**Predictive Modeling of Tensile Strength and Mass Loss Temperature in Wool-Reinforced PLA Composites Using Machine Learning**

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**Abstract**

Polylactic acid (PLA)-based fiber-reinforced polymers (FRPs) often demand detailed characterization of their thermal and mechanical properties to indicate their potential field of application. Conventional characterization methods are usually costly and time-consuming, what creates opportunity for alternative strategy that involves employing machine learning for property prediction. In this study, prediction models were formulated for the tensile strength and 5% mass loss temperature of unmodified and modified wool-reinforced PLA composites. The modeling employed linear and boosted decision tree regression algorithms within the Microsoft Azure Machine Learning Studio. The resulting models facilitated the prediction of numeric values for the targeted properties, in functions of concentrations of wool, coupling agents, and coupling agent types. Model evaluations were conducted to interpret and compare their accuracy. Furthermore, the predicted values for selected formulations were put together with actual average results obtained through laboratory experimentation. This approach aimed to assess the effectiveness of machine learning in predicting selected thermal and mechanical properties of PLA-based FRPs, contributing valuable insights to the field of material science and potential advancements in the domain.

**Keywords:** fiber-reinforced polymers; polylactic acid; maleinized linseed oil; sheep wool fiber; silane coupling agent; machine learning; regression model

# INTRODUCTION

Fiber-reinforced polymers (FRPs) represent modern materials designed to achieve designed properties.[1] The incorporation of such reinforcement is frequently employed for modifying or enhancing plastics to adhere to applicable standards in a significant portion of plastic applications. Consequently, the escalating interest in research pertaining to plastic reinforcement is well-founded, given its impact on the manufacturing of plastic components and the subsequent possibilities for their advancement. [2–4]

The surge in environmental awareness and a commitment to sustainable production processes have fostered a sustained interest in the development of green composites. Unlike materials dependent on fossil fuels, where additional greenhouse gases are emitted into the atmosphere, the life cycle of green composites involves the production and disposal of carbon dioxide and greenhouse gases. Despite the fact that certain biobased materials, such as wool fiber, may necessitate additional production steps, they offer significant advantages over their fossil-dependent or inorganic counterparts. [5,6]

An example of sustainable fiber-reinforced polymers is polylactic acid reinforced with silane-modified sheep wool fibers with an addition of malenized linseed oil. Various formulations of such FRPs have been examined by considering the mechanical, thermal, or microstructural characteristics of the obtained materials. The characteristics allowed us to compare the formulations according to procedures based on testing standards, such as tensile (ISO 527) and other standards.[7–10] Such materials could be successfully processed with broadly used and economically efficient injection-molding processing methods, where detailed processing conditions were described in previous works. [4,11] Although in the previous work, concentrations of polylactic acid and malenized linseed oil were constant, various concentrations of sheep wool fibers and various conditions of silane or alkoxide coupling agents were taken under investigation. Furthermore, several concentrations and different chemical structures were tested to treat the coupling agents' treatment conditions; a detailed characterization of materials allowed for a database of materials' properties. Variable concentrations of sheep wool fiber or coupling agents might be used for creating a regression model for the prediction of formulation properties.

Nowadays, artificial intelligence (AI) is more and more often used for diverse areas of expertise such as technology, finance, industry, commerce, entertainment and many other.[12,13] Research in materials science traditionally requires designed experiments based on trial and error approach with strong chemical intuition. Such approach is often time and effort consuming as it require to verify numerous material combination in laboratory leading to high material and energy consumption to form valuable conclusions The usage of mathematical models helps to analyze specific aspects of an investigation. [13,14] As mathematical approaches such as density functional theory, molecular dynamics or finite elements often provide faster and cheaper alternative to laboratory research, they are still constrained with time what drives it towards limited fields of applications. Alternatively to conventional scientific and computational methods may be applied AI techniques such as deep learning, as they offer faster results as traditional mathematical approaches providing simultaneously comparable to physics-based level of accuracy. The role of artificial intelligence is to provide a computing solution for information processing based on cognitive services to interpret text, hearing, speaking, or vision. Therefore machine learning can be successfully applied to process the information and achieve conclusions or predictions based on gathered data. On the one hand, there is a wide variety of applications where AI computer vision models are applied for manufacturing processes, where vision systems recognize target objects and assist human operations, or where based on vision recognition enhances planning systems.[13,15,16] On the other side, a regression model can be created and trained on the specific numeric value.

Regression models created through mathematical modeling are more often in use due to the increasing amount of data gathered during material characterization and relatively higher costs and time-consuming, broad characterization of material in the laboratory. As a result of the regression model, a quantitative relation between physicomechanical properties with proportions in model material can be established. The similarity of materials in the model and the stability of the obtained results allow applying the data for regression models. [17] Several algorithms can be distinguished for training regression models, among others, linear and boosted decision tree algorithms. [18] The linear regression algorithm establishes a linear function between independent variables providing a numeric value as an outcome. A machine learning instance enhances fitting the line and measuring the error between the function and actual measurement data. Such algorithms are most common and usually used for simple models with low-complexity data sets. The linear regression models have mainly used two different methods. A gradient descent method minimizes error with each step of training a model. Among parameters to control such optimization can be distinguished step size or learning rate. An ordinary least squares method refers to the loss function, where error corresponds to the sum of the square distance between the actual and predicted value. Fitting the model is achieved by minimalizing squared errors. Such a model is best fitting when a linear correlation is assumed. Furthermore, the boosted decision tree algorithm allows the creation of decision tree models corresponding to prior present decision trees. The boosting algorithm learns by fitting a residual of the prior decision tree to improve the model's accuracy. In the regression models, a series of step-wised boosted decision trees is created, and based on arbitrary factors optimal tree is being selected. [13]

The main objective of current research is regression modeling of properties of the polylactic acid-based sheep wool fiber reinforced polymers based on the thermal and mechanical results from previous investigations. [4,11]

# EXPERIMENTAL

In the prior studies, polylactic acid (PLA) was modified with maleinized linseed oil (MLO) and reinforced with unmodified or modified sheep wool fibers with four coupling agents. Labeling of coupling agents remained as follows: (3-(2-aminoethylamino)propyl)-trimethoxysilane (labeled as coupling agent A), trimethoxy (2-(7-oxabicyclo(4.1.0)hept-3-yl)ethyl)silane (labeled as coupling agent B), tris(2-methoxyethoxy)(vinyl) silane (labeled as coupling agent C) and titanium (IV) (triethanolaminate)isopropoxide (labeled as coupling agent D).The experimental phase of this study involved collecting raw data from prior studies, specifically experiments conducted in accordance with industrial testing standards.[4,7,11]

The outcomes derived from the tensile test, specifically the maximum resistance, and the thermogravimetric analysis, specifically the 5% mass loss temperature, were gathered and wrote down in the dataset in form of CSV file. The gathered data was based on various variables for characterization, including the presence of maleinized linseed oil (MLO) in the formulation, the concentration of sheep wool fiber or its surface modification type, and the concentrations of a coupling agent used for modification. A comprehensive summary of the formulations involved within the database is provided in **Table 1**, offering a structured representation of the relevant experimental variables and their corresponding labeling.

**Table 1. The formulations collected in the database**

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Label | PLA [phr] | MLO [phr] | Wool [phr] | Coupling agent type | Coupling agent concentration [phr] |
| PLA-MLO-1W | 100 | 10 | 1 | n.a.\* | 0 |
| PLA-MLO-5W | 100 | 10 | 5 | n.a.\* | 0 |
| PLA-MLO-10W | 100 | 10 | 10 | n.a.\* | 0 |
| PLA-MLO-1W-1C | 100 | 10 | 1 | C | 1 |
| PLA-MLO-5W-1C | 100 | 10 | 5 | C | 1 |
| PLA-MLO-10W-1C | 100 | 10 | 10 | C | 1 |
| PLA-MLO-1W-2.5C | 100 | 10 | 1 | C | 2.5 |
| PLA-MLO-5W-2.5C | 100 | 10 | 5 | C | 2.5 |
| PLA-MLO-10W-2.5C | 100 | 10 | 10 | C | 2.5 |
| PLA-MLO-1W-1A | 100 | 10 | 1 | A | 1 |
| PLA-MLO-1W-1B | 100 | 10 | 1 | B | 1 |
| PLA-MLO-1W-1D | 100 | 10 | 1 | D | 1 |

n.a.\* - not applicable

In the subsequent phase of the study, preprocessing of the prepared data was conducted to adjust its suitability for regression Machine Learning (ML) algorithms. Initially, a targeted selection of columns was performed, focusing on material composition and the experimentally obtained characterization values. All data were converted into numerical form to comply with a supervised regression model type. To achieve this, the selected columns underwent analysis to identify and subsequently eliminate entries that were either empty or non-numerical. Following the process, the normalization of experimental results was applied in order to standardize the data within a range of 0 to 1, regardless of the actual numerical values derived from experiments. The rational values between 0 and 1 maintained the original scale between the experimental results, preserving the relative relationships among the data points.

Conclusively, the prepared data underwent a final step of random partition into two subsets: 75% of the data, comprising 115–116 records, was allocated for model training, while the remaining 25%, consisting of 37–38 records, was reserved for model evaluation. This thorough preprocessing and division of the dataset into training and evaluation subsets aimed to ensure effectiveness of the subsequent machine learning models developed for selected material properties and their modeling approaches. An example of as created pipeline has been shown in Figure 1.

Diagram

Description automatically generated

Figure 1 Training pipeline in Microsoft Azure Machine Learning Studio

For model training were used two types of models: linear least square regression with a regularization rate equal to 0.001 and with an intercept term; and boosted decision tree with a single parameter training mode, maximum of 20 leaves per tree, minimum of 10 samples per leaf node, a 0.2 learning rate and 100 trees constructed per model. The evaluation, as an additional outcome of the machine learning pipeline, included mean absolute error (MAE), root mean squared error (RMSE), relative absolute error (RAE), relative squared error (RSE), and coefficient of determination (R2). [19]

Subsequently, prepared training pipelines were deployed into batch inference pipelines, as presented in Figure 2. In this scenario, a structured CSV data file, organized in a tabular form, was created with a similar structure to the original database used for training. The database used for predictions of degradation temperature (T5) and tensile strength contained variable wool concentration (from 1 to 15phr, with 1phr step), coupling agent concentration (from 0 to 5phr, with 0.5phr step), and coupling agent type (the A, B, C, and D). The prepared prediction dataset underwent a transformation using the previously trained models, and the scoring process followed. The outcome of this scoring procedure involved obtaining predictions for material characteristics based on the updated input parameters, contributing to an enhanced understanding of the influence of wool and coupling agents concentrations on the estimated material properties.

Diagram

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Figure 2 Batch inference pipeline

# RESULTS AND DISCUSSION

The methodology detailed earlier helped to create models capable of simulating numerical values for above mentioned mechanical and thermal properties. The resultant score results were visually represented through graphical presentations for all tested coupling agents. Each coupling agent's evaluation was captured in a set of four plots, corresponding to different properties. These plots, merged with their respective model evaluations, were collectively presented in figures ranging from Figure 3 and Figure 6. Additionally, Figure 7 displayed the average measurement results with their standard deviation for selected formulations. [4,11] This figure also incorporated the corresponding scored results derived from the created models, providing a complete visualization of the relative performance of the models against experimental measurements.

The data export from the inference pipeline summarized simulated tensile strength properties across varied concentrations of wool and coupling agents, including different types of coupling agents. The visual representation of tensile strength predictions is illustrated in both Figure 3 and Figure 4, wherein differences between tested algorithms are defined, with separate plots for each coupling agent.

It is notable that each model type represents a distinct approach; for instance, the boosted decision tree groups data into a more condensed subset, while linear regression exhibits a linear change with changes in parameters. Furthermore, regarding the boosted decision tree model, wherein predictions for tensile strength can exhibit sharp differences between adjacent formulations. In contrast, linear regression consistently shows similar tensile strength predictions for adjacent formulations. This difference highlights the behavior of different models in capturing and representing the complex relationships within the data.

Chart, surface chart

Description automatically generated

Figure 3 Prediction of tensile strength from linear regression model

The linear regression model, shown in Figure 3 , reveals a general trend wherein an increase in the concentration of coupling agents, coupled with a reduction in wool concentration, corresponds to an increase in tensile strength. Nevertheless, this improvement was particularly notable at lower coupling agent concentrations, specifically up to 0.5 phr. Notably, coupling agents B and D, characterized by lower concentrations, demonstrated more significant improvements compared to other surface modifiers, thereby contributing to the overall enhancement of tensile strength.

Chart, treemap chart

Description automatically generated

Figure 4 Prediction of tensile strength from boosted decision tree regression model

An alternative approach for simulating tensile strength involved the use of boosted decision tree regression. Across all examined coupling agents and wool concentrations below 1 phr, the addition of coupling agents resulted in an enhancement of tensile strength. Notably, changes in the concentration of coupling agents did not lead to changes in this property within the boosted decision tree model. In cases of higher wool concentrations, where an increase in wool content tended to decrease tensile strength, the presence of even low concentrations of coupling agents proved beneficial in modifying the adverse impact of wool addition. For wool content exceeding 7 phr, it was concluded to use coupling agent concentrations above 2 phr. Bring into line with the assumptions drawn from the linear regression model for tensile strength, both coupling agents B and D showed the most effective compensation for the negative effects of wool additive compared to the remaining coupling agents investigated in the study.

The evaluation of the tensile strength models valued their precision and facilitated comparisons between them. Mean Absolute Error (MAE) and Root Mean Squared Error (RMSE) evaluations were provided in MPa units, enabling the comparison of models pertaining to the same property. On the other hand, Relative Standard Error (RSE), Relative Absolute Error (RAE), and Coefficient of Determination (R2) are absolute values, helping additional comparisons among models across various properties with diverse value ranges and units. The boosted decision tree regression model displayed lower values for both MAE and RMSE, indicative of superior precision compared to the linear regression model. The MAE value of 1.34 MPa for the boosted decision tree model indicated better average model accuracy, whereas the linear regression model exhibited a value of 2.16 MPa. Although the boosted decision tree model showed higher accuracy, the errors of both MAEs fell within the range of 0.93-2.59 MPa, corresponding to the standard deviation of the original data employed for modeling.[4,11] Additionally, the marginal difference between the MAE and RMSE values for the boosted decision tree regression suggested a similarity in prediction errors. On the contrary, for linear regression, the substantial difference between the MAE and RMSE indicated the presence of larger errors for the predictions.

Furthermore, the R2 values demonstrated that the boosted decision tree regression revealed a good fit to the input data, suggesting its applicability for predicting data points beyond those used in model creation. However, despite the good fitting of linear regression based on the coefficient of determination, greater errors in tensile strength prediction were observed.

The models for the 5% mass loss temperature showed a comparable trend, although the boosted decision tree model tended to group results into subsets more notably than the linear regression model. The results of the assumption for the linear regression model are showed in Figure 5. The model predicts that, following an initial increase, the temperature decrease occurs at coupling agent concentrations higher than 0.5 phr. The model also predicts that the initial increase in mass loss temperature observed for 0.5 phr is reduced by an increase in coupling agent concentration. Depending on the type of coupling agent, the model predicts equal temperatures between 1-2 phr of coupling agent. The highest initial increase in temperature was observed for coupling agent B modification, while 0.5 phr of coupling agent C exhibited the lowest temperature among the modifications.

Graphical user interface, chart

Description automatically generated

Figure 5 Prediction of 5% mass loss temperature from linear regression model

The boosted decision model exhibits comparable patterns in response to varying coupling agent concentrations, as illustrated in Figure 6. Specifically, the model predicts that treatment with up to 0.5 phr of coupling agent leads to an increase in temperature. Subsequently, between 0.5 and 1.5 phr, the temperature stabilizes at a stable level, and beyond this range, it experiences a decline. This behavior can be categorized into three distinct subsets of predictions based on wool concentration: below 0.5 phr, between 0.5 and 7 phr, and above 7 phr.

Furthermore, among the tested coupling agents, coupling agent C is generally associated with the most substantial decrease in the predicted temperature. This observation shows effects of different coupling agents on the predicted thermal behavior, further contributing to the understanding of their impact in varying concentration scenarios.

Chart, treemap chart

Description automatically generated

Figure 6 Prediction of 5% mass loss temperature from boosted decision tree regression model

Analogous to the mechanical models for the 5% mass loss temperature, greater accuracy was achieved for the boosted decision tree regression model. The Mean Absolute Error (MAE) value for this model was noted at 2.86ºC, while the MAE increased to 3.59ºC for the linear regression model. This difference in MAE values highlights the better precision of the boosted decision tree regression model in predicting the 5% mass loss temperature when compared to its linear regression counterpart.

To compare actual data with modeled data, we selected the results of five representative formulations from **Table 1** involving both unmodified and modified types of wool fiber. As predicted from the model evaluations, both the boosted decision and linear regression models enabled the prediction of results that closely resembled the actual average outcomes. However, for certain samples such as PLA-MLO-1W or PLA-MLO-1W-1C, predictions generated by both linear and boosted decision tree regressions did not yield satisfactory results when compared to the actual measured values and exceeded mean absolute errors.

Despite the proximity of numerous predictions to the actual values, it is essential to acknowledge that the presented models are not perfectly fitting and should be regarded rather as reference points. Furthermore, when predicting values, both models should be considered to showcase the range of predictions and provide a comprehensive perspective on the potential variations between the modeled and actual outcomes.

Chart, bar chart

Description automatically generated

Figure 7 A summary of results from laboratory measurements and modeled values.

In conclusion, considering that the model was developed based on two distinct research studies, it is necessary to define the advantages and drawbacks associated with the models. Firstly, both studies utilized identical batches of raw materials, comprising polylactic acid (PLA), maleinized linseed oil (MLO), wool fibers, and coupling agents. These materials were processed using the same machinery. Secondly, both tensile strength and 5% mass loss temperature assessments were conducted employing identical equipment, under the same testing conditions, and by the same technician. It is important, however, that the process of coupling agent modification varied in terms of mixture ingredients and the temperature of modification. Additionally, the investigative phases of both works were separated by a 2-month interval.

Despite the mentioned disadvantages, it is pertinent to highlight that the similarities in raw materials, processing conditions, and testing procedures allow for a relatively accurate comparison between the two studies, minimizing significant errors in the assessment of the developed models.

# CONCLUSIONS

In summary, the presentation of model results facilitates a clear understanding and comparison of different algorithms employed for predictions. Within the boosted decision tree model, changes in the concentration of coupling agents do not induce changes in the property; however, at higher concentrations of wool, even low concentrations of coupling agents assist in partially modifying the undesired reduction in mechanical properties due to wool addition. The evaluation of tensile strength models provided insights into the precision of each model, enabling effective comparisons.

Notably, the boosted decision tree regression model showed lower values for both Mean Absolute Error (MAE) and Root Mean Squared Error (RMSE), signifying superior precision compared to the linear regression model. The coefficient of determination demonstrated that the boosted decision tree regression model exhibits a good fit to the input data, making it potentially applicable for predicting data points beyond those utilized in model creation. On the other hand, the good fit of the linear regression model based on the coefficient of determination is associated with greater errors in predicting tensile strength. This underscores differences between model accuracy and predictive capabilities, highlighting the need for careful consideration in choosing the appropriate model for specific applications.

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