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Machine Learning-Assisted Tensile Modulus Prediction for Flax Fiber/Shape Memory Epoxy Hygromorph Composites

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Abstract: Flax fiber/shape memory epoxy hygromorph composites are a promising area of research in the field of biocomposites. This paper focuses on the tensile modulus of these composites and investigates how it is affected by factors such as fiber orientation (0° and 90°), temperature (20 °C, 40 °C, 60 °C, 80 °C, and 100 °C), and humidity (50% and fully immersed) conditions. Machine learning algorithms were utilized to predict the tensile modulus based on non-linearly dependent initial variables. Both decision tree (DT) and random forest (RF) algorithms were employed to analyze the data, and the results showed high coefficient of determination R^2 values of 0.94 and 0.95, respectively. These findings demonstrate the effectiveness of machine learning in analyzing large datasets of mechanical properties in biocomposites. Moreover, the study revealed that the orientation of the flax fibers had the greatest impact on the tensile modulus value (with feature importance of 0.598 and 0.605 for the DT and RF models, respectively), indicating that it is a crucial factor to consider when designing these materials.

Keywords: composites; machine learning; decision tree; random forest; mechanical properties

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1. Introduction

Composite materials are becoming increasingly popular in various engineering fields due to their exceptional properties [1–3], including high strength-to-weight ratio [4–6], resistance to corrosion [4], and long-lasting durability [7]. These materials are being used in a wide range of applications, from aircraft structures [8] to medical implants [9] and wind turbine blades [10]. In recent years, there has been a growing interest in utilizing natural fibers as replacements for synthetic fibers in polymer composites. This is because natural fibers are a renewable resource, abundant, recyclable, biodegradable, lightweight, and cost-effective, with good mechanical properties [11–16].

Flax fibers, which have been utilized for centuries in various applications such as clothing, paper, and rope due to their natural origin, are an ideal reinforcement component for composite materials. They are extracted from the stems of the flax plant and are known for their robustness, rigidity, and longevity, making them an excellent option where weight reduction and toughness are significant factors. Furthermore, flax fibers are a sustainable and biodegradable option, making them a compelling substitute for synthetic fibers [17].

Shape memory epoxy resins are a type of polymer that can return to their original shape after being exposed to specific triggers such as light or heat. This behavior is due to the presence of shape memory polymers (SMPs) which can undergo reversible shape changes upon the influence of an external stimulus. In the case of shape memory epoxy hygromorph composites, the epoxy resin matrix contains SMPs, enabling the material to display shape memory characteristics [18]. The exceptional responsiveness to stimuli and adjustable stiffness of shape memory polymers (SMPs) have long been a subject of interest, leading to notable advancements in fields such as aerospace, civil engineering, and others. Epoxy resin (EP) is a promising material for such applications owing to its impressive mechanical properties, resistance to fatigue, and radiation endurance [19]. Epoxy resins were one of the initial materials to gain widespread use in industrial applications [20].

The use of steel plates reinforced with epoxy resin adhesive to enhance the load-bearing capacity of prestressed reinforced concrete bridges has become increasingly popular [21]. Kuo et al. have investigated the development of injection molding tools utilizing epoxy resin [22,23].

Flax fiber/shape memory epoxy hygromorph composites are a type of composite material that incorporates flax fiber composites with an epoxy shape memory polymer (SMP) matrix [24]. Hygromorphic materials are a unique class of smart materials that utilize the humidity of their surroundings to activate their properties and ability to morph [11]. Flax fiber/shape memory epoxy hygromorph composites are desirable for various applications due to the combination of flax fibers, shape memory epoxy resin, and hygroscopicity. Hygroscopicity is another essential characteristic of flax fiber/shape memory epoxy hygromorph composites [25]. It refers to the ability of a material to absorb moisture from its environment, which can significantly affect the material's mechanical and physical properties. Moisture absorption can cause a material to expand, resulting in dimensional changes and a loss of strength. However, in some cases, moisture absorption can lead to improvements in specific characteristics, such as hardness and impact resistance.

Dyachkova et al. [26] discussed the development of carbon nanotubes (CNTs) and graphene-based fillers to modify epoxy resin. They used different mass ratios of CNTs and graphene materials and analyzed their effect on particle size and mechanical properties. Optimal dispersions were achieved. The resulting composites remained stable up to 300 °C and exhibited an increase in tensile strength and modulus by 84–88% and 40%, respectively, for certain mass ratios.

Flax fiber/shape memory epoxy hygromorph composites have the potential to be used in self-healing materials. Such materials are capable of repairing damage over time, thereby prolonging their lifespan and reducing the need for maintenance. The shape memory properties of flax fiber/shape memory epoxy hygromorph composites could be utilized to initiate the self-healing process [27].

Nurazzi et al. analyzed research on improving the durability, rigidity, and resilience of hybrid materials, as well as their long-term and short-term effectiveness. Natural fiber–polymer composites are becoming increasingly competitive with synthetic polymer composites in terms of stiffness and cost, and their tensile and impact strength values are approaching those of synthetic materials. Hybrid materials based on natural fiber-reinforced polymer composites are used in various structural and outdoor settings, such as panels installed beneath the floor of vehicles, aircraft components, and constructions intended for use in marine environments [28]. There is significant global interest in the use of natural fibers as substitutes for conventional materials across various industrial sectors, driven mainly by the goal of achieving sustainability and promoting a more environmentally friendly approach. Flax and other natural fibers are combined with polymeric resins to produce novel materials [29].

Machine learning is a powerful of tool that allows algorithms to learn from a dataset and apply that knowledge to new data [30]. Typically, a training phase is required, where a subset of the dataset is used. However, there are potential challenges in applying this method, such as the need for accurate and reliable data input. After the training phase, various algorithms can be utilized, including decision tree (DT) and random forest (RF), which are commonly used in supervised machine learning. These algorithms involve providing both input and corresponding output data to the machine learning model. Decision trees are a machine learning algorithm used for classification and regression analysis. They adopt a tree structure, where internal nodes represent attributes or features of the analyzed data, branches signify decisions or rules, and leaves indicate the resulting outcomes or predictions [31]. Random forest, also known as random choice forest, is an ensemble learning approach employed in diverse problem domains such as classification and regression. This method entails generating numerous decision trees during the training phase [32]. Machine learning has been extensively used in predicting the mechanical properties of composites due to its ability to handle large and complex datasets [33,34].

Moreover, the use of machine learning models has enabled the prediction of these properties with fewer experimental tests, resulting in significant cost and time savings [35].

Li et al. presented a prediction model for the compressive strength of basalt fiber-reinforced concrete (BFRC) using the random forest (RF) algorithm. The model was compared to neural network and support vector regression methods, using metrics such as mean squared error (MSE), root mean squared error (RMSE), and R-squared (R^2). The experiments led to the conclusion that the RF model demonstrated higher accuracy and better regression fit in predicting the compressive strength of BFRC compared to the other models [36].

Lim et al. developed a predictive model for delamination using a combination of the random forest (RF) algorithm and the vibration-based natural frequency shift technique. They established a finite element procedure in the ANSYS Composite PrepPost engine, incorporating delamination, and validated its accuracy by comparing with both numerical and experimentally determined natural frequencies reported in the literature. The validated procedure was then used to determine the free vibration responses of pristine and delaminated composite plates characterized by five delamination parameters. Four RF models were developed and tested to determine the delamination parameters based on natural frequency shifts. The study revealed that the presence of delamination led to a reduction in natural frequencies, particularly noticeable in higher modes. Among the four natural frequency predictors and using a larger dataset, the RF model produced the most accurate predictive results when tested with unseen cases [37].

Liu et al. developed a predictive model using the XGBoost decision tree-based machine learning technique to calculate the residual tensile strength and modulus of pultruded fiber-reinforced polymer (FRP) composites exposed to water, high humidity, and alkaline solutions. The methodology of the XGBoost decision tree was presented, and the predictions were validated using a dataset. The model demonstrated excellent agreement with experimental results, achieving R-squared (R^2) values of 0.93 for predicted tensile strength and 0.85 for modulus. The XGBoost decision tree model provided good interpretability, quantitatively analyzing the importance of input data attributes, including exposure time, exposure temperature, pH value of the environment, fiber volume fraction, plate thickness, fiber type, and matrix type [38].

Gupta et al. introduced a supervised classification model for predicting the high stress abrasive wear behavior of unidirectional epoxy composites reinforced with sisal fiber. The model utilized 192 wear volume measurements across 3 fiber orientations, with 153 used for training and 39 for testing. The study explored two alternatives for the classification model: decision tree induction and sequential covering rule induction. The decision tree-based classification model outperformed sequential covering rule induction in terms of accuracy and stability. Once trained, this efficient model can be applied to sophisticated tribological experiments for predicting, reducing overall complexity and effort compared to strict analytical techniques. Furthermore, it can be applied to various computationally costly models [39].

The mechanical properties of flax fiber/shape memory epoxy hygromorph composites are highly influenced by various experimental conditions, such as fiber orientation, temperature, and humidity. Therefore, studying and predicting the impact of these factors on the mechanical behavior of flax fiber-reinforced composites is essential.

This work focuses on predicting the tensile modulus of flax fiber/shape memory epoxy hygromorph composites under different experimental conditions. The tensile modulus is a crucial mechanical property that measures the stiffness of the material under tensile loading. Accurate prediction of the tensile modulus can greatly assist in designing and optimizing composite structures for various applications.

For the first time, machine learning algorithms were employed to predict the tensile modulus of flax fiber/shape memory epoxy hygromorph composites, considering three initial experimental variables: the orientation of the flax fibers, humidity, and temperature. These variables are known to have a non-linear relationship.

To accomplish this objective, two machine learning algorithms, namely decision tree (DT) and random forest (RF), were utilized. The experimental data collected by Li et al. [24], which pertains to the tensile modulus of the composites under various fiber orientations, temperatures, and humidities, served as the basis for this study.

A comparison of the performance between DT and RF in predicting the tensile modulus of the composites was conducted. The results demonstrate that both algorithms achieve high accuracy in predicting the tensile modulus, with RF slightly outperforming DT in terms of prediction accuracy. Furthermore, a feature importance analysis was conducted using both DT and RF models to identify the most influential factors affecting the tensile modulus. The analysis revealed that fiber orientation has the most significant influence on the tensile modulus, followed by temperature and humidity.

2. Materials and Methods

The large dataset of mechanical properties of flax fiber/shape memory epoxy hygromorph composites used in this work was obtained from the study conducted by Li et al. [24]. The dataset can be accessed at the Mendeley Data repository located at [40]. In their study, flax fiber/shape memory epoxy hygromorph composites were mechanically deformed with consideration of two fiber orientations (0° and 90°) at five temperatures (20 °C, 40 °C, 60 °C, 80 °C, and 100 °C) and two humidity conditions (50% and fully immersed). The details of the tensile tests can be found elsewhere [21,24]. The samples, which were saturated, had dimensions of $t_{0^\circ} = 0.56$ mm and width $w_{0^\circ} = 15$ mm for flax fiber orientations at 0° EL, and $t_{90^\circ} = 0.56$ mm and width $w_{90^\circ} = 25$ mm for flax fiber orientations at 90°. The mechanical properties of the unidirectional laminates were determined using a Shimadzu universal testing machine with a crosshead speed of 1 mm/min, according to ISO 527-4 standards. To prevent moisture loss during testing, samples under relative humidity of 50% and immersed conditions were wrapped with polymer films. In addition, a heating chamber was used to control the temperature prior to testing, and thermocouples were used to measure the temperature near the samples [24]. In this work, the Scikit-learn package was used to implement the algorithms in custom homemade programs. TensorFlow [41] is a widely-used open-source, high-level neural network application programming interface written in Python. It is another powerful open-source framework for creating and deploying machine learning models. It is frequently used for a broad range of tasks, including time series analysis, audio and image recognition, and natural language processing. Deep neural network models can be built, trained, and deployed using a variety of tools and frameworks provided by TensorFlow. It can handle enormous datasets since it supports both CPU and GPU computation, making it scalable and efficient. Data scientists choose TensorFlow because of its versatility and ease of use [42]. Scikit-learn provides a wide range of supervised and unsupervised machine learning algorithms for classification, regression, and clustering. These algorithms can be used to solve a variety of real-world problems, such as image classification, sound signal analysis, and natural language processing. Some of the popular algorithms provided by Scikit-learn include decision trees, random forests, logistic regression, K-nearest neighbors, support vector machines, and neural networks. The simplicity and ease of use of Scikit-learn are two of its greatest benefits. The library's user interface is clear and consistent. Scikit-learn offers a variety of preprocessing algorithms, as well as tools for cross-validation, hyperparameter adjustment, and data separation. These features assist users in data preparation and cleaning, model creation, and performance evaluation. Moreover, Scikit-learn provides a variety of visualization tools, such as heatmaps, histograms, scatter plots, and line graphs, to aid users in understanding their data and model output. These visualizations can be utilized to examine the data, identify trends, and gain insights. Scikit-learn has a large and active user and development community, making it easy to seek assistance and support. The program is well-documented, and the official website offers comprehensive tutorials and examples. Additionally, Scikit-learn has a GitHub repository where users can report bugs and contribute to the package's development. The speed and effectiveness of Scikit-learn are also major benefits [43].

For the implementation of the models, the data are rescaled to a range of [0–1] to improve the training results. The algorithms and equations used in this study are outlined elsewhere [43], with the maximum depth of a tree set to "None," the minimum number of samples in a leaf node set to 1, and the minimum number of samples required in a leaf node also set to 1. A total of 100 values were considered in the machine learning algorithms, with 80% of them used for training (80 values) and the remaining 20% for testing (20 values).

The objective of this work is to employ machine learning algorithms to investigate and predict how experimental variables, such as temperature, humidity, and the orientation (direction) of flax fibers, affect the tensile modulus of flax fiber/shape memory epoxy hygromorph composites.

3. Results and Discussion

To assess the correlation between the features in this study, a correlation matrix map was generated in Figure 1 using Pearson correlation coefficients [44]. Pearson correlation coefficients indicate the strength of the linear relationship between variables [45,46]. A correlation matrix is a table that displays the correlation coefficients between different variables in a dataset, specifically the mechanical characteristics of flax fiber/shape memory epoxy hygromorph composites used in this work. The correlation coefficient between two variables is shown in each cell of the table, with values ranging from −1 (indicating a perfect negative correlation) to 1 (indicating a perfect positive correlation). A correlation coefficient of 0 suggests no association between the variables. In the field of machine learning, correlation matrices are frequently utilized to analyze the relationships between variables and identify those that are most strongly linked to the target variable. By studying the correlation matrix, users can select features and develop models based on the information gained, enabling them to identify the factors that are likely to be accurate predictors of the target variable. Additionally, correlation matrices can be used to identify multicollinearity, which occurs when two or more variables are strongly correlated with each other. In certain situations, removing one or more of the correlated variables may be necessary to prevent overfitting and ensure that the model generalizes effectively to new data [32].

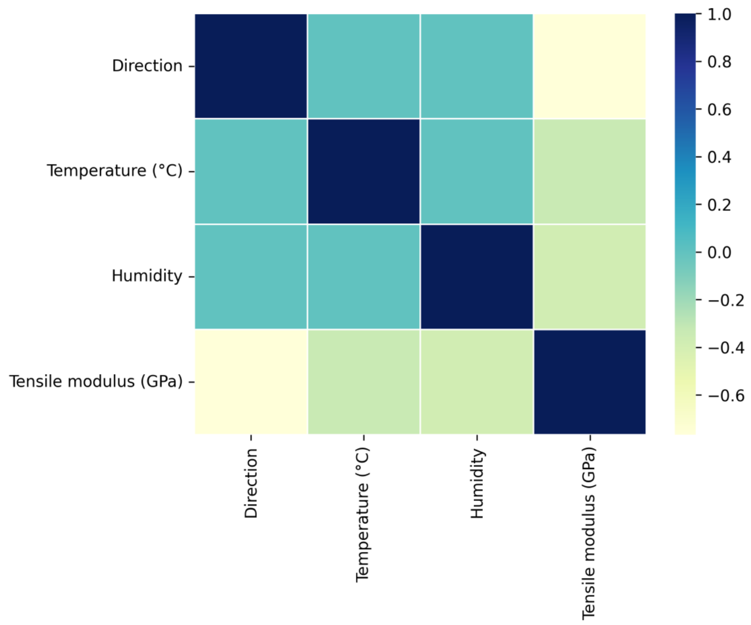


Figure 1. Correlation matrix between variables (Direction, Temperature, and Humidity) and the output (Tensile modulus) of the models.

The related Pearson correlation coefficient between two variables, X and Y, is defined as follows:

$$\rho(X,Y) = \frac{cov(X,Y)}{\sigma_X \sigma_Y} \quad (1)$$

where σ_X and σ_Y represent the standard deviations of X and Y, and $cov(X,Y)$ represents the corresponding covariance [47].

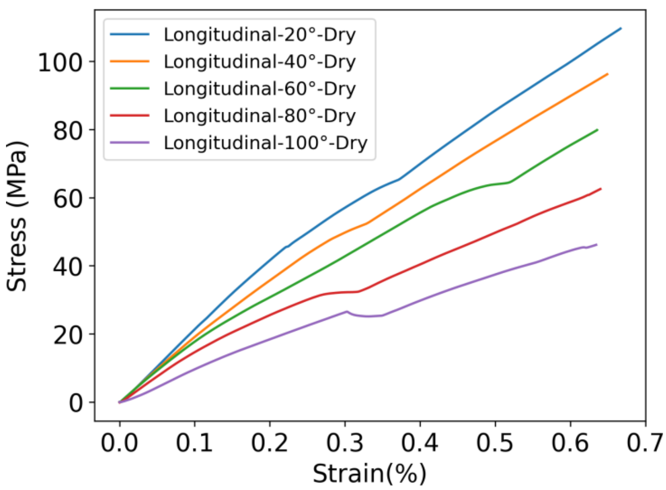
As depicted in Figure 1, the correlation matrix reveals that the relationship between temperature, humidity, and flax fiber orientation (direction) is close to 0, indicating no clear correlation between them. However, there is a negative correlation between temperature, humidity, flax fiber orientation, and the tensile modulus.

Table 1 summarizes the range of input and output values of the models. The range of tensile modulus values obtained in the work of Li et al. [24] was quite broad, with a minimum value of 0.016 GPa and a maximum value of 16.302 GPa. The minimum value was recorded when the composite was fully immersed, the wax fiber was oriented transversely, and the tensile test was conducted at a high temperature of 80 °C. On the other hand, the maximum value was obtained when the composite had a relative humidity of 50%, the wax fiber was oriented longitudinally, and the tensile test was conducted at a lower temperature of 20 °C.

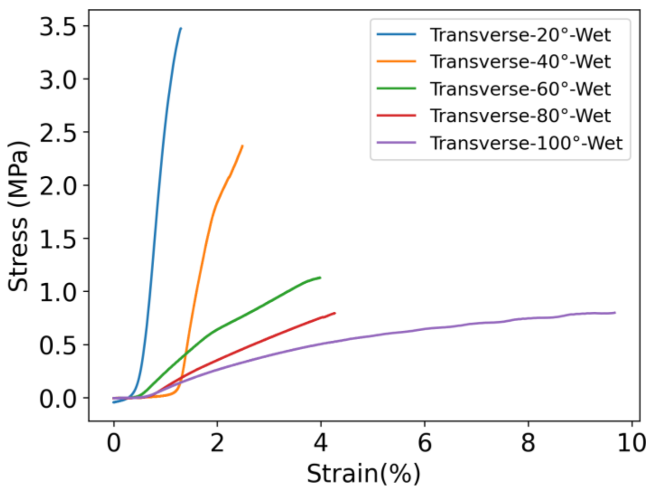
Table 1. Range of input and output values of the models.

Attribute	Range
Direction	0 (Longitude)/90 (Transverse)
Temperature (°C)	20–100
Humidity	50 (Dry)/100 (Immersed)
Tensile Modulus (GPa)	0.016–16.302

Figure 2 depicts the strain–stress curves obtained in the longitudinal direction (Figure 2a) and transverse direction (Figure 2b) at various temperatures and under different humidity conditions (dry and wet, respectively). The complete experimental curves can be obtained from another source [24].



(**a**)



(**b**)

Figure 2. Strain–stress curves obtained: (**a**) in the longitudinal direction; (**b**) in the transverse direction.

Figure 3 depicts the histogram illustrating the frequency distribution of the tensile modulus values. The data reveal that a significant portion, approximately 40%, of the tensile modulus falls within the range of 0 to 2 GPa. However, it is not possible to classify the frequency distribution of the tensile modulus as a normal distribution (Gaussian distribution), uniform distribution, or lognormal distribution. Nevertheless, the data were fitted to an exponential function with an adjustment constant, and the parameters were fined-tuned to achieve the optimal exponential curve that best aligns with the experimental data. An R-square (R^2) of 0.94 was obtained. R^2 serves as a metric to assess how well a model explains the variation in a dependent variable. Its range is from 0 to 1, with 0 indicating that the model explains none of the variance and 1 indicating a full explanation. In this case, the exponential fit corresponds well to the distribution of the tensile modulus.

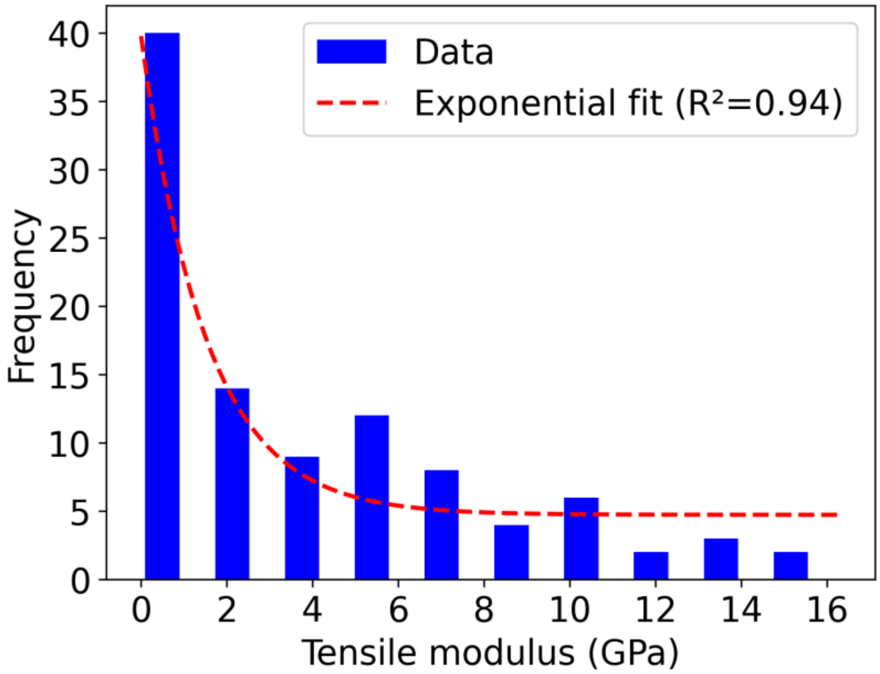
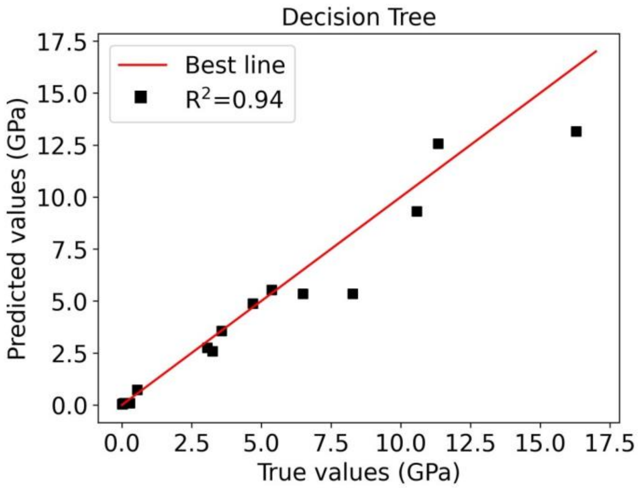
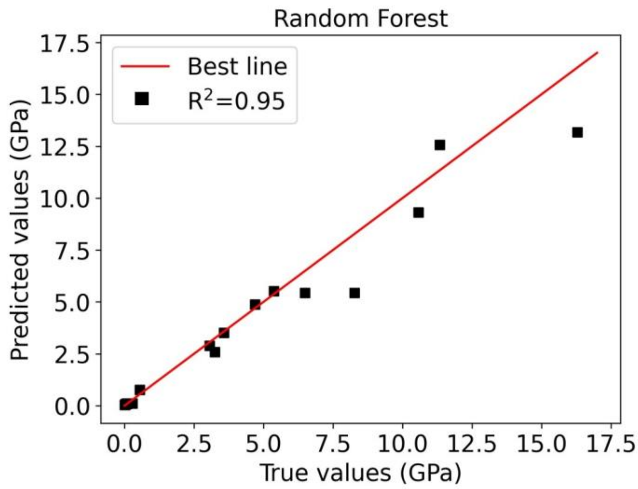


Figure 3. Histogram distribution of the frequency of the tensile modulus.

These results emphasize the significant influence of wax fiber orientation and environmental conditions on the mechanical properties of hygromorph composites. Such insights can help optimizing the design and fabrication of these composites for specific applications. The utilization of ML models can further enhance our understanding of the relationship between various parameters and the resulting mechanical properties of hygromorph composites. Figure 4 depicts the comparison between the true and predicted tensile modulus of the test data, considering the decision tree model (Figure 4a) and the random forest model (Figure 4b). The plots demonstrate a strong agreement between the predicted and actual values of the tensile modulus for flax fiber/shape memory epoxy hygromorph composites, with the majority of the data points lying close to the 45-degree line (the best line), indicating nearly perfect predictions. To compare the performance of the two machine learning models, a statistical measure called the coefficient of determination, R^2, was employed. R^2 is commonly used to evaluate the effectiveness of machine learning models [48]. According to Chicco et al., R^2 is often the most informative statistic in many cases compared to other measures such as mean absolute error (MAE) and its percentage variation (MAPE), symmetric mean absolute percentage error (SMAPE), mean square error (MSE), and the square root of mean square error (RMSE) [33]. MAE calculates the average absolute difference between the actual and expected values of a dataset. Therefore, R^2 is recommended as the standard measure for evaluating regression analyses across various scientific disciplines [48].



(a)



(b)

Figure 4. True and predicted values of the tensile modulus by: (**a**) the decision tree model; (**b**) the random forest model.

An R^2 value of 0.94 was obtained for the decision tree model, while the random forest model achieved a value of 0.95. These high R^2 values indicate that both models effectively capture a substantial portion of the data's variation and can be considered strong predictors of the tensile modulus outcome.

Composite materials, such as flax fiber/shape memory epoxy hygromorphs, exhibit significant variations in their mechanical properties. This inherent variability can introduce noise and make it challenging to further increase the R^2.

Table 2 provides a classification of the feature importance in the models, indicating the extent to which each input feature contributes to the overall predictive power of the models. In both cases, the flax fiber orientation (direction) emerges as the parameter with the most significant influence on the tensile modulus. It is followed by the humidity and temperature levels that were considered during the tensile tests.

Table 2. Features importance of the models.

Feature	Decision Tree	Random Forest
Direction	0.598	0.605
Humidity	0.224	0.215
Temperature (°C)	0.178	0.180

This study highlights the potential of machine learning algorithms in accurately predicting the mechanical behavior of flax fiber-reinforced composites under various experimental conditions. The predicted tensile modulus values offer valuable insights for designing and optimizing composite structures for diverse applications. Future research can expand upon this approach by investigating other mechanical properties, including compression strength, impact resistance, and fatigue performance, and examining the influence of additional experimental conditions, such as moisture content and loading rate on the mechanical behavior of composites. Furthermore, there is potential to explore the optimization of fiber orientation to attain desired mechanical properties, while considering factors such as processing conditions and manufacturing constraints. In addition, exploring alternative machine learning algorithms could be beneficial to determine whether they yield more accurate predictions or provide deeper insights into the behavior of biocomposites. It would also be worthwhile to explore the application of these techniques to other materials, such as Ni–W with a composite-like microstructure [49–51], dissimilar welding joints [52], or harmonic alloys [53,54].

4. Conclusions

This work successfully demonstrates the effectiveness of machine learning (ML) models in studying the impact of various factors on the tensile modulus of flax fiber/shape memory epoxy hygromorph composites. The study specifically investigates the influence of fiber orientation, humidity, and temperature on the mechanical properties of these composites. The results reveal that ML models provide accurate predictions of the tensile modulus, as indicated by high R-squared values of 0.94 and 0.95 for the decision tree (DT) and random forest (RF) algorithms, respectively. Among the variables studied, the orientation of the flax fibers emerges as the most influential factor affecting the tensile modulus, followed by humidity and temperature. These findings underscore the potential of ML models in predicting and optimizing the properties of hygromorph composites for specific applications. The practical implications of this research are significant for the development of advanced materials with tailored properties. The findings suggest that optimizing the orientation of the flax fibers, as well as controlling humidity and temperature, can substantially enhance the tensile modulus of these composites.

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