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PROCESS VARIABLES IN MIXTURE EXPERIMENTAL DESIGN APPLIED TO WOOD PLASTIC COMPOSITES

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Abstract:

The inclusion of process variables in mixture experimental design is crucial for optimizing final products with precision. Unlike standard response surface designs, which are limited by the requirement that proportions must sum to 100%, mixture-process experiments enable a thorough evaluation of how operational factors, such as particle size and mixing time, interact with mixture components. This approach enhances the understanding of how processing conditions affect product properties and leads to more accurate predictive models, thereby improving production consistency and reliability. Regression analysis reveals that interactions between PET and both particle size and mixing time significantly impact the response variable. The model demonstrates strong predictive accuracy, with R-squared and adjusted R-squared values of 92% and 86%, respectively, and a low root mean square error (S) of 0.2818. The PRESS value of 3.38 confirms the model's ability to accurately predict new data. The absence of high multicollinearity, as indicated by variance inflation factor (VIF) values below 5, further supports the model's stability and interpretability. Contour plots illustrate the effect of varying mixture proportions on the response, such as tensile strength, showing a positive impact of both particle size and mixing time. The highest tensile strength is achieved at maximum levels of these variables, indicating a synergistic effect. Response variable optimization identifies the optimal mixture composition as 90% PET, 10% wood, and no coupling agent. To maximize tensile strength, the largest particle size and longest mixing time should be used, though extrapolating beyond the studied parameters should be done with caution.

Keywords: wood plastic composite; PET; polyethylene terephthalate; design of experiments for mixtures; process variables.

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1. Introduction

Wood plastic composites, known as WPC are materials that have a polymeric component, with an additional filler component embedded within it. These days it is quite common to mix materials with natural fibers (Najafi, 2013); these filaments are generally wood.

Wood fiber fillers were originally employed to reduce the material's density and, in some cases, the cost of products. However, it has been demonstrated that adding coupling agents can significantly improve certain mechanical properties of WPC (Adhikary et al., 2008). This happens due to the coupling agent improving the interaction between the polymer base and the wooden fibers. Some mechanical properties in WPC, such as tensile strength, compression strength, and flexural strength, have been improved in thermoplastics like PET (polyethylene terephthalate) (Cruz-Salgado et al., 2015; Cruz-Salgado et al., 2023). The reinforcement of specific thermoplastics with natural filaments can result in properties that are like those of fiberglass. (Herrmann et al., 1998). Additionally, it can be said that some thermoplastic polymers are environmentally beneficial, because the polymer matrix and the natural filler are generally recycled materials (Cheung et al., 2009)

Additionally, PET is widely known as the prevalent type of semi crystalline and see through thermoplastic in the polyester group because of its strong rigidity and resistance, to both mechanical stress and chemicals properties (Ashwani et al., 2021). Due to the wide applicability of PET, its demand has been increasing over the years. In 2016, a demand of 8400 kilotons was reported, with an expected annual growth rate of 6.9% (Velásquez et al., 2019). Due to the growing need for it and increasing demand these days brings about a surge in the buildup of waste poses a significant environmental challenge to address. Conversely, it is widely accepted that engaging in recycling efforts and advocating for an economy stands out as the most effective approach to mitigate the environmental repercussions and pave the way towards sustainable progress and growth. (Dahlbo et al., 2018). This increase in PET waste has

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generated significant interest in post-consumer recycling, representing an area of opportunity, both economically and environmentally (Velásquez et al., 2019).

Similarly, the primary difficulty in producing WPC lies in optimizing its formulation, specifically in determining the appropriate proportions of thermoplastic polymer, wood filler, and coupling agent that should constitute the mixture. (Cruz-Salgado et al., 2015; Cruz-Salgado et al., 2023). These are generally the components that make up the composite. To optimize the mechanical properties of WPC, it is essential to adjust the proportions of the components and thus assess how these variations influence the properties of the composite. Since the composite represents the mixture of the three components that form a WPC, the components cannot be varied independently. For example, reducing the proportion of PET would require reducing one or more of the remaining components, this to have mixtures of the same size for a valid comparison (Cruz-Salgado, 2015; Cruz-Salgado et al., 2016; Cruz-Salgado, 2016).

On the other hand, design of experiments (DOE) is an experimental strategy that efficiently helps to find the best arrangement of factors that affect one or more response variables. Within DOE, there is a topic called experimental designs for mixtures. In this type of experimental design, the factors under study are the proportions of the components in a mixture. The hypothesis is that different proportions of the mixture affect a response variable of interest. Applied to the case of WPC, the hypothesis is that different proportions of the polymer matrix, the coupling agent, and the wood filler, significantly modify certain mechanical properties of the composite (Cruz-Salgado et al., 2023).

These proportions are connected through a linear constraint of the type:

$$x_1 + x_2 + \cdots + x_q = 1 \qquad (1)$$

where represent the proportion of the component of the mixture (Cornell, 2002).

In some cases, the components proportions in the mixture may be subject to additional constraints of the type:

$$a_i \leq x_i \leq b_i \qquad (2)$$

for one or more components (Cornell, 2002). In the case of WPC, these additional constraints represent, for example, that the proportion of wood filler must be less than or equal to 20% of the total mixture, or that the proportion of the coupling agent must not exceed 5% of the total mixture.

Experimental designs for mixtures are generally modeled using Scheffé polynomial models (Scheffé, 1958), which are fitted to the experimental data using the method of least squares. Some alternative forms of models include the so-called intercept model, which is obtained by replacing one component of the mixture with a constant term (Cruz-Salgado, 2016). The justification for using the intercept model is that it offers a lower degree of collinearity in the terms of the fitted model, which represents less numerical instability in the estimation of the model coefficients (Piepel et al., 2021). Three articles discuss several advantages of this approach, as well as recommendations for its use and interpretation (Cruz-Salgado et al., 2015; Cruz-Salgado, 2016; Kang et al., 2016). It is important to highlight that experiments for mixtures whose components have additional constraints of type (2) can result in an extremely small range in terms of the mixture. Not only is the region of the experimental design restricted, but the model used in the mixture design must also satisfy restriction (2). This can lead to problems in model fitting, stemming from poor conditioning of the information matrix required for least squares fitting (Kang et al., 2016). In other words, the columns of the corresponding model matrix can be nearly linearly dependent. Some consequences of poor conditioning include: estimated parameters by least squares have large standard errors and tend to be correlated, and the estimates tend to be highly dependent on the precise location of the experimental design points (Prescott et al., 2002).

Process variables are factors in an experiment that are not part of the mixture itself but whose variations can influence the blending properties of the ingredients. In addition to analyzing the effect of varying the proportions of a mixture, one might be interested in determining the effect when changing the level of one or more operational factors (Cornell, 2002). Some examples of operational factors are different particle sizes, different mixing times, or different types of coupling agents. Standard response surface designs, such as factorial designs or central composite designs, are not ideally suited for mixture problems because these designs typically assume that individual factors can be adjusted independently of the levels of other factors (Cornell, 2002). In mixture experiments, altering the proportion of one ingredient directly affects the proportions of the other ingredients, as the total proportions must sum to 100%. Consequently, mixture–process experiments are needed. In these experiments, the investigator also examines additional variables, such as mixing speed or preparation temperature, which can be varied independently of each other and of the mixture components. (Anderson-Cook et al., 2004).

The inclusion of process variables refers to incorporating factors that influence the processing or production conditions along with the mixture components themselves. Process variables can interact with the mixture components, affecting the final product's properties. For instance, in a composite material, factors like temperature, pressure, or curing time might interact with the proportions of PET, wood powder, and coupling agents. Including process variables allows for a more comprehensive optimization of the mixture design. By accounting for these variables, can better understand how different conditions affect the performance and quality of the final product. When process variables are included, the experimental design can lead to more accurate predictive models. This helps in predicting the outcomes under various conditions and improving the consistency and reliability of the process production (Cornell, 2002).

The novelty of this study lies in its incorporation of process variables into the experimental design for optimizing wood plastic composites (WPC). While traditional mixture designs focus solely on the proportions of components such as the polymer matrix, wood filler, and coupling agent, this study introduces operational factors—such as particle size, mixing time, and temperature—that can be varied independently of the mixture composition. This approach allows for a more comprehensive analysis of how these process variables interact with the mixture components to influence the mechanical properties, particularly tensile strength. The unique contribution of this research is its integration of mixture and process variables in the experimental design, providing a more precise understanding of the complex relationships between material composition and production conditions. By accounting for these interactions, the study offers an innovative framework for improving the consistency, performance, and overall quality of WPCs, which is relatively underexplored in current literature. This combined optimization model enhances the predictive accuracy of the mechanical behavior of WPCs under varying conditions, making it a valuable advancement in the field of composite material development.

In this article, we discuss mixture designs that include process variables in the analysis applied to wood plastic composites optimization. Relatively little research has been conducted on incorporating process variables into experimental designs that involve mixture variables. The goal is to determine both the proportion of the mixture components and the levels of the process variables that maximize the tensile strength property of a WPC.

2. Materials and methods

2.1. Materials

Test specimens of the composite material were molded using virgin low viscosity (0.75) PET as the polymer matrix and wood powder as the natural filler. The PET was obtained from INVISTA™ POLYPROPYLENE (Mexico City). Produced from the industry processing with surface planners, wooden shavings of Pinus elliottii were purchased from sawmills located at a local lumberyard at León Guanajuato México. The additive that acts as the coupling agent (maleic anhydride) from Du Pont Company was purchased by Taotao Plastic Raw Materials Co., Ltd. (China).

2.2. Mixture preparation

The sawdust was sieve utilizing an AS 200 Analytical Sieve Shaker and a test sieve for particle size analysis. The objective was to dimension the filler particle size to mitigate the potential effect of different wood sizes on the mechanical properties of the WPC. The particle size used in the experiment was 1.4 mm and 2.4 mm.

Three materials, PET, wood powder, and the coupling agent, were dehydrated in a drying oven at a temperature of 100 °C for 8 hours. This procedure was carried out for the total amount of material needed for the entire experiment. The range of mixing times used was 5 and 10 minutes.

After dehydration, the materials were weighed to prepare the different formulations for each treatment of the experimental design. A Brabender single-screw laboratory extruder (PL2200 PLASTICORDER, Mexico) was used to mix the materials. The Brabender Single-Screw Laboratory Extruder is a product manufactured by Brabender GmbH & Co. KG, a German company specializing in instruments and equipment for material testing and quality control, particularly in plastics, food, and chemical industries. The screw speed was set to ninety RPM. The temperature profile across the various zones of the extruder was 240, 250, 260, and 260 °C, respectively.

The room temperature during mechanical testing was set according to ASTM D638 and ISO 527, which are commonly used for tensile testing of plastics and composites, the room temperature was maintained at 23°C (±2 °C), with a relative humidity of around 50% (±10%).

All formulations were prepared using an extruder, which is supplied through a hopper. The components enter the extruder via this hopper and are transported through four heating zones by means of a screw mechanism. As the polymer comes into contact with the walls of the extruder cylinder, it begins to melt, encapsulating the wood. The coupling agent, which has a lower melting point than the polymer, melts more quickly, allowing it to adhere to the polymer and wood before the wood is fully encapsulated. After passing through the four warming zones, the material is thoroughly melted and mixed, enabling it to be molded into various shapes. Following extrusion, the compound is ground into a fine powder using a 5HP PAGANI blade mill with a 5 mm mesh diameter. The ground material is then used to prepare samples for mechanical testing.

2.3. Mechanical tests

The molded specimens were analyzed following the ASTM D 638 standard for tensile properties (ASTM, 2008). The tests were carried out using an Instron Universal Testing Machine (model 1196), at a speed of 1 mm/min. The analyzed response variables were measured in kilograms-force (kg·f/mm^2). The Instron Universal Testing Machine, Model 1196 is a product manufactured by Instron, a well-known company specializing in materials testing equipment. Instron is headquartered in the United States, with its main office located in Norwood, Massachusetts.

2.4. Experimental mixture design

As mentioned earlier, Scheffé polynomial models are commonly used in mixture experimental design. The linear Scheffé model has the following form:

$$E(Y) = \sum_{i=1}^{q} \beta_i x_i \tag{3}$$

Similarly, the quadratic Scheffé model can be described as follows:

$$E\left(Y\right)=\sum_{i=1}^{q}\beta_i x_i+\sum_{i=1}^{q-1}\sum_{j=i+1}^{q}\beta_i j x_i x_j \quad (4)$$

where β_i and β_j are unknown parameters that must be estimated, typically using least squares. Due to the mixture constraint in Eq. (1), the form of the quadratic Scheffé model only contains linear terms and cross-product terms (Cornell, 2002).

There may be instances where in addition to the dependent mixture components, we have other factors and/or process variables that can be controlled independently of one another and of the mixture components. For example, consider a chemical production system. The composition of the WPC formula involves mixture variables (x_i) while the settings on the manufacturing equipment are process variables (z_i). The response of interest, Y, can be modelled as a function of the mixture and process variables

$$Y=f\left(x,z\right)=x^T\beta+x^T\Lambda z+\varepsilon \quad (5)$$

where β is a vector of coefficients representing the main effects, interactions, and possibly cubic terms in the mixture components and Λ is a matrix of coefficients representing the interactions between the mixture and process variables (Anderson-Cook et al., 2004).

Let us consider a mixture experiment involving q components and n process variables, where n can be any positive integer. For the mixture components, each x_i can vary from 0 to 1, defining a (q–1)-dimensional simplex as the mixture region of interest. The combined region of interest, incorporating both the mixture components and process variables, is q–1+n dimensional. To set up design configurations for the process variables and mixture components, one approach is to establish a mixture design at each configuration point for the process variables, or alternatively, to position a factorial arrangement within the process variable settings at each composition point of the mixture components. For instance, if we select a q=3 simplex-centroid design for fitting a special cubic model to the mixture components (Anderson-Cook et al., 2004)

$$\begin{aligned} Y\left(x\right)=&\ \beta_1x_1+\beta_2x_2+\beta_3x_3+\beta_{12}x_1x_2+\\ &+\beta_{13}x_1x_3+\beta_{23}x_2x_3+\beta_{123}x_1x_2x_3 \end{aligned} \quad (6)$$

Let the number of process variables be 2, thus, with z_1=±1 and z_2=±1, a 2^2 factorial design is contemplated for fitting the model in the two process variables:

$$Y\left(z\right)=\alpha_0+\alpha_1z_1+\alpha_2z_2+\alpha_{12}z_1z_2 \quad (7)$$

The combined simplex-centroid by 2^2 factorial design is shown in Figure 1.

The combined design shown in Figure 1 is utilized for gathering data to fit the integrated model encompassing both the mixture components and the process variables. The combined model is

$$\begin{aligned} Y\left(x\right)=&\ \beta_1\left(z\right)x_1+\left(z\right)\beta_2x_2+\left(z\right)\beta_3x_3+\left(z\right)\beta_{12}x_1x_2+\\ &+\left(z\right)\beta_{13}x_1x_3+\left(z\right)\beta_{23}x_2x_3+\left(z\right)\beta_{123}x_1x_2x_3 \end{aligned}$$

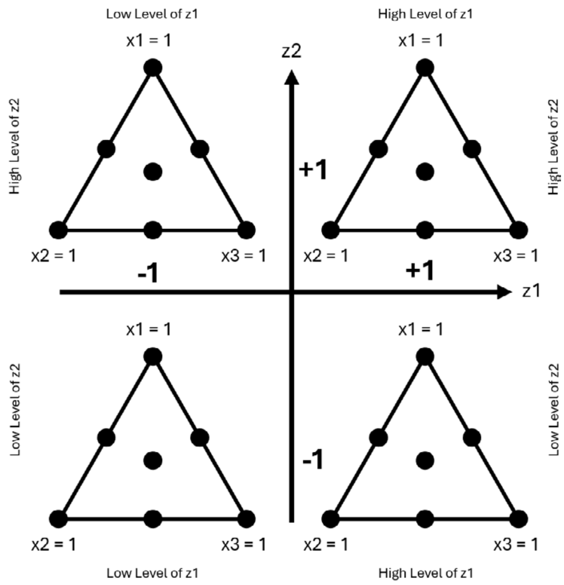

Figure 1: Combined design.

Or in an equivalent form

$$\begin{aligned} Y\left(x\right)=&\ \beta_1\left(\alpha_0+\alpha_1z_1+\alpha_2z_2+\alpha_{12}z_1z_2\right)x_1+\\ &+\left(\alpha_0+\alpha_1z_1+\alpha_2z_2+\alpha_{12}z_1z_2\right)\beta_2x_2+\\ &+\left(\alpha_0+\alpha_1z_1+\alpha_2z_2+\alpha_{12}z_1z_2\right)\beta_3x_3+\\ &+\left(z\right)\beta_{12}x_1x_2+\left(z\right)\beta_{13}x_1x_3+\\ &+\left(\alpha_0+\alpha_1z_1+\alpha_2z_2+\alpha_{12}z_1z_2\right)\beta_{23}x_2x_3+\\ &+\left(\alpha_0+\alpha_1z_1+\alpha_2z_2+\alpha_{12}z_1z_2\right)\beta_{123}x_1x_2x_3 \end{aligned} \quad (8)$$

3. Results and discussion

The components of the composite analyzed are PET (x_1), wood powder (x_2) and E-GMA coupling agent (x_3). As mentioned earlier, the sum of the proportions of these three components must be equal to 1. Furthermore, the components have additional constraints of the type shown in Eq. (2). For the coupling agent, it is assumed that its effective range for promoting bonding between PET and wood is between 0% and 3%. The lower and upper constraints of the component proportions in the mixture are shown in Table 1. The process variables analyzed were particle sizes of 1.4 mm and 2.4 mm; and mixing times of 5 and 10 minutes (see Table 1).

Table 1: Proportions constraints of the mixture components and factors level.

Component	Component and factors
PET (x_1)	$0.60 \le x_1 \le 0.90$ (wt%)
Wood (x_2)	$0.10 \le x_2 \le 0.40$ (wt%)
E-GMA (x_3)	$0 \le x_3 \le 0.03$ (wt%)
Particle size (z_1)	1.4 to 2.4 mm
Mixing times (z_2)	5 to 10 min

To determine the proportions of the compound and the levels of the process variables, a D-optimal mixture design experiment was generated by computer. The design used is an extreme vertices design, with

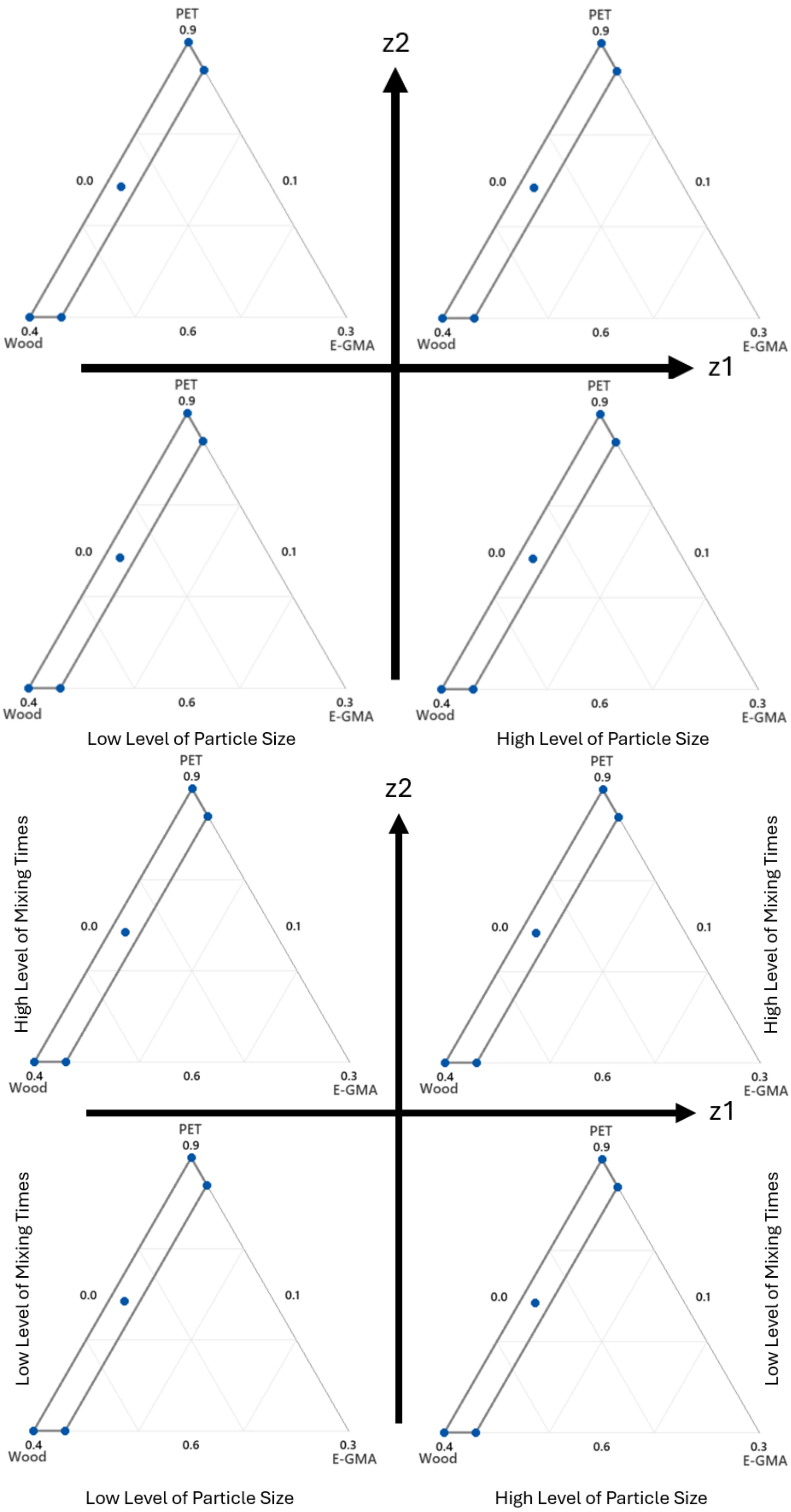

Figure 2: Combined experimental region.

20 design points, combined with a factorial design 2^2. The experimental design and its analysis were performed using the statistical software Minitab®, version 21.1.1. The experimental region and the experimental points, which represent the different formulations made, and the process variables levels, are shown in Figure 2.

Table 2: Experimental Design.

Point	PET (x_1) wt%	Wood (x_2) wt%	E-GMA (x_3) wt%	Particle size (z_1) mm	Mixing times (z_2) min	Tensile* (Y) (kg·f/mm^2)
1	0.6000	0.4000	0.000	-1	-1	1.37
2	0.6000	0.3700	0.030	-1	-1	1.42
3	0.9000	0.1000	0.000	-1	-1	1.68
4	0.8700	0.1000	0.030	-1	-1	1.66
5	0.7425	0.2425	0.015	-1	-1	1.54
6	0.6000	0.4000	0.000	1	-1	1.68
7	0.6000	0.3700	0.030	1	-1	1.72
8	0.9000	0.1000	0.000	1	-1	2.46
9	0.8700	0.1000	0.030	1	-1	2.40
10	0.7425	0.2425	0.015	1	-1	2.03
11	0.6000	0.4000	0.000	-1	1	2.07
12	0.6000	0.3700	0.030	-1	1	2.08
13	0.9000	0.1000	0.000	-1	1	2.70
14	0.8700	0.1000	0.030	-1	1	2.66
15	0.7425	0.2425	0.015	-1	1	2.35
16	0.6000	0.4000	0.000	1	1	3.49
17	0.6000	0.3700	0.030	1	1	3.57
18	0.9000	0.1000	0.000	1	1	3.60
19	0.8700	0.1000	0.030	1	1	3.55
20	0.7425	0.2425	0.015	1	1	3.01

*Experimental results (kg·f/mm^2).

The experimental design, which shows the different formulations developed and experimental results are given in Table 2.

In mixture experiments, ANOVA (Analysis of Variance) is a statistical method used to decide the significance of the effects of different factors on a response variable, for example tensile strength. Specifically, it helps in understanding whether the variations in the response variable can be attributed to the differences in the proportions of the components in the mixture, two or more process variables or if they are due to random variation. ANOVA divides the total variability in the response variable into components attributable to different sources, such as the mixture components, process variables and their interactions (Cornell, 2002). It tests null hypotheses that state there are no effects due to the mixture components, process variables or their interactions. If the null hypothesis is rejected, it indicates that at least one of the components, process variables or the interactions has a significant effect on the response (tensile strength (Y)). ANOVA helps in fitting appropriate statistical models (e.g., linear, quadratic) to describe the relationship between the mixture components, process variables and the response. It provides a basis for selecting the model that best explains the observed data. By computing F-statistics and p-values, ANOVA assesses the significance of each factor in the mixture experiment and the factorial design. Factors with low p-values (typically <0.05) are considered to have a significant effect on the response. The estimated coefficients for each term in the model, along with their standard errors, t-values, and p-values, help in understanding the effect size and significance of each predictor variable and interaction term.

In Table 3, the estimated coefficients of the regression model are presented, along with the p-values and the usual statistics. The coefficient of 2.71 (PET) suggests that for each unit increase in the PET variable, the tensile strength (Y) is expected to increase by 2.71 units, holding other factors constant. The asterisks (*) in the T-value and P-value columns indicate that this effect is statistically significant. The coefficient of 1.26 (Wood) implies that an increase in the Wood content leads to a 1.26 unit increase in tensile strength, holding other variables constant. Like PET, the significance markers (T-value and P-value as *) indicate that this is statistically significant. With a coefficient of 2.03, E-GMA appears to increase tensile strength by 2.03 units for each unit increase, but given the large standard error (4.63) and the P-value marked as *, it suggests high variability and potential insignificance. PET* z_1 interaction term has a coefficient of 0.37 with a T-value of 2.42 and a P-value of 0.03. This indicates a statistically significant interaction between PET and z_1 on tensile strength, suggesting that changes in the variable z_1 modify the effect of PET on the response (Y). The coefficient of 0.46 (Wood*z_1) suggests an interaction between Wood and z_1, but the T-value of 1.22 and P-value of 0.24 indicate this interaction is not statistically significant. For E-GMA*z_1 the coefficient is 0.46, but with a very high standard error (4.63) and a P-value of 0.92, suggesting no significant interaction between E-GMA and z_1. With a coefficient of 0.47 (PET*z_2), a T-value of 3.08, and a P-value of 0.01, this interaction term is statistically significant. This means that the relationship between PET and tensile strength is influenced by changes in z_2. The coefficient of 0.78 (Wood*z_2) indicates a strong interaction between Wood and z_2, with a T-value of 2.07 and a P-value of 0.06. While this is not conventionally significant at the 0.05 level, it is close enough to suggest a possible effect worth investigating further. The coefficient for this interaction E-GMA*z_2 is 0.59, but the T-value of

0.13 and P-value of 0.90 suggest this interaction is not statistically significant. Variance inflation factor (VIF) helps in understanding whether the predictor variables are highly correlated and potentially inflating the variance of the coefficient estimates. High multicollinearity can inflate the variance of coefficient estimates and make the model unstable and difficult to interpret. High correlation, indicating potential multicollinearity problems (VIF > 5). Table 3 shows VIFs values lower than 5.

Table 3: Estimated regression coefficients for tensile strength (Y).

Terms	Coef	SE Coef	T-Value	P-Value	VIF
PET	2.71	0.15	*	*	3.46
Wood	1.26	0.38	*	*	2.76
E-GMA	2.03	4.63	*	*	2.18
PET*z_1	0.37	0.15	2.42	0.03	3.46
Wood*z_1	0.46	0.38	1.22	0.24	2.76
E-GMA*z_1	0.46	4.63	0.10	0.92	2.18
PET*z_2	0.47	0.15	3.08	0.01	3.46
Wood*z_2	0.78	0.38	2.07	0.06	2.76
E-GMA*z_2	0.59	4.63	0.13	0.90	2.18

Coefficients are calculated for coded process variables.

Model Summary provides a quick summary of how good the statistical model fits the data. Key metrics such as R-squared and adjusted R-squared are included to indicate the proportion of variance in the response variable explained by the model. A higher R-squared value suggests a better fit. In this case, the coefficients of determination R-squared and adjusted R-squared are 92% and 86%, respectively (see Table 4). This indicates a satisfactory fit of the regression model. The root mean square error (S) assesses the model's predictive accuracy. These metrics indicate how well the model predicts new observations. A small value of S suggests a good ability to make predictions. In Table 4, the value of S is equal to 0.2818, which is a relatively small value. PRESS (Prediction Error Sum of Squares) is a statistic used to evaluate the predictive capability of a regression model. It provides a measure of how well the model predicts new data points and is particularly useful for validating the model's performance. A lower PRESS value indicates a model with better predictive accuracy. It suggests that the model has a good fit and is capable of accurately predicting new observations. Comparatively, a higher PRESS value may indicate a model that is overfitting the training data and may not generalize well to new data. Table 4 shows a PRESS value equal to 3.38.

Table 4: Model Summary.

S	R-sq	R-sq(adj)	PRESS	R-sq(pred)
0.281861	92.03%	86.23%	3.38052	69.17%

By computing F-statistics and p-values, ANOVA assesses the significance of each factor in the mixture experiment. Factors with low p-values (typically <0.05) are considered to have a significant effect on the response. In Table 5, it can be confirmed that the global regression shows significant statistical significance as it has a p-value close to 0. The linear part of the mixture components also has a significant effect on the response (Y), as it has a p-value of 0.041. Regarding the interaction effect between the mixture components and the operational variable (particle size), only the interaction between PET and particle size has a significant effect on the response (Y), as it has a p-value of 0.034. Regarding the interaction effect between the mixture components and the operational variable (Mixing times), only the interaction between PET and Mixing times has a significant effect on the response (Y), as it has a p-value of 0.010.

Table 5: Analysis of variance for tensile strength (Y).

Source	DF	Seq SS	Adj SS	Adj MS	F-Value	P-Value
Regression	8	10.0912	10.0912	1.2614	15.88	0.000
Component Only						
Linear	2	0.6905	0.6905	0.34525	4.35	0.041
Component*z_1						
Linear	3	3.1866	3.1866	1.06221	13.37	0.001
PET*z_1	1	3.062	0.4647	0.46465	5.85	0.034
Wood*z_1	1	0.1238	0.1178	0.11776	1.48	0.249
E-GMA*z_1	1	0.0008	0.0008	0.00079	0.01	0.922
Component*z_2						
Linear	3	6.2141	6.2141	2.07137	26.07	0.000
PET*z_2	1	5.8588	0.7541	0.75406	9.49	0.010
Wood*z_2	1	0.354	0.3388	0.33882	4.26	0.063
E-GMA*z_2	1	0.0013	0.0013	0.0013	0.02	0.900
Residual Error	11	0.8739	0.8739	0.07945		
Total	19	10.9651				

In experimental design, a main effects plot is a graphical representation used to show the effect of each factor on the response variable, assuming other factors are held constant. The main effects plot helps visualize how changes in a particular process variable affect the response variable (Y). This is useful for understanding the individual contributions of each factor to the overall variation in the response. On the plot, the x-axis typically represents the levels of the factor being analyzed, while the y-axis shows the mean response for each level. For each process variable, the plot displays the average response at different levels of that factor, providing a clear view of how the response changes with changes in the process variable levels. If the lines in the plot are parallel or nearly parallel, it suggests that the factor has a consistent effect across its levels. If the lines are not parallel, it indicates that the effect of the factor varies depending on its level. The slope of the lines reflects the strength and direction of the factor's effect on the response. At the top of Figure 3 is shown that the two operational variables, particle size and mixing time, have a significant effect on the response (tensile strength). For both operational variables, the response value reaches its maximum at the high level.

An interaction plot is a graphical tool used to investigate the effects of different combinations of mixture components on the response variable. It helps to visualize how the interaction between different components influences the outcome. The purpose is to identify and understand how different components of a mixture interact with each other and how these interactions affect the response variable. This is crucial for optimizing mixture formulations. X-axis represents the levels or proportions of one component of the mixture. Y-axis shows the response variable. If the lines in the interaction plot are parallel, it suggests that there is no significant interaction between the components. Each component has a consistent effect on the response. If the lines intersect are not parallel, it indicates an interaction between the components. This means that the effect of one component on the response depends on the level of the other component. At the bottom of the Figure 3 is shown that there is an interaction effect between the two process variables, as the lines are perpendicular.

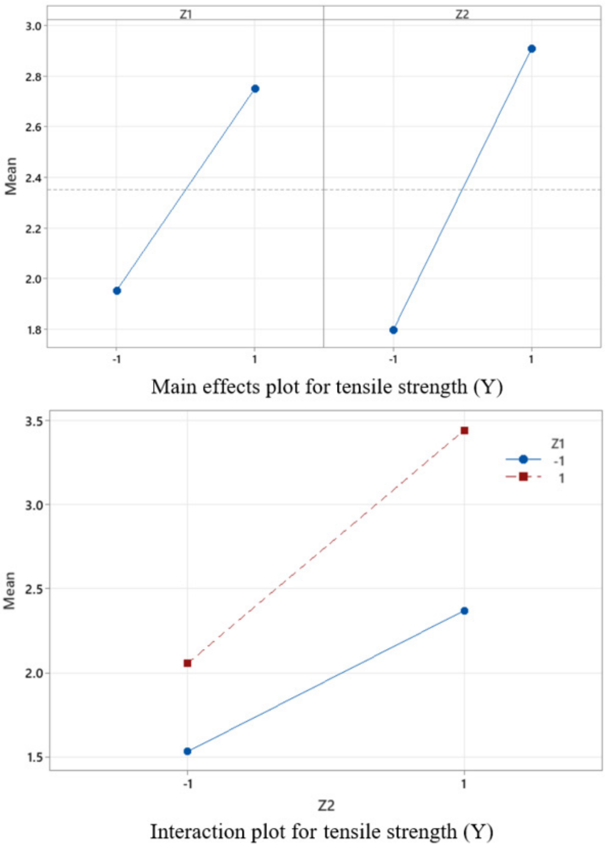

Figure 3: Main and interaction plots for tensile strength (Y).

A cube plot is a graphical representation used to visualize the effects of multiple factors and their interactions on the response variable in a factorial experiment. The purpose is to display the relationship between factors and the response variable in a multi-dimensional space, typically involving three or more factors. It helps in understanding how different combinations of factor levels influence the response. Each axis of the cube represents one of the factors in the experiment. For a three-factor design, the cube has three axes, each corresponding to a different factor. The vertices of the cube represent the different combinations of factor levels. Each vertex corresponds to a unique combination of high and low levels of factors. The response variable values are plotted at the vertices or within the cube, showing how the response changes with different factor combinations. Figure 4 shows the cube plot for tensile strength (Y).

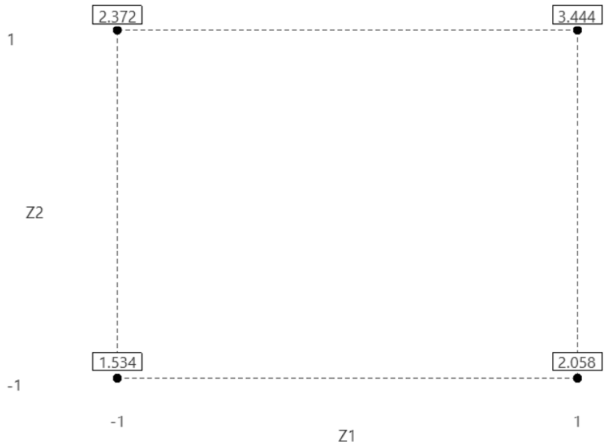

Figure 4: Cube plot (data means) for tensile strength (Y).

A mixture contour plot is a graphical tool used to visualize the effects of different combinations of mixture components on the response variable. It provides a two-dimensional representation of how varying proportions of components affect the response. The purpose is to illustrate how the response variable changes with different combinations of component proportions in a mixture. It helps in understanding the optimal proportions of components to achieve desired outcomes. The plot typically uses the proportions of three mixture components as the axes. The spacing and shape of contour lines indicate how sensitive the response is to changes in component proportions. Closely spaced lines suggest a steep gradient, meaning small changes in component proportions can lead to significant changes in the response. The contour plot helps identify regions of the plot where the response is optimized. This is useful for determining the best mixture composition. Figure 5 shows the mixture contour plot for tensile strength (Y). Darker colors represent higher values of the response Y. Similarly, the gray area shows the region limited by the constraints listed in Table 1.

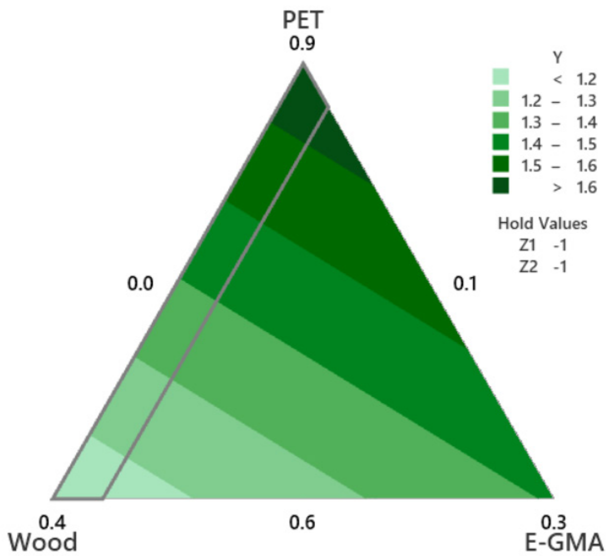

Figure 5: Mixture contour plot for tensile strength (Y).

A mixture surface plot is a graphical tool used to visualize the relationship between mixture components and the response variable across different combinations of component proportions. It provides a three-dimensional view of how varying the proportions of two or more components affects the response. The purpose is to illustrate how the response variable changes with different proportions of mixture components. It helps to understand the effects of component interactions on the response and identify optimal formulations. The shape of the surface indicates how the response varies with changes in component proportions. For instance, peaks and valleys on the surface show areas where the response is maximized or minimized. Figure 6 shows that the highest value of the response is located at the PET vertex.

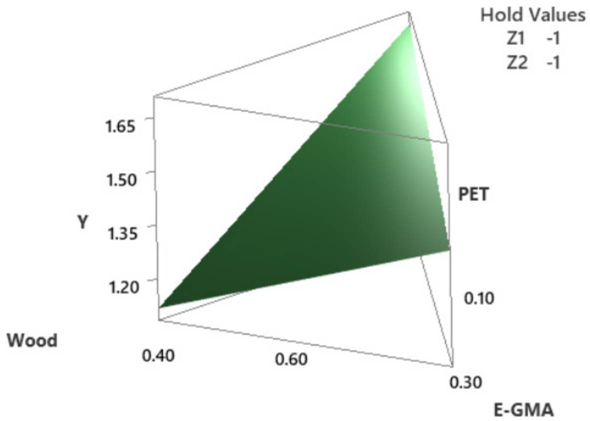

Figure 6: Mixture surface plot for tensile strength (Y).

A contour plot is a graphical representation that shows the relationship between two independent variables and a dependent variable (Y). It uses contour lines to connect points of equal response values, effectively displaying a three-dimensional surface on a two-dimensional plane. The purpose is to visualize how the response variable changes across different levels of two independent variables. Figure 7 shows the process variables contour plot for tensile strength (Y). This contour plot shows the tensile strength (Y) as a function of particle size (z_1) and mixing time (z_2). From the plot, the following interpretations can be made as particle size (z_1) increases (movement to the right on the X-axis), tensile strength (Y) also increases. This is evidenced by the darker areas on the plot, which represent higher values of Y, located on the right side of the z_1 axis. Similarly, as mixing time (z_2) increases (movement upward on the Y-axis), tensile strength (Y) also increases. The darker areas are located at the top of the z_2 axis. The darker areas, which indicate the highest values of tensile strength (Y), are found at the high levels of both z_1 and z_2. This suggests a positive synergistic effect when both factors, particle size and mixing time, are at their high levels.

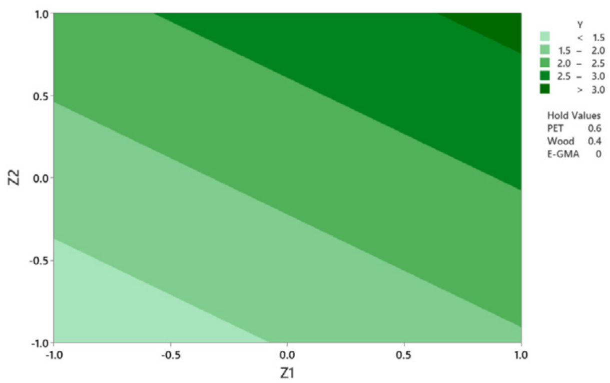

Figure 7: Process variables contour plot for tensile strength (Y).

Figure 8 shows the process variables surface plot for tensile strength (Y). The surface plot showing tensile strength (Y) as a function of particle size (z_1) and mixing time (z_2) provides a three-dimensional view of how these two factors affect the response. From the plot, the following interpretations can be made. The shape of the surface indicates how tensile strength (Y) varies with changes in particle size (z_1) and mixing time (z_2). The crests on the surface represent areas where tensile strength is at its maximum, while the valleys represent areas where it is at its minimum. As particle size (z_1) increases, the surface shows an increase in tensile strength (Y). This indicates that, in general, larger particle sizes tend to improve tensile strength. As mixing time (z_2) increases, an increase in tensile strength (Y) is also observed. The surface rises when mixing time is increased, suggesting that a longer mixing time contributes to higher tensile strength. The shape of the surface can show how the effects of z_1 and z_2 interact. If the surface shows a clear peak in the high region of both factors, this indicates a positive synergistic interaction, where the combination of high levels of both factors results in maximum tensile strength. The plot can help identify the optimal point for both factors, where tensile strength reaches its maximum value. This point is where the surface reaches its highest peak.

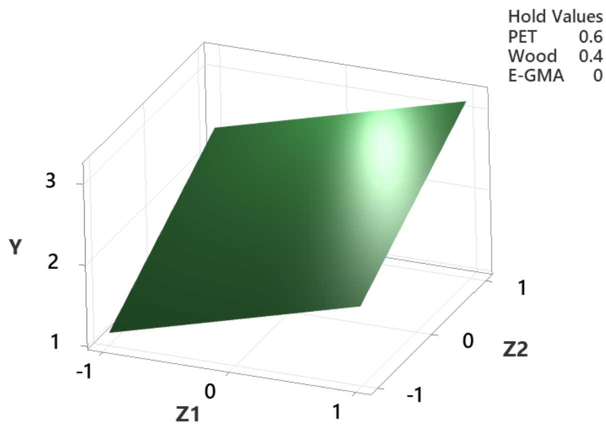

Figure 8: Process variables surface plot for tensile strength (Y).

The observed improvement in tensile strength with larger particle sizes and longer mixing time can be explained through several factors related to the interaction between the polymer matrix and wood fibers, the distribution of fibers, and the composite's structural integrity. When larger wood particles are used in the composite, the available surface area for interaction between the polymer matrix and wood fibers increases. Larger particles typically provide more contact points with the polymer, enabling better mechanical interlocking between the wood and the polymer chains. This stronger interfacial adhesion helps transfer stress more effectively from the polymer matrix to the wood fibers, resulting in improved tensile strength. Extended mixing times allow for more uniform dispersion of the wood fibers within the polymer matrix. With longer mixing, the polymer has more time to infiltrate the wood particles' surface irregularities, filling voids and creating stronger bonds. This improved dispersion also helps to reduce the formation of agglomerates (clusters of wood particles), which can act as stress concentrators and lead to mechanical failure. Therefore, a more homogeneous distribution contributes to a more effective load transfer across the entire composite, increasing tensile strength.

4. Optimization of the composite formulation

Response variable optimization refers to the process of finding the best combination of mixture components that results in the optimal value of the response variable. The primary objective of response variable optimization is to identify the proportions of the mixture components that maximize or minimize the response variable, depending on the desired outcome. Table 6 shows the response optimization for tensile strength (Y).

To attain the maximum tensile strength (Y) within the experimental region used, the compound requires the following formulation: 90% PET as the polymer matrix, 10% wood as the natural filler, and 0% E-GMA, meaning no coupling agent should be used. Additionally, from the analysis discussed in the results section, it is known that the two process variables, particle size (z_1) and mixing time (z_2), have a significant interaction effect on tensile strength (Y). Therefore, to achieve the highest value of the response (Y), the largest particle size and the longest mixing time should be used.

It is important to mention that the regression model suggests that using particle sizes greater than 2.4 mm and mixing times longer than 10 minutes could achieve a higher value of tensile strength. This should be considered with caution, as it would be extrapolating, meaning these would be predictions for areas outside the analyzed experimental region.

Table 6: Response optimization for tensile strength (Y).

Mixture components	Levels	Process variables	Leves	Tensile strength	Predicted response
PET	0.9	z_1	1	Y	3.46
Wood	0.1	z_2	1		
E-GMA	0.0				

5. Conclusion

The inclusion of process variables in mixture experimental design is essential for a more comprehensive and precise optimization of final products. While standard response surface designs are not suitable for mixture problems due to the constraint that proportions must sum to 100%, mixture-process experiments allow for a more detailed evaluation of how operational factors —such as particle size and mixing time— interact with the mixture components. This inclusion of process variables not only enhances the understanding of how processing conditions affect the final product's properties but also facilitates the creation of more accurate predictive models. This improves the consistency and reliability of the production process, enabling more effective optimization of product properties.

The analysis of the regression model indicates that the interaction terms between PET and particle size (z_1), as well as between PET and mixing time (z_2), significantly impact the response variable (Y). The model demonstrates a strong fit, with R-squared and adjusted R-squared values of 92% and 86%, respectively, and a low root mean square error (S) of 0.2818, suggesting good predictive accuracy. The PRESS value of 3.38 supports this, indicating that the model accurately predicts new data points. Furthermore, the absence of high multicollinearity, as evidenced by variance inflation factor (VIF) values below 5, confirms the stability and interpretability of the model. Both particle size and mixing time are shown to significantly influence tensile strength, with the highest response observed at elevated levels of these operational variables.

Contour plots are valuable graphical tools for understanding how varying proportions of mixture components affect the response variable. By providing a two-dimensional representation of component proportions, these plots reveal how changes in the mixture composition influence the response, such as tensile strength. The contour lines illustrate sensitivity to changes, with closely spaced lines indicating significant effects. Figures showing the contour plots for tensile strength demonstrate that both particle size and mixing time positively impact the response. Specifically, higher values of tensile strength are achieved when both particle size and mixing time are at their maximum levels, indicating a synergistic effect. This insight helps in optimizing the mixture composition to achieve desired outcomes.

Response variable optimization aims to identify the optimal proportions of mixture components that maximize or minimize the desired outcome. For maximizing tensile strength (Y) within the studied parameters, the optimal formulation is 90% PET, 10% wood, and no coupling agent (E-GMA). Furthermore, the interaction between process variables, such as particle size and mixing time, plays a crucial role in enhancing tensile strength. To achieve the highest tensile strength, the largest particle size and the longest mixing time should be utilized. However, predictions for particle sizes beyond 2.4 mm and mixing times longer than 10 minutes should be approached with caution, as they involve extrapolation beyond the analyzed experimental range.

The lack of significant effects of the coupling agent on the performance of wood plastic composites (WPC) can be attributed to several factors. First, certain thermoplastic polymers, such as polyethylene terephthalate (PET), may exhibit inherent compatibility with wood fillers, reducing the necessity for a coupling agent. In these cases, the polymer naturally adheres well to the wood fibers, and the addition of a coupling agent may provide limited or no additional benefit. Moreover, the proportion of wood filler within the composite plays a crucial role in determining the efficacy of the coupling agent. When the wood filler content is low (e.g., less than 10%), the coupling agent may not significantly enhance the interaction between the polymer matrix and the wood fibers. The mechanical improvements typically associated with coupling agents, such as increased tensile strength or durability, are more pronounced when the wood filler proportion is higher, as the agent enhances fiber-matrix bonding. Furthermore, PET's high crystallinity and strong mechanical properties often negate the need for a coupling agent. In applications where PET alone provides sufficient mechanical strength, the inclusion of a coupling agent to improve fiber-matrix interaction may be unnecessary. The inherent rigidity and resistance of PET make the addition of a coupling agent redundant, particularly in cases where optimal performance is already achieved without it.

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