Effect of Fiber-matrix Interface Decohesion on the Behavior of Thermoset and Thermoplastic Composites Reinforced with Natural Fibers: A Comparative Study

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In this article, a comparative study was carried out on two types of thermosetting and thermoplastic matrices to study the effect of the fiber-matrix interface damage on the behavior of thermosetting and thermoplastic composites reinforced by the same natural alfa and wood fibers. The genetic modeling was based on the probabilistic formalism of Weibull. The results have been compared with those obtained by the nonlinear acoustic technique, the two results found to coincide perfectly. The numerical simulation also shows a good concordance with the real behavior of the materials studied, and shows that thermosetting composites are the most resistant to applied thermal stress by 21% compared to thermoplastic composites. Statistical analysis demonstrates that the correlation coefficient values found are very close to 1 (0.964 and 0.973), these values are very satisfactory, and confirm that the results obtained by the genetic model and the nonlinear acoustic technique are in very good agreement with the statistical analysis data. The experimental work presented by Antoine Le Duigou et al. and the work of Bodros et al. have shown that the use of natural fibers greatly improves the mechanical properties of composite materials.

Keywords: alfa fiber, wood fiber, phenolic composite, polyetherimide (PEI) composite, interface, damage.

1. INTRODUCTION

The composite materials are made up of the arrangement of several materials (fibers: synthetic or natural, matrices: thermoset or thermoplastics, fillers, etc.) that are immiscible to form new materials with better mechanical and physicochemical properties. They thus have many advantages allowing them to compete directly with so-called conventional materials such as metals or alloys.

The automotive industry is looking to replace glass and carbon fibers with plant fibers. Indeed, in recent years research on composite polymers based on plant fibers is growing, particularly in the automotive, aeronautics, sports, construction, and packaging industries [1, 2]. The challenge is to develop sustainable technologies and to manufacture more environmentally acceptable materials, the use of natural fibers in the automobile is due to their low density: the density of flax is 1.4 g/cm³ while that of fiberglass is 2.5 g/cm³ [1, 2]. The natural fibers reduce energy consumption in the process, reduce the cost of manufacturing the composite and improve the mechanical properties of these materials [1, 3, 4]. To improve the mechanical properties of composites materials, the surface treatment of these materials reinforced with natural fibers is more than necessary to remove amorphous constituents such as hemicellulose and lignin, in order to obtain more cellulose possible with higher crystallinity levels. Superior mechanical properties and resistance to thermal degradation may be associated with a cellulose content at the fiber level [5, 6]. Another important characteristic concerning the reinforcement of a polymer with natural fiber is the thermal degradation of fibers, which is on average around 200 °C, limiting parameter to the treatment temperature. Fiber size, morphology, and fiber orientation are also characteristics that directly influence the final properties of composites [5, 7, 8]. The plant fibers generally have good mechanical properties, but their use is limited because of their hydrophilic nature and their low thermal stability; the fiber-matrix interface, through the incorporation of a coupling agent, will depend not only on the nature of the polymer matrix but also on the type of fibers. The biggest difference between a composite and a biocomposite is its end of life. Indeed, biocomposite must comply with standards of biodegradability and / or recyclability [1,10].

Our contribution is to model the behavior of the fiber-matrix interface with the genetic approach of four biocomposite materials by using two natural fibers alfa and wood to reinforce two matrices of different types; thermoset matrix (phenolic) and a thermoplastic matrix (polyetherimide (PEI):), these materials were under the effect of the same mechanical tensile stress ranging from 300 to 700 N/m² and the same thermal stress ranging from 300 to 1300 N/m².

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2. METHODS AND MODELS

In this section, we will present the analytical and numerical models and the physical properties of fibers and matrices used in our computational model as well as the genetic approach which we have chosen to model the effect of thermal stress on the fiber-matrices interface of biocomposite materials alfa/polyetherimide (PEI), alfa/phenolic, wood/polyetherimide (PEI) and wood/phenolic.

2.1. Cox's micromechanical model

Cox developed his model by applying tensile stress to a representative elementary volume (R.E.V) [11]. When the fiber is stressed along its axis, the forces are transmitted to the fibers via the shear forces at the interface. The interface shear stress (IFSS – Inter Facial Shear Strength) is expressed by Eq. 1 [11]:

$$\tau = \frac{E_f * a * \varepsilon}{2} \beta \left(\tanh \left(\beta * \frac{1}{2} \right) \right), \tag{1}$$

$$\beta = \frac{2G_m}{E_f * r_f^2 * \ln(\frac{R}{r_f})}$$

where ε is the deformation; a is the radius of the fiber; R is the distance between fibers; G_m is the shear modulus of the matrices; E_f is the Young's modulus of the fiber; τ is the shear stress of the interface; r_f is the distance between fiber and the matrices.

2.2. The theory of nonlinear acoustic for materials

Nonlinear acoustics is emerging as a very promising new avenue for the non-destructive testing and evaluation of materials and structures. Indeed, if the acoustic non-linearities can be very high for inhomogeneous materials such as rocks, [12 – 17], they increase significantly in the presence of damage. Nonlinear phenomena associated with damage are also observed for much more homogeneous materials such as metal alloys. Thus, many potential applications exist in nonlinear acoustics both for the characterization of macroscopic cracks and for the evaluation of diffuse damage due to the presence of microcracks [12, 15 – 17].

The classical nonlinear elastic behavior of materials is usually described by the addition of a non-linear term β in Hooke's law which is then written in Eq. 2.

$$\sigma = E\varepsilon(1 + \beta\varepsilon), \tag{2}$$

where σ and ε are respectively the stress and strain in a given direction x, resulting from the ultrasonic field. E is Young's modulus and β the parameter of non-linearity in the x direction. In the case where the material is damaged, E and β are effective sizes. E and β can be determined from acoustic measurements. Young E's modulus is identified by considering the propagation speeds in longitudinal and transverse waves possibly in variable incidence if the material is anisotropic [12]. The nonlinearity parameter that can be expressed in function of elasticity constants (linear) of order 2 and (non-linear) of order 3 is determined from acoustoelastic measurements, that is to say from variations

in ultrasound speeds as a function of applied static elastic stress [12, 19].

2.3. Thermal stresses

Thermal stresses resulting from the differential expansion of the fibers and the matrices during cooling after preparation of the composite at high temperature [20]. They are presented by the Eq. 3:

$$\sigma_f^T = E_f \frac{a}{a+1}(M_2 - M_0), \tag{3}$$

with:

$$M_0(T) = \int_{T_0}^{T_e}(\alpha_m - \alpha_f)dT;$$

$$M_2(T) = \int_{T_e}^{T}(\alpha_m - \alpha_f)dT,$$

where T_0 is the room temperature; T_e is the temperature of development; T is the test temperature; α_f and α_m are the expansion coefficients of the fiber and matrices.

2.4. Weibull approach

Our results were obtained by a genetic simulation based mainly on the Weibull formalism to calculate the damage at the interface of the four materials using the genetic operator's selection, crossing and mutation.

Damage to the matrices, when the stress is uniform, is given by Eq. 4 Weibull [21 – 24]:

$$D_m = 1 - exp\left\{-\frac{V_{eff}}{V_0}(\frac{\sigma_f}{\sigma_0})^m\right\}, \tag{4}$$

where σ_f is the applied stress; V_{eff} is the volume of the matrices; m and σ_0 are Weibull parameters; V_0 is the initial volume of the matrices.

The fiber is assumed to be a component of an assembly of links each having its breaking strength. Frequency fracture occurs when the weakest link breaks. The discharge of fiber over its entire length is due mainly to its repetition. This rupture can be described by a law similar to that of the matrices [21 – 25].

$$D_f = 1 - exp\left\{-A_f * L_{equi} * (\frac{\sigma_{max}^f}{\sigma_{of}})^{m_f}\right\}, \tag{5}$$

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

2.5. Mechanical and physical properties of fibers and matrices used

2.5.1. Phenolic thermoset matrix

Phenolic resins are thermosetting resins produced by condensation of aldehydes with phenols. However, substituted phenols and higher aldehydes are used to produce phenolic resins with specific properties, including specific reactivities and functionalities [26 – 28]. The sequences must be considered in the preparation of traditional phenolic resins: addition of formaldehyde to the phenol, chain growth or formation of the pre-polymer (resin), and crosslinking or curing reaction. There are essentially three types of phenolic resins: traditional resoles,

novolacs, and newer polybenzoxazines [29]. The physical and mechanical properties of the phenolic resin used in the genetic modeling are presented in Table 1.

Milanese et al. described phenolic matrix by the SEM (scanning electron microscope) technique the fracture of the phenolic resin by traction: cracks through heterogeneous material and microvoids (500^X) and cracks originating from microvoids (1000^X) [36].

2.5.2. Polyetherimide (PEI) thermoplastic matrix

Polyesterimide (PEI) resins are used in the impregnation industry to provide mechanical and electrical support to the coils of rotating machines. Unsaturated polyesterimides are used due to their good mechanical properties and electrical insulation at high temperatures. [37, 38]. The properties of the polyesterimides resin used in the genetic modeling are presented in Table 1.

Jianget al. [41] described by the SEM (scanning electron microscope) technique the freeze fracture surface of the PEI FDM parts with 90° raster at: 350°C, 360°C, 370°C, and 380°C. PEI: polyetherimide; fused deposition modeling (FDM).

2.5.3. Alfa fiber

The natural fiber Alfa (from the Arabic halfa), or Sparta (Stipatenacissima L.) is a plant, with cylindrical stem, perennial herbaceous of the poaceae family, native to arid and semi-arid regions Mediterranean excluding desert sectors: North Africa (Algeria, Tunisia, Morocco, and Libya) and Southern Europe (Spain and Italy) [42]. This species, with tapering leaves, grows in tufts about one meter high, forming large aquifers in areas of medium aridity. The territorial distribution known to date is estimated Dallel [42, 43]: Algeria: 4.000.000 ha; Morocco: 3.186.000 ha; Tunisia: 600.000 ha; Libya: 350.000 ha; Spain: 300.000 ha.

The approximate chemical composition (mass %) of Alfa is as follows: cellulose (44 – 47 %), hemicellulose (22 – 35 %), lignin (19 – 24 %) and ash (2 – 5 %). The plant is of ecological and economic interest. Indeed, it does not need insecticides or pesticides harmful to the environment and it consumes very little water. Researchers have always chosen to treat Alfa's plant with soda, which is one of the cheapest and easiest treatments to perform [42, 44, 45]. The mechanical properties of Alfa fibers used in the genetic modeling are presented in Table 2.

Ajouguim et al. [47] presented by the SEM (scanning electron microscope) technique the Alfa fiber after alkali treatment in 10 hours. The physical surface treatment was carried out by using dispersing agents and additives (waxes, paraffin) which are the most widely used in the field of WPC composites. The wood fiber used in our polymers is medium density panel (MDF) type in the form of sawdust panel factory. In general, these panels are made up of mixtures of mainly conifers (spruce, fir, pine) and hardwoods [51].

Choi et al. and Moon et al. [52, 53] described in detail by the SEM (scanning electron microscope) technique image of the wood fiber.

3. RESULTS AND DISCUSSION

3.1. Study by genetic algorithm and nonlinear acoustic technique

In the present study, we calculated the damage to the interface of thermosetting and thermoplastic composite (alfa/polyetherimide (PEI), alfa/phenolic, wood/polyetherimide (PEI) and wood/phenolic), under the effect of mechanical and thermal stress varying from 300 to 700 N/m² and from 300 to 1300 N/m² respectively. To this end, we used genetic modeling to evaluate this damage at each iteration. Our contribution in this article is to cross between matrix and fiber damage defined by the Weibull model (Eq. 4, Eq. 5) to generate interface damage. The objective function is defined by the Cox model. Our algorithm will randomly generate an initial population of 2800 individuals, then we improve each initial population by a set of genetic operators (selection, crossing and mutation) and in each case; we have used the values of the variables shown in Table 1 and Table 2. The damage to the interface is determined at each iteration by the crossing between the matrix damage D_m fiber damage D_f. The population consists of chromosomal genes. To exploit the maximum mechanical and thermal (Eq. 3) and to see the progress of our genetic algorithm, the selection used is of roulette type and with a mutation value selected equal to 0.25. The calculations were performed by the optimal iteration values found of the damage at the interface, which allowed the optimization of the results of our genetic model. The results of genetic modeling relate to the effect of mechanical and thermal stress on the resistance of thermosetting and thermoplastic composite materials are represented by black and blue figures.

Table 1. Physical and mechanical properties of the phenolic resin and polyestersimide resin used in our genetic modeling [30 – 35, 39, 40]

Properties	Density, g/cm^3	Tensile modulus, GPa	Tensile strength, MPa	Elongation at break – tensile mode, %	Compression strength, MPa	Cure temperature, °C
Phenolic matrix	1.29	2.8	35	1.5	210	25 – 300
Polyetherimide matrix	1.27	3.1	86	12	110	210

Table 2. Mechanical properties of Alfa and wood fibers [42, 46]

Properties	Density, g/cm^3	Elongation at break – tensile mode, %	Tensile strength, MPa	Avg. fiber length, mm	Young modulus, GPa
Alfa fiber	1.4	1.5	134	2.9	15
Wood fiber	1.5	/	666	2.9	26

To validate our found results, we calculated the effect of the mechanical and thermal stress on the interface damage of the materials studied, using the nonlinear parameter *B* defined by Eq. 2. These results have been represented by the figures in red.

3.1.1. Interface damage of wood/polyetherimide (PEI)

For the wood/polyetherimide material, the effect of thermal and mechanical stress is represented by Fig. 1, Fig. 2 and Fig. 3, where it was found that the value of the damage at the interface was 0.4 for a mechanical and thermal stress of 300 N/m² and reaches a value of 0.46 for a mechanical stress of 700 N/m² and a thermal stress of 1100 N/m².

Fig. 1. Effect of thermal stress on damage interface for $\sigma = 300$ N/m²

The results gained are in good agreement with those found by the nonlinear acoustic technique represented in the same figures with red color.

Fig. 2. Effect of thermal stress on damage interface for $\sigma = 500$ N/m²

3.1.2. Interface damage of alfa/polyetherimide (PEI)

For the alfa/polyetherimide (PEI) material, the effect of thermal and mechanical stress is represented by Fig. 4, Fig. 5 and Fig. 6, where it was found that the value of the damage at the interface was 0.22 for a mechanical and thermal stress of 300 N/m² and reaches a value of 0.3 for a

mechanical stress of 700 N/m² and a thermal stress of 1100 N/m². The results obtained are in a good agreement with the results found by the nonlinear acoustic technique represented in the same figures with red color.
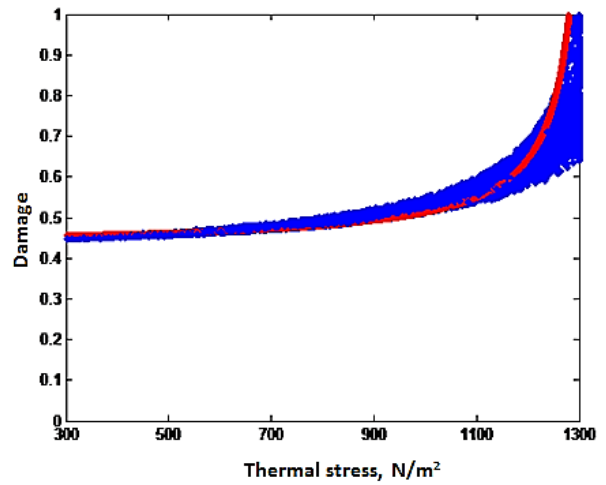

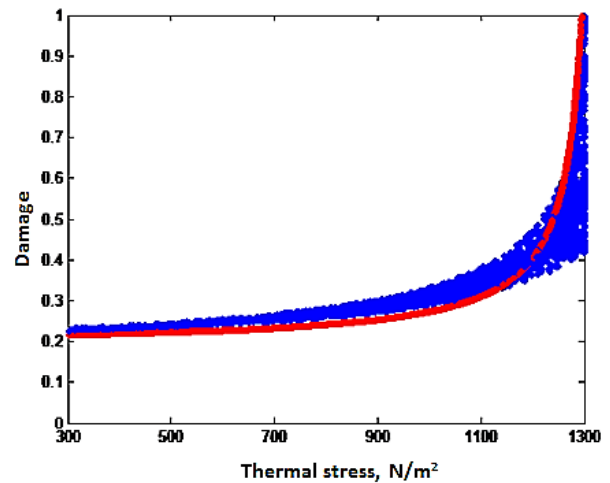**Fig.3.** Effect of thermal stress on damage interface for $\sigma = 700$ N/m²

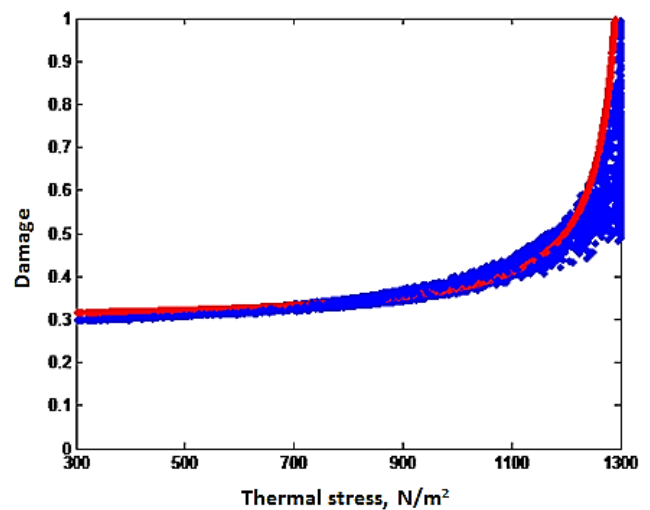**Fig. 4.** Effect of thermal stress on damage interface for $\sigma = 300$ N/m²

Fig. 5. Effect of thermal stress on damage interface for $\sigma = 500$ N/m²
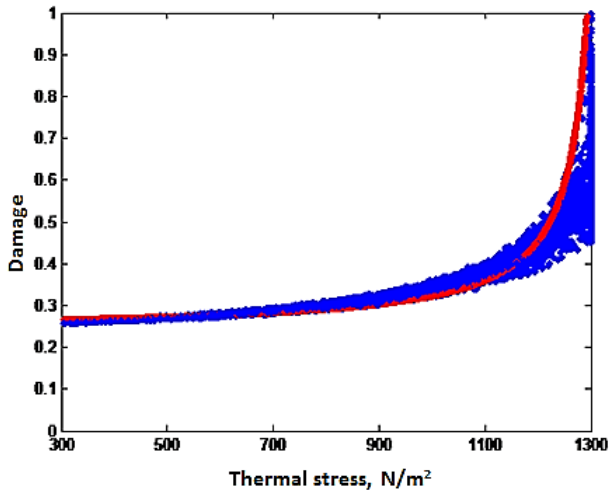
Fig. 6. Effect of thermal stress on damage interface for $\sigma = 700$ N/m^2

3.1.3. Interface damage of wood/phenolic

For the wood/phenolic material, the effect of thermal and mechanical stress is represented by Fig. 7, Fig. 8 and Fig. 9, where it was found that the value of the damage at the interface was 0.1 for a mechanical and thermal stress of 300 N/m² and reaches a value of 0.15 for a mechanical stress of 700 N/m² and a thermal stress of 1100 N/m².
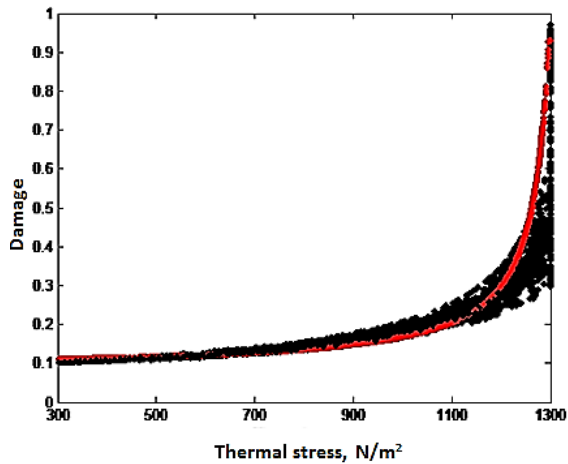

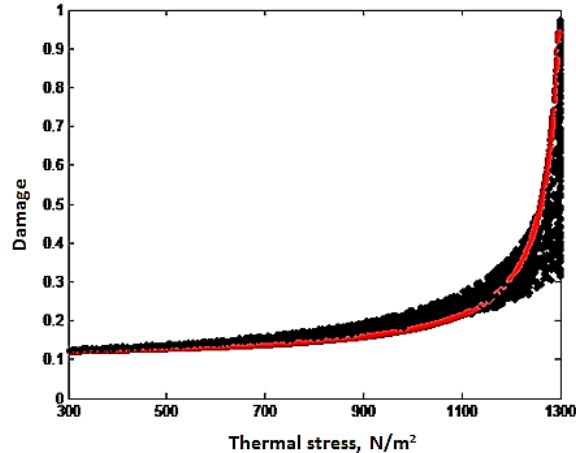**Fig. 7.** Effect of thermal stress on damage interface for $\sigma = 300$ N/m^2

Fig. 8. Effect of thermal stress on damage interface for $\sigma = 500$ N/m^2

These results are in good agreement with the results found by the nonlinear acoustic technique represented in the same figures with red color.
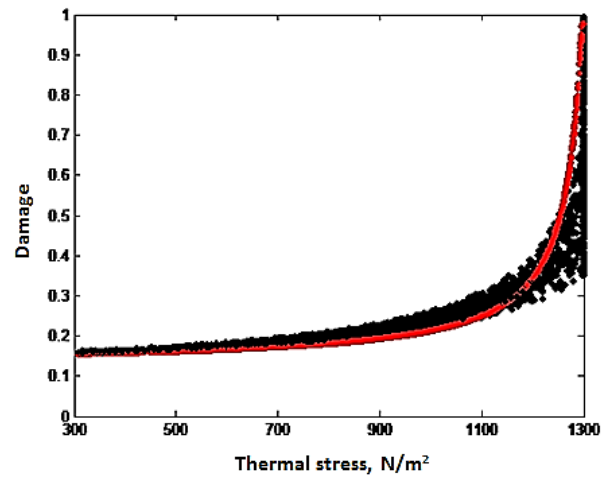

Fig. 9. Effect of thermal stress on damage interface for $\sigma = 700$ N/m^2

3.1.4. Interface damage of alfa/phenolic

For the alfa/phenolic material, the effect of thermal and mechanical stress is represented by Fig. 10, Fig. 11 and Fig. 12, where it was found that the value of the damage at the interface was 0.04 for a mechanical and thermal stress of 300 N/m² and reaches a value of 0.09 for a mechanical stress of 700 N/m² and a thermal stress of 1100 N/m².

The results are in good agreement with the results found by the nonlinear acoustic technique represented in the same figures with red color.

The results presented by the two simulation models (genetic and nonlinear acoustics); that they are in good agreement with each other, confirm that the fiber-matrix interface of composite materials based on the thermoset phenolic matrix is more resistant to mechanical and thermal stresses applied by 21 % compared to thermoplastic polyetherimide (PEI) matrix, and that its results show the real behavior of the four materials studied, namely that the thermoset resin is made up of linear chains cross linked between them. The chains are linked in space by strong bonds of the covalent type [54].
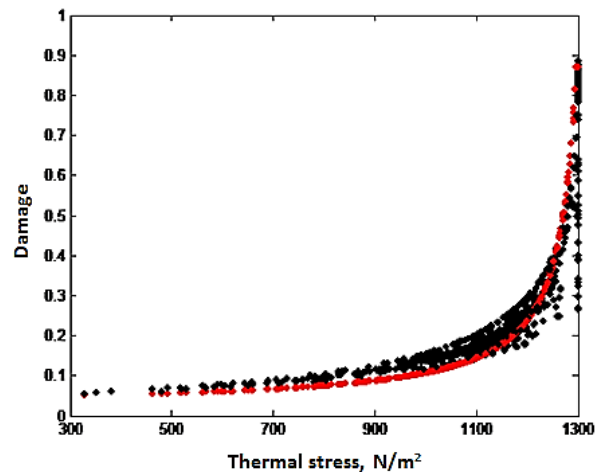

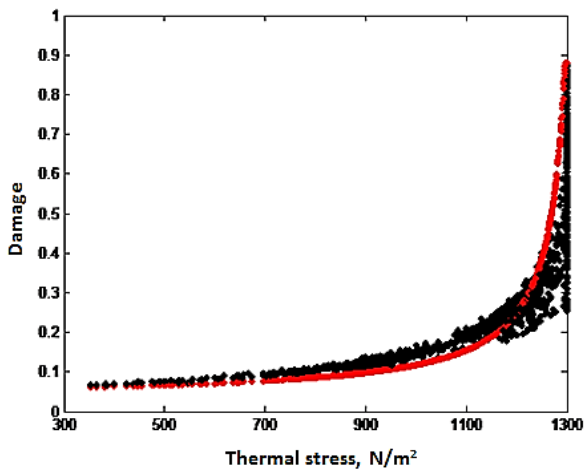**Fig. 10.** Effect of thermal stress on damage interface for $\sigma = 300$ N/m^2

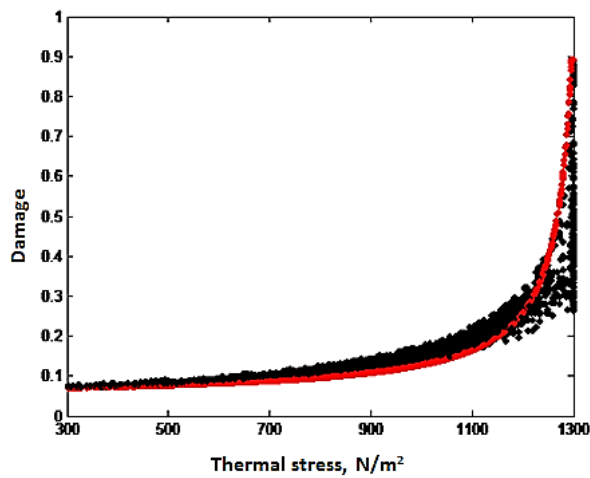**Fig. 11.** Effect of thermal stress on damage interface for $\sigma = 500$ N/m^2

Fig. 12. Effect of thermal stress on damage interface for $\sigma = 700$ N/m^2

We are therefore in the presence of an insoluble and infusible three-dimensional network, unlike the thermoplastic resin; it is made up of linear or branched chains with covalent bonds. These chains are linked together by weak bonds of the Vander Waals and hydrogen type [54]. The experimental work presented by Antoine Le Duigou et al [55] and the work of Bodros et al [56] have shown that the use of natural fibers greatly improves the mechanical properties of composite materials. The values of the obtained correlation coefficients are very close to 1 (0.964 and 0.973), and they are very satisfactory and confirm that the results found by the genetic model and the nonlinear acoustic technique coincide perfectly with the statistical analysis data.

3.2. Convergence study of the two approaches by a statistical analysis

To study the convergence of the genetic model and the nonlinear acoustic technique, we have used a statistical analysis based on the calculation of the correlation coefficient between the interface damage (Y_i) obtained by the genetic approach and the nonlinear acoustic technique and the different values of the thermal stress(X_i). The results found are shown in Table 3.

$$\tau_1(x;y) = \frac{cov(x;y)}{\sigma_x\sigma_y} = \frac{\sigma_{xy}}{\sigma_x\sigma_y} = \frac{1537}{2126.03*0.75} = 0.964; \qquad (6)$$

$$\tau_2(x;y) = \frac{cov(x;y)}{\sigma_x\sigma_y} = \frac{\sigma_{xy}}{\sigma_x\sigma_y} = \frac{1417.3}{2126.03*0.685} = 0.973. \qquad (7)$$

The Eq. 6 and Eq. 7 were used to calculate the correlation coefficients for alfa / polyetherimide (PEI). The values found (0.964 and 0.973) are very acceptable, and coincide perfectly with those obtained by the genetic model and the nonlinear acoustic technique.

4. CONCLUSIONS

In this study, the effect of thermal stress on the fiber-matrices interface of thermoset and thermoplastic composite materials reinforced with alfa and wood natural fibers (alfa/polyetherimide (PEI), alfa/phenolic, wood/polyetherimide (PEI) and wood/phenolic) has been investigated and studied using genetic algorithm and nonlinear acoustics approach simulation. The results presented by the two numerical simulation models that they are in good agreement with each other, confirm that the fiber-matrix interface of composite materials based on the thermoset phenolic matrix are more resistant to mechanical and thermal stresses, and show that thermosetting composites are the most resistant to applied thermal stress by 21 % compared to thermoplastic composites.

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Table 3. The values of interface damage obtained by genetic algorithm and nonlinear acoustic technique

Materials										
Alfa/polyetherimide (PEI)		Interface damage obtained by genetic algorithm								
	X_i	300	400	500	600	700	800	900	1000	1100
	Y_i	0.22	0.23	0.25	0.26	0.26	0.27	0.28	0.3	0.3
		Interface damage obtained by nonlinear acoustic technique								
	X_i	300	400	500	600	700	800	900	1000	1100
	Y_i	0.21	0.22	0.22	0.23	0.24	0.25	0.26	0.27	0.27

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