results faithfully showed the real behaviour of the three materials according to their mechanical properties, notably the values of Young's Modulus, deformation and stress at break (see Table 2). The study concludes that the epoxy matrix exhibits the highest Young's modulus value, resulting in the greatest strength at its fiber-matrix interface. These findings align well with the results reported by the authors [57,58,68,69].

#### 4. CONCLUSION

The present study aims to investigate the impact of alfa and date palm natural fibers as reinforcement for epoxy, vinyl ester, and polyimide polymers (Alfa-Date palm/epoxy, Alfa-Date palm/vinyl ester, and Alfa-date palm/polyimide) using a genetic approach. By employing a genetic simulation grounded in the Weibull formalism, the study intricately assessed the damage at the fiber-matrix interface across the examined materials. The genetic algorithm results accurately captured the intrinsic behaviour of these materials, highlighting key mechanical properties such as Young's Modulus, deformation, and stress at break. Moving forward, there are several avenues for further research in this field. Future studies could explore the application of the methods and techniques utilized in this study to predict mechanical characteristics and apply them to other types of composite materials. Additionally, considering the extensive repetition cycles and long monitoring periods involved in these experiments, the incorporation of fuzzy logic and neural networks could enhance predictive accuracy. Furthermore, it is important to consider the implications of this research on the costs of operation and maintenance of materials made from composite materials. Additionally, further

investigation into the application of additives mentioned in the paper and their impact on the performance and longevity of composite materials could yield valuable insights. Overall, the conclusions drawn from this study align with contemporary advancements in natural fiber-reinforced composite materials tailored for diverse industries and eco-friendly applications, highlighting the potential for future research and practical applications in this field.

#### Conflicts of Interest

The authors declare no conflict of interest.

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